

Long-term fate of arsenic pollution in subtropical agricultural soils

Ganlin Zhang

g1zhang@issas.ac.cn

Institute of Soil Science, Chinese Academy of Sciences <https://orcid.org/0000-0001-5729-1062>

Yamin Chen

Northeast Forestry University

Shunhua Yang

Ruiqiang Liu

Northeast Forestry University

Hongda Song

Guiyao Zhou

Hua-Yong Wu

Institute of Soil Science, Chinese Academy of Sciences

Jerome Nriagu

University of Michigan

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Abstract

The historical use of arsenical compounds in agriculture has introduced substantial quantities of arsenic (As) into soils, where it represents a potential threat to food safety and groundwater quality. A common misconception is that As is immobilized in the surface-soil layer and vadose zone, and subsequently eroded and transported via streams to the ocean over time. Knowledge of As redistribution and retention in deep soils and regolith remains poorly understood. Here we investigated As redistribution along 10-m regolith profiles under contrasting land uses in a subtropical red-soil critical zone of China. Agricultural land use substantially increased As stocks relative to natural woodland, with enrichment concentrated below 100 cm, where more than 80% of total As was stored. The reticulate red clay (RRC) horizon emerged as the dominant subsurface As reservoir. Elevated As/Pb ratios in the RRC horizon indicate downward migration of land-use-specific legacy As inputs, rather than simple lithogenic accumulation. Importantly, Fe_2O_3 was the strongest predictor of As enrichment in the RRC horizon, where its concentration was significantly higher than in both the surface soil layer and deep weathered saprolite. These findings identify Fe-oxide-rich RRC horizons as critical long-term sinks for agricultural As pollution in deep red-soil critical zones.

1. Introduction

Arsenic (As) and, to a lesser extent, lead (Pb) compounds have been used in agriculture for centuries.^[1–5] The earliest available evidence for the use of As sulfides as a pesticide dates to the early 10th century AD in China and the Middle Ages in Western countries.^[1] From the late 1800s until the discovery of dichlorodiphenyltrichloroethane (DDT) in 1942, different As compounds (including lead arsenate, calcium arsenate, magnesium arsenate, zinc arsenate, zinc arsenite, and Paris green) were the primary pesticides and herbicides used in agriculture in most countries.^[2–5] These compounds were applied to fruit trees in orchards, crops and turf grass (e.g., golf courses, sod farms) and widely used in livestock dips to control ticks, fleas, and lice and in controlling potato beetles and mosquitoes.^[1, 3–6] A recent report estimated that 14% to 17% of cropland is affected globally and that between 0.9 and 1.4 billion people live in regions of heightened human health and ecological risks from toxic metal pollution.^[7] For As specifically, most of the contaminated areas are in fertile alluvial deposits of large deltas and/or along major river basins – the major food baskets of the world.^[8–12] Understanding As migration behavior in these regions is therefore essential for assessing the potential risks to groundwater and food security.^[9–14]

Arsenic behavior in tropical/subtropical soils is distinct because these ecosystems (such as the Oxisols and Ultisols) are typically highly weathered, acidic, and rich in iron (Fe) and aluminum (Al) oxides.^[15–19] It is widely believed that these mineralogical characteristics make tropical soils exceptionally effective at sequestering and accumulating As, while often resulting in lower bioavailability to life forms compared to temperate soils.^[16–20] Under agricultural conditions, however, the biogeochemistry of many toxic metals can be modified to make the trace metals more bioavailable and mobile, thereby orchestrating their

migration into surface and subsurface waters.^[17–20] While the losses of trace metals from agricultural systems have been of interest for more than a century, few studies have addressed such diagenetic migration and storage of As in the vadose zone.^[8–12] The behavior of toxic metals in surface soils (< 100 cm) has been widely investigated, but the controls on their redistribution and retention in deep soil and regolith remain poorly understood.^[21–25]

This topsoil-centered perspective overlooks the deep critical zone, where weathering, water flow, and pedogenesis create strong vertical heterogeneity in regolith structure and reactivity.^[21–25] In tropical/subtropical red-soil landscapes, the regolith commonly consists of an upper uniform red clay (URC), a reticulate red clay (RRC) horizon, and underlying weathered sandstone (WS).^[26–28] Among these layers, the RRC horizon may be particularly important for As retention because of its clay-dominant texture enriched in Fe and Al oxides, heterogeneous structure, high water-retention capacity, and abundant reactive mineral surfaces.^[26–30] These properties may allow the RRC horizon to function as a deep reactive sink that accumulates As and slows its migration.^[29–33] At the same time, the stored As may become amenable to remobilization if hydrological pathways, redox conditions, or competing ions change.^[34–36] Therefore, identifying the role of RRC in deep As storage is essential for understanding whether tropical/subtropical red-soil regolith acts primarily as a long-term barrier or as a delayed source of As to groundwater.^[34–37]

A key uncertainty is how to distinguish the sources of As accumulated in deep regolith. Elevated As in the vadose zone may reflect downward leaching of anthropogenic As from historical agricultural inputs, but it may also arise from natural weathering of As-bearing parent materials or from mineralogical changes during pedogenesis.^[8–12] Historically, arsenical compounds, including Pb arsenate, were widely used in some agricultural systems, providing a potential source of both As and Pb to surface soils.^[1–5] However, Pb is much less mobile than As and is generally retained in surface soil layers after anthropogenic input.^[4, 5, 38–41] We therefore used Pb as a relatively conservative reference element to evaluate whether deep As enrichment was primarily inherited from parent-material weathering or resulted from preferential downward migration of As.^[38–41] If As and Pb were mainly derived from the same weathering source, their concentrations should covary vertically and the As/Pb ratio should remain relatively stable with depth. In contrast, an increase in As/Pb ratios in the deep regolith would indicate preferential As enrichment relative to Pb, consistent with leaching of anthropogenic As into the vadose zone.^[4, 5, 29–31]

2. Results

2.1 Relationship between regolith thickness and arsenic and lead storage

Across the agricultural boreholes, total As storage ranged from 415.9 to 2306.8 kg ha⁻¹ and increased significantly with regolith thickness ($R = 0.80, p < 0.001$; Fig. 1a). Total Pb storage also increased with regolith thickness, although the relationship was weaker than that observed for As ($R = 0.65, p < 0.001$; Fig. 1b). The As/Pb storage ratio showed a similarly positive relationship with regolith thickness ($R = 0.79, p < 0.001$; Fig. 1c), indicating that thicker regolith profiles stored proportionally more As relative to Pb.

