Detection and Monitoring of Bioavailable Heavy Metals in Soil: Current Status, Problems, and Prospects (Postprint)

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Abstract

With economic and social development, heavy metal pollution in soils poses a significant threat to food security and human health. However, the currently widely adopted total content detection method cannot accurately reflect the biological toxicity of soil heavy metals. To better understand the contamination status and biological toxicity of soil heavy metals, rapid and effective detection of the occurrence forms and available content of soil heavy metals is required. This paper introduces the definition of soil available heavy metals and their transfer in the soil-organism system. Based on domestic and international literature in related research fields, detection methods for soil heavy metal availability are classified into direct detection methods (chemical and physical detection methods) and indirect detection methods (bioindicator methods) according to detection objects. The paper systematically elaborates on the research progress, applicable scope, advantages, and disadvantages of extraction methods in chemical detection, diffusive gradients in thin-films (DGT) technique and spectroscopic analysis in physical detection, as well as indicator plant detection, microbial detection, and animal detection in bioindicator methods. It summarizes that the main limitation of current detection methods is their inability to simultaneously satisfy both large-area monitoring and precise detection requirements. The paper also identifies that rapid, large-area, in-situ determination of pollutant types and contents, along with implementing corresponding measures to ensure food security and human health, are urgent issues requiring resolution. This paper conceptualizes soil-plant-human as an integrated system to understand the migration and enrichment mechanisms of heavy metal availability within this system. It proposes that selecting appropriate data processing methods and modeling parameters, combined with remote sensing, geographic information systems, and global positioning systems for in-situ three-dimensional monitoring of soil heavy metal pollution, represents the

primary development direction for future research on detection and monitoring of soil heavy metal available content.

Full Text

The Detection and Monitoring of Available Heavy Metal Content in Soil: A Review

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Abstract: With rapid economic and social development, heavy metal contamination in soil poses an increasingly serious threat to food security and human health. However, conventional total concentration detection methods cannot accurately reflect the actual toxicological bioavailability of heavy metals in soil. To better understand heavy metal pollution levels and their biological toxicity, rapid and effective detection of heavy metal speciation and available content is essential. This paper introduces the definition of available heavy metals and their transfer in soil-organism systems. Based on domestic and international literature, detection methods for available soil heavy metals are categorized into direct methods (chemical and physical detection) and indirect methods (biological indicators). We systematically review research progress, applicability, and advantages/disadvantages of chemical extraction methods, diffusive gradients in thin-films (DGT) technology, spectral analysis, plant indicator methods, microbial detection methods, and animal detection methods. The main limitation of current methods is their inability to simultaneously satisfy both large-area monitoring and precise detection requirements. Rapid, large-scale, in-situ determination of pollutant types and concentrations is urgently needed to safeguard food security and human health. This paper treats the soil-plant-human system as an integrated whole to understand heavy metal migration and enrichment mechanisms, suggesting that appropriate data processing methods and modeling parameters combined with remote sensing, geographic information systems, and global positioning systems for in-situ three-dimensional monitoring represent the primary future direction for research on available heavy metal detection and monitoring.

Keywords: Soil contamination; Heavy metal speciation; Bioavailability; Content detection; Remote sensing monitoring

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With increasing population and accelerated urbanization and industrialization, heavy metal pollutants have entered soils through various pathways, including industrial "three wastes" emissions, municipal sewage, and improper application of pesticides and fertilizers. Heavy metals cannot be biodegraded and exhibit strong concealment and high toxicity. Once in soil, particularly farmland, they affect crop yield and quality while posing significant health risks to humans, making them a research priority for environmental scientists worldwide.

Previous research on heavy metal availability focused primarily on total content effectiveness. However, due to the complex and heterogeneous composition of soil environments and intricate chemical reaction mechanisms, heavy metals exist in multiple forms with varying bioavailability. Therefore, the biological toxicity of heavy metals depends not only on their total content but largely on their speciation distribution. Detecting and monitoring available heavy metal content provides an effective approach for accurately assessing pollution levels, predicting ecosystem impacts, and evaluating human health risks, while also forming the theoretical basis for remediation strategies. Nevertheless, precise definitions of heavy metal availability remain controversial across soil science, biology, and toxicology disciplines, and no standardized measurement methods currently exist for risk assessment.

This paper introduces definitions of available heavy metals across different disciplines and their transfer in soil-organism systems. Based on relevant literature, we categorize detection methods into direct and indirect approaches according to detection targets, systematically reviewing research progress, applicability, advantages, and disadvantages of various methods to provide references for future research, particularly regarding large-scale farmland safety.