2.2 Effects of land use on vertical arsenic and lead distributions

Vertical profiles of As concentration in regolith differed markedly among land-use types (Fig. 2a–c). Woodland profiles generally showed lower As concentrations (Figure S1a–b) and shallower As-enriched zones than agricultural profiles (Figure S1c). In contrast, cropland, orchard, and paddy-field profiles commonly exhibited thicker As-enriched intervals, higher peak As concentrations, and deeper peak positions. Orchard profiles showed the deepest As peaks (Fig. 2a–c), with the mean peak depth reaching 277 cm, approximately 77%, 31%, and 3% deeper than those in woodland, cropland, and paddy-field profiles, respectively. Pb enrichment commonly occurred within or near the same deep horizons where As was enriched. The As/Pb concentration ratio varied with both depth and land use (Fig. 2g–i). Intervals with elevated As/Pb ratios indicate enhanced As enrichment relative to Pb. Despite substantial site-to-site variation within each land-use type, agricultural use was generally associated with a deeper downward extension of the As-enriched zone.

2.3 Regolith properties controlling arsenic concentration and As/Pb ratios

Regolith physicochemical properties were significantly associated with variations in As concentration (Figure S9). Arsenic concentration was negatively correlated with pH, soil bulk density (SBD), and sand concentration, but positively correlated with Fe₂O₃ concentration, Al₂O₃ concentration, soil water concentration (SWC), and clay and silt concentration (Figure S9, $p < 0.05$). random forest analysis further quantified the relative importance of these properties in explaining the variation in As concentration (Fig. 3). For the full dataset, the model explained 78% of the variation ($p < 0.01$). Mineral-related variables contributed significantly to the model association, with Fe₂O₃ identified as the primary predictor, followed by Al₂O₃. The concentrations of Fe₂O₃ and Al₂O₃ in the reticulated red clay layer were significantly higher than those in the uniform red clay and weathered layers (Figure S4).

The As/Pb ratio showed similar associations with regolith properties. In the full random forest model, Fe₂O₃ concentration, Al₂O₃ concentration, SWC, clay and silt, pH, and SBD together explained 53% of the variation in the As/Pb ratio. Land-use-specific models explained 60%, 41%, and 37% of the variation in cropland, orchard, and paddy-field profiles, respectively (Figure S5). Both Al₂O₃ and Fe₂O₃ concentrations

were positively correlated with the As/Pb concentration ratio, with a stronger relationship for Fe_2O_3 ($R = 0.38$, Figure S3b) than for Al_2O_3 ($R = 0.18$, Figure S2b).

3. Discussion

Our results show, for the first time, that deep regolith, rather than surface soil alone, is the dominant reservoir of legacy As pollution in tropical/subtropical red-soil critical zone (Fig. 4). Across the profiles, 81% of total As stock occurred below 100 cm (Fig. 2), indicating that assessments restricted to the conventional surface-soil layer would substantially underestimate the magnitude of As storage and overlook the zone most relevant to long-term contaminant retention and groundwater vulnerability.^[21–25] A comparison of the As and Pb profiles helps to distinguish among alternative explanations for deep As enrichment. If the observed profile patterns were controlled solely by upward release from weathered parent material, agricultural and woodland profiles developed in the same critical-zone setting would be expected to show similar As and Pb enrichment patterns. Instead, we found the elevated As/Pb ratios in deep soil horizons (Fig. 2g–i), supporting a downward legacy-input signal of the As. The differences in translocation are consistent with the fact that this comparative framework is useful because As is sensitive to Fe redox cycling ($R = 0.87$, Figure S3a), environmental change, and hydrological redistribution, whereas Pb is generally less redox-sensitive ($R = 0.42$, Figure S3d) and more strongly retained by mineral and organic surfaces.^[34–36, 38]

The RRC horizon appears to be the key pedogenic unit regulating deep As enrichment. Peak As depth increased significantly with the midpoint depth of the RRC, indicating that profile architecture, rather than depth alone, determines where As is preferentially retained (Figure S7). In highly weathered red soils, the RRC horizon is characterized by fine-textured materials, abundant reactive mineral surfaces, relatively high water-retention capacity, and slower vertical transport compared with adjacent layers (Figure S8).^[26, 27, 37] These properties favor As sorption and storage by increasing contact between dissolved As and Fe/Al oxide-rich surfaces.^[29–33] The RRC horizon can therefore be interpreted as a deep reactive retention zone that intercepts downward-moving As (Fig. 4). Importantly, regolith structure does not merely provide physical space for contaminant accumulation; it shapes the depth, intensity, and persistence of As storage through coupled pedogenic, mineralogical, and hydrological properties.^[21–25]

Land-use history further explains why agricultural profiles, especially orchard and paddy-field profiles, exhibited stronger and deeper As accumulation than woodland, with the orchard mean peak depth reaching 277 cm (Fig. 2a–c and Figure S1). In orchards, enhanced As accumulation may reflect both larger cumulative inputs from historical pesticide or fertilizer use and the biological effects of perennial fruit-tree systems.^[1–5] Compared with annual upland crops, fruit trees develop deeper and longer-lived root systems, which can influence As redistribution through deep root uptake, fine-root turnover, rhizosphere acidification, organic ligand release, and root-induced preferential flow paths.^[20, 40, 41] These processes may repeatedly modify the mobility and retention of As within deeper horizons, thereby reinforcing As accumulation in orchard regolith (Fig. 4). Paddy fields likely promote As redistribution

through a different pathway dominated by hydrological and redox processes.^[42–48] Periodic flooding creates reducing conditions that can dissolve Fe oxides and release sorbed As, whereas drainage and re-oxidation can promote Fe precipitation and As re-adsorption at new depths.^[48–52] Repeated flooding–drainage cycles can therefore generate vertical As redistribution that is not fully captured by static texture variables alone.^[48–52] This mechanism is consistent with the relatively lower variance explained by the paddy-field random forest model (Fig. 3d) and with the divergence between As and Pb in some paddy intervals (Fig. 2c, f, i).

Among the measured regolith properties, mineral-related geochemical indicators emerged as the strongest predictor of As concentration, with Fe_2O_3 showing the highest relative importance in the random forest model (Fig. 3a). The positive correlations of As with Fe_2O_3 , Al_2O_3 , clay and silt, and soil water concentration, together with the negative correlations with sand, pH, and bulk density, indicate that As preferentially accumulates in finer, wetter, and less compact portions of the profile (Figure S9). Fine particles provide larger specific surface areas and are more likely to host Fe- and Al-oxide coatings (Figures S2 and S3) that retain arsenate through surface complexation and related sorption reactions.^[29–33] The negative relationship with pH is consistent with stronger oxyanion adsorption on variable-charge mineral surfaces under acidic conditions, which are characteristic of strongly weathered red soils.^[34, 35, 53] The negative correlation with bulk density is likely indirect, reflecting the tendency of lower-density horizons to have greater porosity and water retention, both of which may increase contact time between dissolved As and reactive mineral surfaces.^[21–25]

The relationship between deep As accumulation and aquifer depth is critical for risk assessment. Previous hydrological observations at the study site indicate that the groundwater table typically fluctuates between approximately 4 and 6 m depth.^[27] Many As-enriched horizons occurred above, within, or near this interval, especially where the RRC horizon was well developed (Figures S7 and S12). Although the concentrations of As and Pb in the soil were below the risk screening values for agricultural land soil contamination in China ($\text{As} < 55 \text{ mg kg}^{-1}$ and $\text{Pb} < 170 \text{ mg kg}^{-1}$),^[54] indicating no exceedance of current soil-risk screening thresholds, this does not necessarily imply the absence of groundwater risk.^[8–13] Under such conditions, the RRC horizon may retard As transport by groundwater by retaining As within fine-textured and reactive deep regolith (Fig. 4). However, this barrier function should not be equated with permanent stability. Seasonal rise in water-table, prolonged saturation, increased dissolved organic carbon, acidification, or reductive dissolution of Fe oxides could remobilize retained As and enhance its downward transfer.^[48–52] Thus, deep regolith should be viewed both as an important As sink and a potential delayed source, with groundwater vulnerability depending on the interaction among As accumulation depth, RRC architecture, and water-table dynamics.^[21–25]

Although this study provides new evidence for substantial As accumulation in deep regolith and highlights its potential implications for groundwater vulnerability, several interpretive constraints should be acknowledged. We quantified total As and major bulk regolith properties, but did not directly determine As speciation, Fe/Al mineral phases, microbial processes, preferential flow pathways,

porewater or groundwater As concentrations. These unmeasured processes are important because As speciation, microbial Fe reduction, and groundwater flow can strongly alter As release, retention, and transport.^[16, 18] Therefore, the proposed mechanisms of As release, retention, paddy-field redox redistribution, orchard biological recycling, and Pb-assisted source discrimination should be treated as process-based interpretations rather than direct measurements. In addition, the discrete sampling intervals used here may have overlooked fine-scale geochemical transitions within individual horizons. Future work should combine high-resolution mineralogical and speciation analyses with hydrological, plant, and microbiological measurements to better resolve the mechanisms controlling As release, retention, and remobilization in deep regolith.

4. Conclusion

In conclusion, this study shows that in subtropical agricultural red-soil landscapes, legacy As pollution is stored predominantly in deep regolith rather than in surface soil. The RRC horizon was the principal As accumulation zone, and fine-textured, water-retentive regolith properties were closely associated with As concentration, whereas total As storage increased with regolith thickness. Pb profiles and As/Pb ratios provided supporting evidence that deep As enrichment reflects both downward legacy inputs and As-specific redistribution, rather than derived from weathering parent rocks. These findings highlight deep regolith architecture, especially the RRC horizon, as a key control on the long-term fate of agriculturally derived As. Although the RRC horizon may currently act as a deep reactive barrier that retards As transport toward groundwater, this retained As should not be regarded as permanently immobilized. Changes in hydrology, redox conditions, acidity, or dissolved organic matter could remobilize As and transform deep regolith from a sink to a delayed source of groundwater contamination.

5. Experimental Section

5.1 Study area

The study was conducted at the Sunjia Critical Zone Observatory site in China (Fig. 5). This 4 km² (2 × 2 km) site features undulating hilly terrain, with elevations ranging from 38 to 74 m and slopes between 0–8°. The region has a subtropical monsoon climate, with a mean annual precipitation of 1795 mm and a mean annual temperature of 17.8°C.^[26, 27] The land cover consists of paddy-field (28%), cropland (23%), woodland (19%), orchard (9%), and other uses (21%). Primary crops include peanuts (*Arachis hypogaea*), sweet potatoes (*Ipomoea batatas*), cassava (*Manihot esculenta*), oilseed rape (*Brassica napus*), turnips (*Brassica rapa*), and watermelons (*Citrullus lanatus*). The paddy-fields utilize a double-rice (*Oryza sativa* L.) cropping system.^[27] Orchards are predominantly planted with Mandarin citrus (*Citrus reticulata* Blanco) and chestnut (*Castanea mollissima* Bl.). Woodland areas are dominated by camphor (*Cinnamomum camphora*) and masson pine (*Pinus massoniana*). The soils are derived from Quaternary Red Clay and are broadly equivalent to Acrisols and Ferralsols in the WRB system.^[55, 56]

5.2 Sample collection and arsenic and lead measurements

In March 2018, 21 10-m regolith profiles were established to represent differences in land use, cultivation history, and geomorphic position (Figure S8), including woodland ($n = 3$), orchard ($n = 3$), cropland ($n = 7$), and paddy-field ($n = 8$). Continuous regolith cores were collected from the surface to the weathered bedrock using a rotary corer with an internal diameter of 13 cm. Samples were collected at 10-cm intervals from 0 to 100 cm, at 20-cm intervals from 100 to 500 cm, and at 50-cm intervals below 500 cm. Undisturbed cores for bulk density (BD) determination were obtained using a 5.0×5.0 cm cutting ring. For each depth interval, subsamples were separated into air-dried and field-moist fractions for subsequent analyses and transported to the laboratory at 4°C.