1 Definition of Available Heavy Metal Speciation

The chemical speciation of heavy metals strongly influences their behavior. Heavy metals can exhibit different toxicity and environmental behavior based on speciation differences. Their mobility in the environment, absorption by organisms, and toxic effects all serve as bases for characterizing and understanding heavy metal speciation. However, no unified definition of "availability" currently exists, with slight variations across disciplines. Environmental scientists define it as environmental bioavailability and environmental availability—the forms that can be absorbed by organisms or cause toxic effects. Toxicologists focus on toxicological bioavailability—the portion that enters an organism' s digestive system and can be dissolved and absorbed. The International Standards Organization (ISO) describes bioavailability as a dynamic process [Figure 1: see original paper] comprising three steps: (1) available heavy metals in soil (environmental availability); (2) heavy metals absorbed by organisms (environmental bioavailability); and (3) heavy metals accumulated in organisms causing toxic effects (toxicological bioavailability). These three forms can transform under specific conditions. In summary, although no unified concept exists, all definitions essentially investigate the potential relationship between heavy metals and organisms, linking organisms with their surrounding environment.

2 Review of Detection Theories, Methods, and Achievements for Available Soil Heavy Metal Content

Numerous studies on available soil heavy metal detection span physics, chemistry, and biology. Based on detection targets, methods are divided into direct and indirect approaches.

2.1 Direct Detection Methods

Direct methods analyze contaminated soil directly, primarily including chemical extraction and physical analysis.

2.1.1 Chemical Extraction Analysis Methods Chemical extraction employs appropriate reagent solutions (single or mixed) or sequential solutions with increasing extraction capacity at specific soil-to-solution ratios to extract heavy metals, with subsequent measurement of extracted content. Methods are categorized as single extraction and sequential extraction.

Single extraction uses one reagent to extract plant-available heavy metal forms, showing good correlation with plant uptake and suitable for assessing short- to medium-term hazards. Common extractants include chelating agents (e.g., 0.05 $\text{mol} \cdot \text{L}^{1}$ EDTA, DTPA), neutral salts (e.g., 0.05 $\text{mol} \cdot \text{L}^{1}$ CaCl , 1 $\text{mol} \cdot \text{L}^{1}$ NaNO), and acids (e.g., 0.43 $\text{mol} \cdot \text{L}^{1}$ HOAc, 0.1 $\text{mol} \cdot \text{L}^{1}$ HNO). Extraction efficiency varies by reagent mechanism and heavy metal speciation, requiring case-specific selection.

Sequential extraction, also called stepwise or fractionation extraction, uses different reagents in a procedure where each targets specific metal forms. Widely used methods include Tessier (5-step) and BCR (3-step) procedures and their modifications. These methods fractionate heavy metals differently: Tessier defines five forms (exchangeable, carbonate-bound, Fe-Mn oxide-bound, organic-bound, residual), while BCR defines water-soluble, exchangeable/carbonate-bound, Fe-Mn oxide-bound, and organic/sulfide-bound forms. Different methods yield slightly different speciation results that cannot be directly compared, though each identifies the most active metals (typically Cd, followed by Pb and Zn).

While chemical extraction is simple and precise, it is primarily an ex-situ method requiring field sampling and laboratory analysis, which is time-consuming and labor-intensive. Extracted available content only correlates positively with plant uptake rather than representing the exact portion absorbed by plants, necessitating further correlation analysis.

2.1.2 Physical Analysis Methods Physical methods analyze heavy metal speciation using physical principles, primarily diffusion-based (DGT) and optical (spectral analysis) approaches.

Diffusive Gradients in Thin-Films (DGT) Technology. Invented by Davison et al. in 1994, DGT is based on Fick’ s first law of diffusion, quantifying ions passing through a specific diffusion membrane thickness over time. The device comprises a diffusion phase (hydrogel allowing free ion diffusion) and a binding phase (polymer compounds with chelating functional groups). DGT introduces a dynamic concept: measured available concentration includes both soil solution content and dynamically released content from solid phases during measurement. Since plant root uptake depletes soil solution concentrations, promoting replenishment from solid phases, this dynamic process is crucial for bioavailability assessment. DGT has been widely applied for in-situ sampling and bioavailability studies in water, sediment, and soil. However, its accuracy for bioavailability assessment may be affected by plant species. Studies comparing DGT with traditional chemical methods show varying correlations with plant uptake across different metals and plant species. While not necessarily more accurate than chemical extraction, DGT offers advantages as an in-situ technique that is simple to operate, selectively absorbs specific metal forms, predicts bioavailability well, and provides information on release kinetics. Its measurements are less affected by soil physicochemical properties, though the technique remains largely laboratory-based and cannot fully account for all plant growth factors.

Spectral Analysis. Spectral techniques determine material structure and chemical composition using spectroscopic principles, widely applied in soil heavy metal detection. Traditional methods include atomic absorption, UV-Vis, infrared, and plasma spectroscopy (the most widely used), which are sensitive but can only measure total content or extracted fractions, requiring sample pretreatment. Modern structural/speciation techniques like electron probe microanalysis, X-ray diffraction (XRD), and synchrotron X-ray fluorescence spectroscopy (SXRFS) enable rapid analysis without pretreatment but have lower sensitivity, primarily applied in mineral research rather than farmland soil detection. Hyperspectral remote sensing, developed since the 1980s, predicts heavy metal content through high spectral resolution and continuous bands, enabling non-destructive, non-contact rapid measurement based on heavy metal adsorption by soil components. While traditional spectral methods are fast and sensitive, they are rarely used alone for available heavy metal detection. Hyperspectral remote sensing remains exploratory due to complex soil composition and interference from texture, moisture, organic matter, and iron oxides, though its rapid, non-destructive, large-area monitoring potential is promising.