Total arsenic (As), total lead (Pb), total aluminum (Al) and total iron (Fe) concentrations were determined using a portable X-ray fluorescence spectrometer (P-XRF; Thermo Fisher Niton XL3t 960, Thermo Fisher Scientific Inc., Waltham, MA, USA). The total aluminum (Al) and iron (Fe) measured by P-XRF were used as proxies for Al and Fe-oxide concentration. It was supported by data from 8-m regolith profiles (down to bedrock) at the center of the study area, where free oxides consistently accounted for $50.34\% \pm 5.62\%$ (Al_2O_3) and $50.29\% \pm 0.32\%$ (Fe_2O_3) (mean $\pm$ SD) of the sample soils, respectively^[57] and the ratio remains consistent. Before measurement, samples were air-dried at room temperature, ground with an agate mortar and pestle, passed through a 100-mesh nylon sieve (150 μm), and oven-dried at 105°C for 2 h to minimize moisture interference during XRF analysis. The instrument was operated at a tube voltage of 50 kV, a power output of 2 W, and a measurement time of 120 s, using a silver anode X-ray tube and a silicon drift detector for fluorescence signal collection.^[58–60] Each sample was analyzed in triplicate, and elemental concentrations were quantified using the instrument's built-in fundamental-parameters algorithm. Pb was measured as an auxiliary element to help distinguish shared legacy inputs from As-specific redistribution. Measurement precision was assessed through triplicate analyses, and analytical accuracy was verified using certified reference materials. The detection limits of P-XRF for total As, total Pb, total Al and total Fe in dry soil were $< 5 \text{ mg kg}^{-1}$, 14 mg kg^{-1} , 1.5 g kg^{-1} and 0.5 g kg^{-1} , respectively. In addition, the relative standard deviations of replicate measurements were low (As < 0.09 , Pb < 0.17 , Al < 0.07 and Fe < 0.04).^[59, 60] It indicated that the test results for the samples in this study were reliable.

5.3 Regolith stratification

Based on field observations and borehole descriptions, the regolith generally consisted of three horizons (Figure S8): an upper uniform red clay (URC), an intermediate reticulate red clay (RRC), and a lower weathered sandstone (WS). Across the 21 boreholes, the thickness of the URC, RRC, and WS horizons ranged from 100–335, 110–838, and 30–1545 cm, respectively. Regolith architecture was described by the vertical sequence and thickness of these horizons together with the total regolith thickness. Horizon boundaries were identified in the field according to changes in color and degree of consolidation. The RRC, located beneath the URC, was characterized by a reticulate structure composed of a red matrix with

pale vein-like networks. Previous hydrological observations at the site showed that the groundwater table typically fluctuates between 4 and 6 m depth.^[27]

5.4 Accumulation-layer definition and evaluation metrics

To characterize vertical As enrichment, we defined the arsenic accumulation layer (AAL) as the continuous depth interval in which As concentrations exceeded a baseline value of 24.1 mg kg⁻¹ dry soil. This baseline was calculated as the mean of the maximum As concentrations measured in the woodland profiles, which were used as the reference condition. Five indices were used to describe the AAL: ¹ peak As concentration (PC), ² peak depth (PD), ³ mean As concentration within the AAL, ⁴ As stock within the AAL, and ⁵ AAL thickness (AALT).

Because the RRC horizon was hypothesized to be the principal zone of As retention, we further quantified the upper and lower boundaries of the RRC, RRC thickness, the overlap between the RRC and AAL, and the contribution of the RRC to total profile As stock. Borehole O1 was used as an example to illustrate the calculation of these indices (Fig. 6). Pb concentration profiles and As/Pb concentration ratios were used as auxiliary indicators to evaluate whether deep As enrichment reflected shared legacy inputs or As-specific redistribution.

5.5 Fundamental properties and As and Pb stock quantification

Soil bulk density was determined by oven-drying undisturbed core samples at 105°C to constant mass following ISO 11272.^[61] Soil water concentration (SWC) was determined gravimetrically from field-moist subsamples after oven-drying at 105°C to constant mass. Particle-size distribution, including sand, silt, and clay fractions, was measured using a laser diffraction particle-size analyzer (LS230, Beckman Coulter, USA), clay and silt refer to fine particles, while sand refers to coarse particles. Soil pH was measured using an electronic pH meter in a 1:2.5 soil-to-deionized water suspension.

As and Pb stocks for each depth increment were calculated as:

$$S = C \times SBD \times d \times 0.1$$

where S is As or Pb stock per unit area (kg ha⁻¹), C is As or Pb concentration (mg kg⁻¹), SBD is soil bulk density (g cm⁻³), and d is the thickness of the sampling interval (cm). Total As and Pb stocks for each profile were obtained by summing stocks across all depth increments.

5.6 Data analysis

Because long-term monitoring data were unavailable, a space-for-time substitution approach was used to compare sites with similar environmental settings but different land-use histories. Land-use comparisons were conducted using boreholes as the statistical units, whereas depth-resolved analyses

were used to evaluate profile-scale associations between As concentration, Pb concentration, As/Pb ratios, regolith structure, and physicochemical properties.

One-way analysis of variance (ANOVA), followed by least significant difference (LSD) tests, was used to compare AAL metrics and regolith properties among land-use groups at $p < 0.05$. Pearson correlation analysis and simple linear regression were used to examine relationships among As concentration, As/Pb ratios, regolith physicochemical properties, and regolith structural indices. Random forest models were built in R using the “randomForest” package to assess the relative importance of Al_2O_3 , Fe_2O_3 , clay and silt, sand, SWC, SBD, and pH in explaining vertical variation in As concentration, Pb concentration and As/Pb concentration ratios. Model performance was evaluated using the percentage of variance explained, and predictor importance was quantified using relative importance metrics. All statistical analyses were performed in R.

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Figures

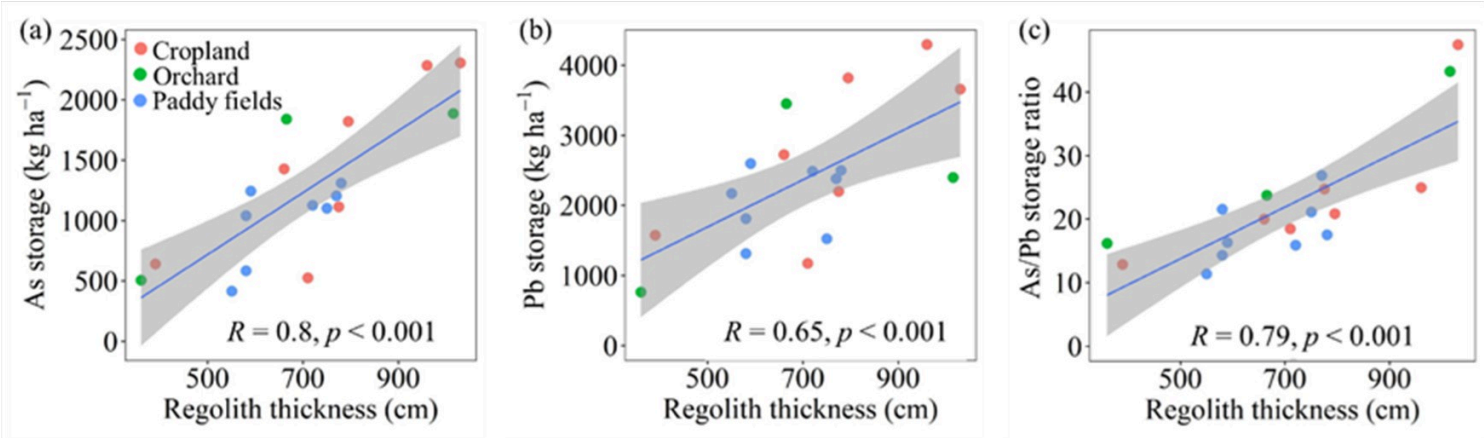


Figure 1

Linear relationships between regolith thickness and (a) arsenic (As) storage, (b) lead (Pb) storage, and (c) As/Pb storage ratio across agricultural regolith profiles. Significance levels are shown in each panel.