2.2 Indirect Detection Methods

Indirect methods assess heavy metal availability through biological responses in contaminated environments, primarily biological detection using individual, population, or community reactions to evaluate pollution.

2.2.1 Plant Indicator Method Plant indicator methods are economical, simple, and reliable for assessing heavy metal availability. They evaluate availability through: (1) plant damage symptoms (root, stem, leaf color/shape changes, chlorophyll and nitrogen content variations); and (2) pollutant content in plant tissues.

Visual symptom assessment is rapid and non-destructive but cannot quantify available content, and heavy metal stress is often difficult to identify macroscopically due to pollution concealment. Hyperspectral remote sensing shows promise for indirect detection, as plant leaf reflectance spectra correlate with morphological and physiological characteristics that change under pollution stress. However, environmental factors affect spectra, systematic data processing methods are lacking, and quantitative estimation accuracy requires improvement. Currently, this method is only suitable for severely polluted soils, with monitoring values serving as references rather than definitive assessments for pollution surveys.

Tissue content analysis has been widely reported. Methods using rye seedlings, wheat, and other plants to determine bioavailability have shown applicability, considering soil-plant relationships and reflecting specific metal-plant bioavailability. However, like chemical methods, this requires sampling and is limited by plant growth cycles, serving only as an auxiliary method.

2.2.2 Microbial Detection Method Microorganisms intimately contact soil microenvironments, making their biomass and activity ideal pollution indicators. Microbial detection uses characteristic indicators like microbial biomass, community structure, and metabolic quotient changes to reflect contamination. Soil microorganisms are highly sensitive bioindicators, as minor soil changes alter their diversity. However, major obstacles include limited understanding of relationships between general soil processes and microbial community structure, and unclear baselines and thresholds for specific indicators.

2.2.3 Animal Detection Method Animal methods primarily assess toxicological bioavailability, mostly using total heavy metal content. Studies show soil animal community diversity decreases with increasing pollution. Currently, animal detection is mainly used for qualitative pollution research.

3 Problems and Development Trends

3.1 Problems in Available Heavy Metal Detection

Direct methods provide instantaneous pollution information at specific times and profiles with high precision but are destructive and exhibit "lag effects" – pollution is detected only after impacts have occurred. They also have weak comprehensive analysis capabilities for farmland ecosystems. Indirect methods consider soil-biological relationships and have practical value but suffer from long cycles, relatively low precision, and inability to directly identify pollutant types. Both traditional and modern methods have applicable scopes and limitations, requiring selection based on evaluation requirements, time constraints, and conditions. Combining multiple methods can improve accuracy.

3.2 Development Trends

The soil-plant system is fundamental to terrestrial ecosystems and environmental elements. Heavy metal accumulation threatens sustainable agricultural development, ecological security, and food safety through food chain transfer. Therefore, detecting and monitoring soil heavy metal pollution to safeguard food security and human health is urgently needed. Future research should focus on:

1. **In-situ detection and monitoring.** Most current methods are ex-situ, time-consuming, costly, and soil-destructive. In-situ techniques reflect actual field conditions and represent an important development direction for real-time heavy metal identification.

2. **Research on heavy metal stress mechanisms and morphological impacts on plants.** Understanding the physiological and biochemical processes from soil absorption to morphological symptoms will help identify appropriate detection methods. While modern molecular biology enables mechanistic exploration, further research is needed.

3. **Systematic soil-plant-human research.** Bioavailability is a comprehensive effect of soil, heavy metals, plants, and humans. Treating this as an integrated system to study migration mechanisms and key influencing factors is crucial for reasonable pollution assessment and remediation strategies.

4. **Large-area three-dimensional monitoring.** Previous research focused on point-scale sampling and laboratory detection with limited spatiotemporal representation. For China' s prominent farmland pollution issues, future emphasis should develop rapid, precise, large-area monitoring technologies using portable hyperspectral instruments, aerial, and spaceborne remote sensing. Hyperspectral remote sensing based on RS, GIS, and GPS will be important for rapid, large-scale, three-dimensional monitoring of available heavy metals.

5. **Development of hyperspectral remote sensing and data processing technologies.** Imaging spectrometers now enable hyperspectral data acquisition from portable to satellite platforms. Data processing techniques are increasing, including continuum removal, logarithmic reciprocal, derivative, and wavelet transforms for spectral preprocessing, and spectral matching and mixed pixel decomposition for imagery. Current methods analyze spectral data to identify sensitive parameters and indices, using regression, principal component analysis, partial least squares regression, and fuzzy neural networks to model relationships with heavy metal content. However, research remains exploratory due to varying plant absorption characteristics and lack of standardized models. Designing standardized hyperspectral data transformation and processing methods is crucial. Additionally, scale effects limit direct application of leaf-scale indices to canopy or regional scales due to atmospheric conditions, illumination, and

landscape heterogeneity. Improving estimation accuracy requires collaborative efforts from remote sensing scientists and related researchers.

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Note: Figure translations are in progress. See original paper for figures.