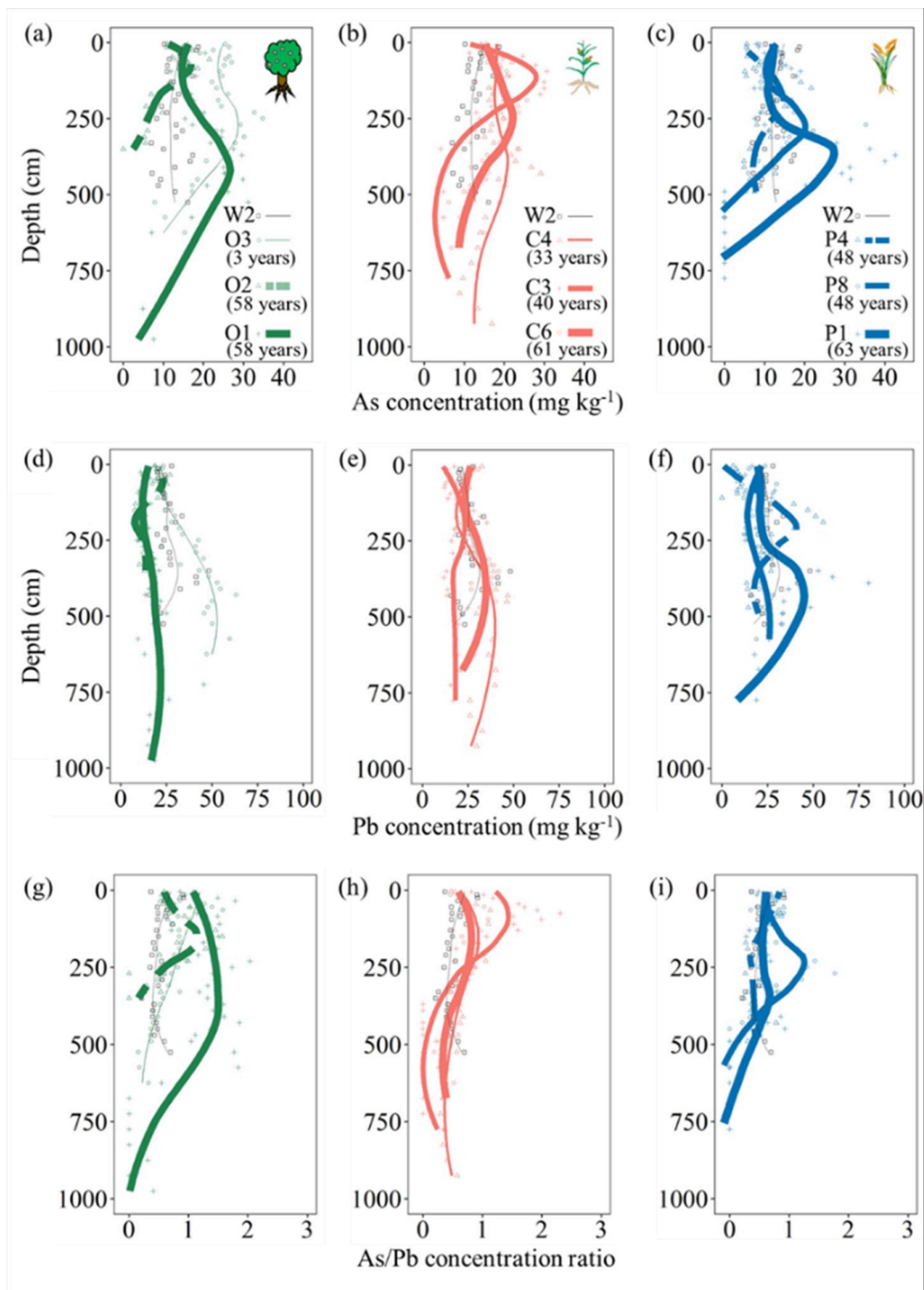


Figure 2

Vertical profiles of regolith As concentration, Pb concentration, and As/Pb concentration ratio under different land-use types. Panels (a–c) show the As concentration profiles under orchard, cropland, and paddy-field, respectively, with woodland as the reference. Panels (d–f) show the corresponding Pb concentration profiles. Panels (g–i) show the As/Pb concentration ratios. C, O, P, and W represent cropland, orchard, paddy-field, and woodland, respectively.

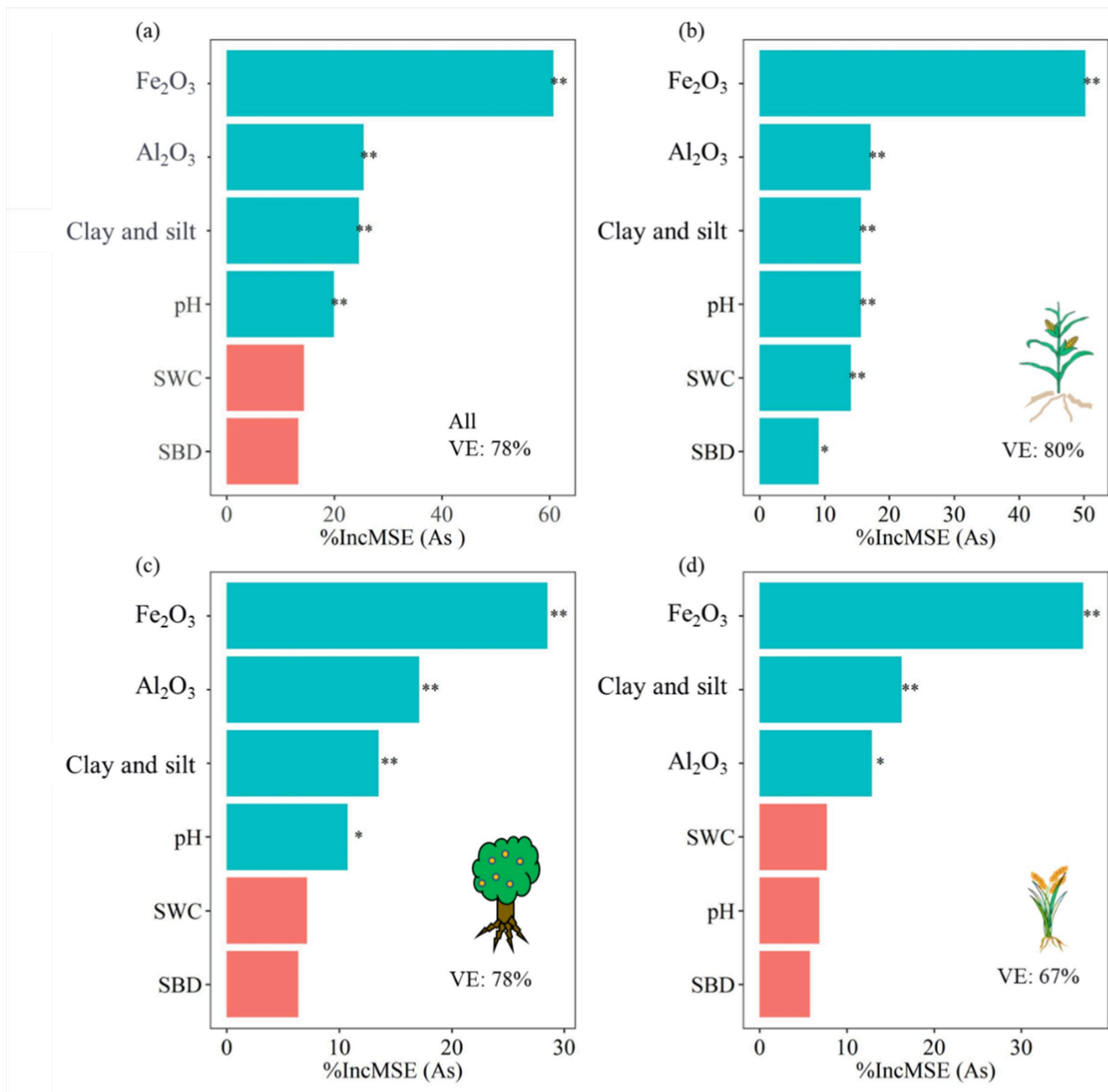


Figure 3

Relative importance of regolith properties, including Al₂O₃, Fe₂O₃, clay and silt, soil water concentration (SWC), soil bulk density (SBD), and pH, in explaining As concentration in the Random Forest models for (a) the full dataset, (b) cropland, (c) orchard, and (d) paddy-field. Asterisks indicate significant predictors (*p < 0.05). VE represents the percentage of variance explained by the model.

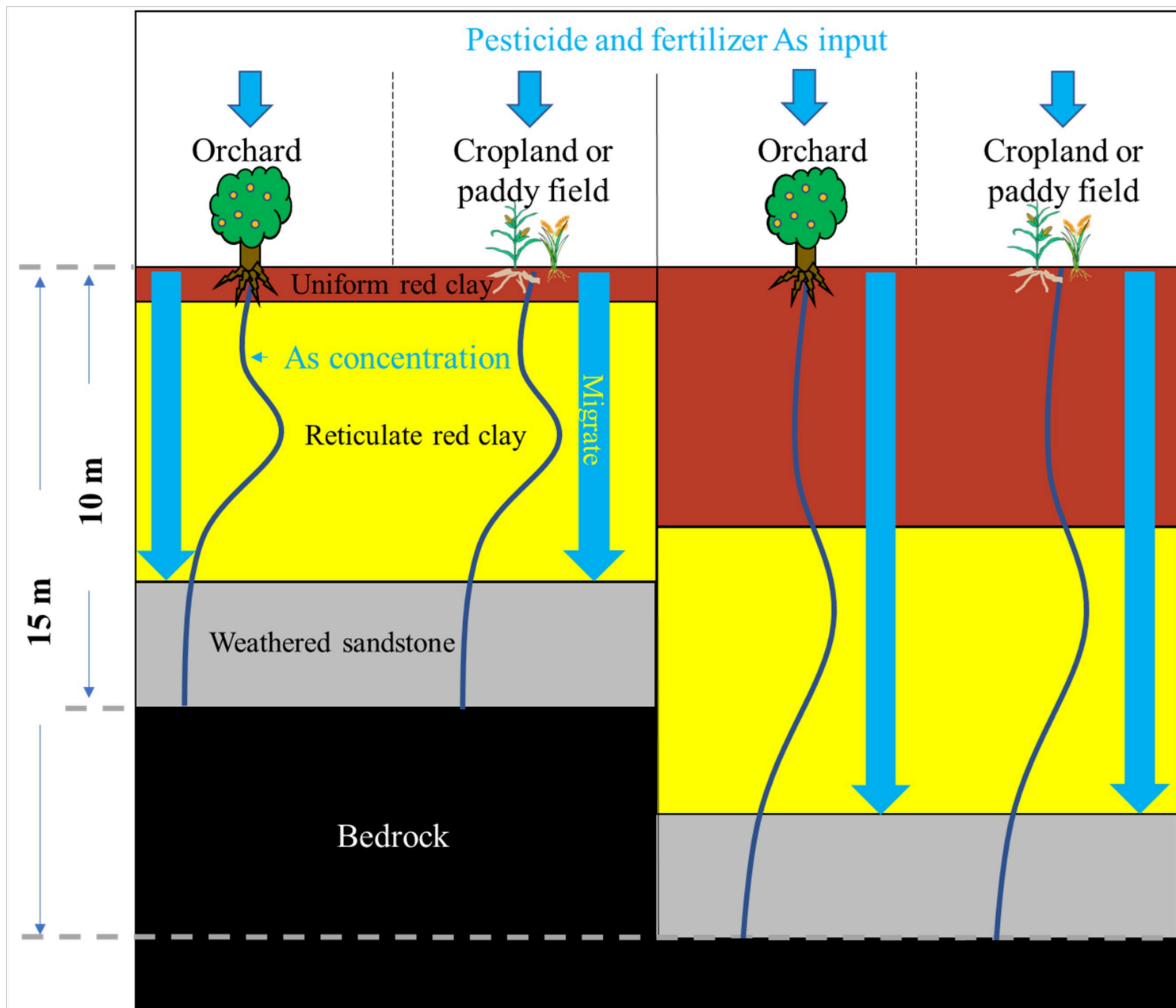


Figure 4

Conceptual diagram illustrating how regolith architecture and land-use history jointly regulate deep As accumulation in subtropical red-soil profiles. The RRC horizon acts as a deep reactive retention zone, while orchard and paddy-field management may enhance As redistribution through distinct biological and hydrological pathways.

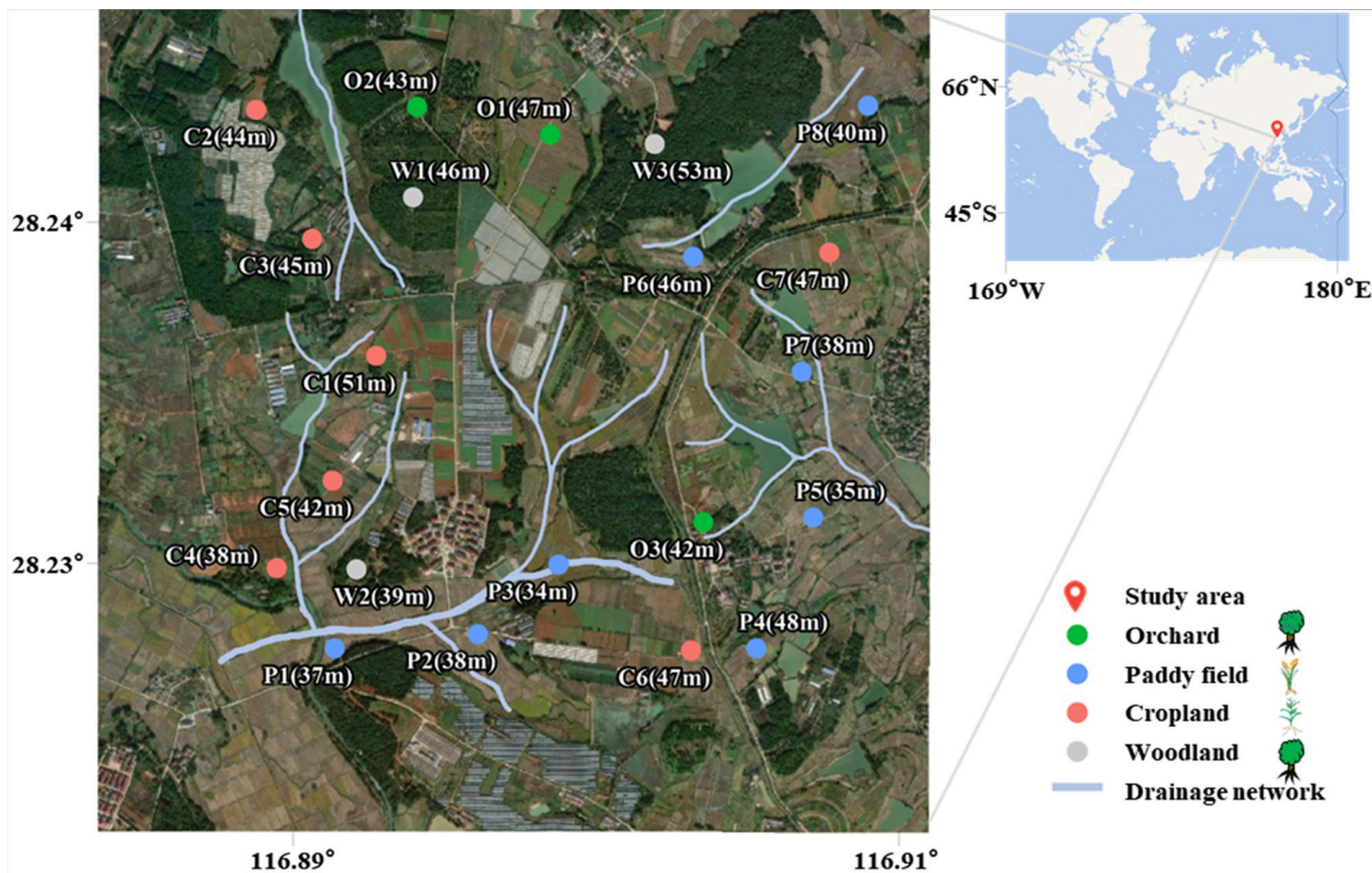


Figure 5

Location of the 4 km² (2 × 2 km) study area and spatial distribution of drilling boreholes. C, O, P, and W refer to cropland, orchard, paddy-field, and woodland, respectively. Numbers indicate the sampling order during field investigation, and elevation for each borehole is shown in brackets. All boreholes were used for profile description; boreholes included in each statistical analysis are indicated in the corresponding results.

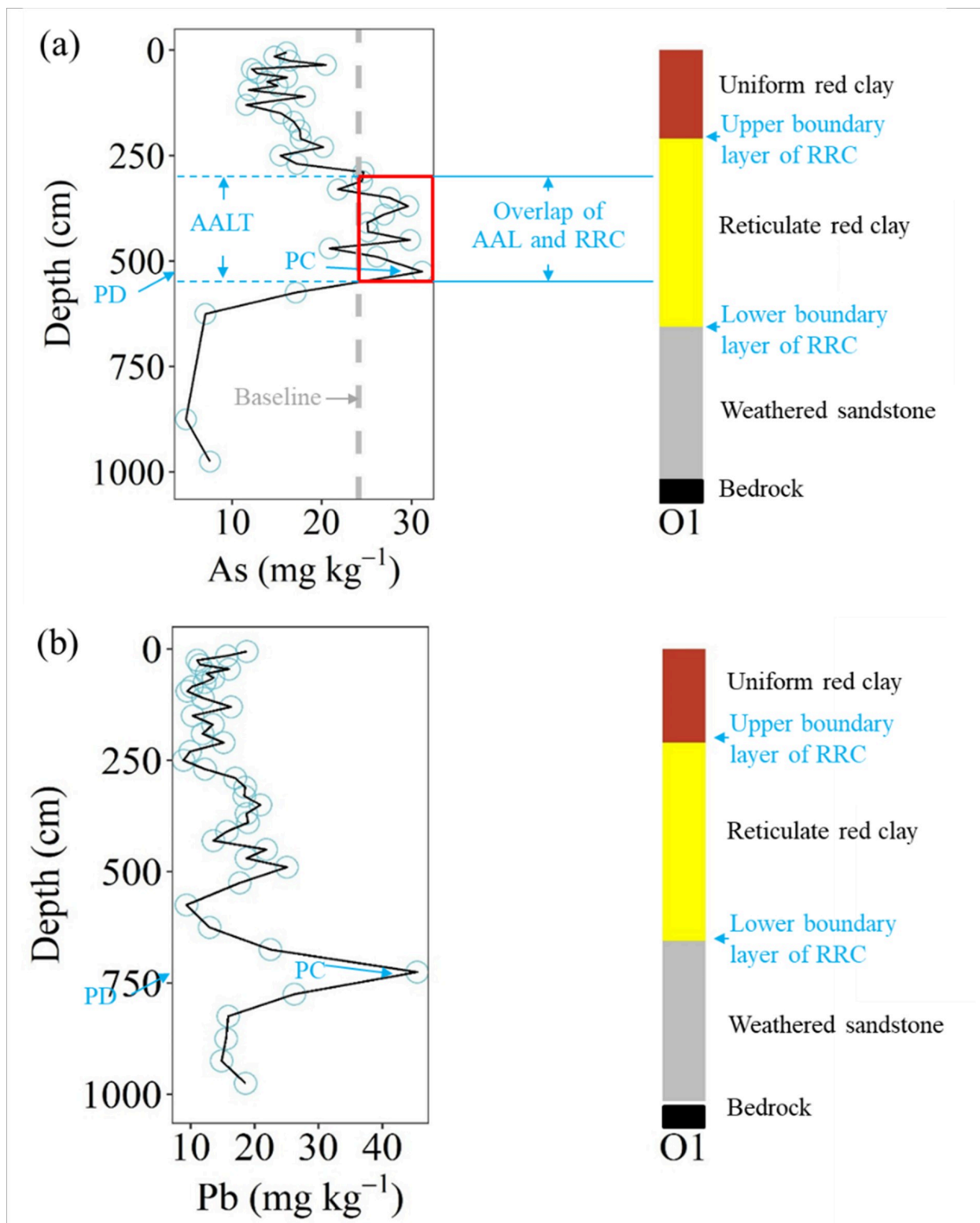


Figure 6

Illustration for the (a) As and (b) Pb accumulation layer (AAL) evaluation indices (peak concentration (PC), peak depth (PD) and the thickness of the AAL (AALT)) in the entire regolith profile. The arsenic storage within the AAL and the mean arsenic concentration within the AAL were calculated using the data within the AAL. The red box represents the AAL. Data of borehole O1 is used as an example.

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