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SPECIAL FEATURE: ORIGINAL ARTICLE

Recent researches on Thermal Treatment and Emission Control (9th i-CIPEC)

Lead contamination in surface soil on roads from used lead–acid battery recycling in Dong Mai, Northern Vietnam

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Abstract Used lead–acid battery (ULAB) recycling has caused numerous health and environmental issues in developing countries. Surface soil pollution from ULAB recycling activities has been linked with elevated levels of lead in human blood. We measured surface soil lead in and surrounding the ULAB recycling village of Hung Yen in northern Vietnam in 2011, 2013, and 2014. The data were analyzed statistically and discussed with respect to distance from the contamination source, year of measurement, contamination pathway, and countermeasures against the contamination. Transportation routes from the smelter or collection site displayed the greatest concentration of surface soil lead (median 6400–10,000 mg/kg). Surface soil lead decreased significantly with distance along the road from the ULAB recycling site, although such a decrease was not observed for rice fields, agricultural roads, or garden soil. Re-suspension and adherence by traffic were identified as key pollution pathways. Distance from the source, covering of the surface of roads, construction of walls, and position relative to the source were shown to be the most effective factors in the reduction of surface soil lead pollution. Application of a combination of these measures should result in improvement in the health of residents.

Keywords Used lead–acid battery · Lead · Surface soil · Field portable X-ray fluorescence

Introduction

Lead (Pb) is known to have serious toxic effects. Health problems from extensive environmental Pb contamination have occurred in many parts of the world. Lead mines, primary and secondary lead smelters, and used lead–acid battery recycling plants scatter high-concentration lead to the surrounding environment and threaten the health of plant workers, local residents and, in particular, children [1–3]. Although developed countries have largely eliminated these sources of Pb contamination, lead poisoning remains a serious concern in developing countries. Among the various sources of Pb contamination in developing countries today, lead–acid battery manufacturing and recycling plants are associated with several of the most

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critical issues relating to lead poisoning. Reviewing papers in 37 countries published from 1993 to 2010, Gottesfeld and Pokhrel identified that the blood lead levels of battery plant workers were substantially higher in developing countries than in the US, selected to be representative of developed countries [4].

Several environmental matrices related to raised blood lead levels have been reported, including air, soil, and house dust [5–7]. Zahran et al. used a broad data set of 367,839 children (aged 0–10) to demonstrate a clear relationship between children's blood lead levels and re-suspension of Pb-contaminated soil (i.e., pathway soil → air dust → child) [7]. Furthermore, an atmospheric transport and dispersion model demonstrated that the deposition profile derived from fugitive dust Pb emission from a battery recycling site was significantly correlated with the observed spatial pattern of residential soil Pb [8]. In some cases, dust Pb emission rates were greater than stack Pb emissions [9]. Hence, as major pollution sources of both human and environmental matrices, it is important to measure the contamination levels of soil Pb in and around battery recycling sites. Furthermore, surface soil Pb is readily re-suspended by wind and traffic.

Recently, high levels of Pb concentration in human blood, urine, and hair were reported at the used lead–acid battery (ULAB) recycling village of Hung Yen, Vietnam [10]. To identify measures to prevent irremediable health effects, knowledge of the nature and pathways (re-suspension, adherence, and deposition) of the environmental lead contamination is required. However, before this study, no systematic data set existed for lead levels in surface soil from this area. In addition, there is a need to assess and implement effective countermeasures based on quantitative Pb measurement to address contamination in the near future. In this study, we examined in detail lead pollution in surface soil in and around the ULAB recycling village.

Focusing on the types of road and relative positions from the site thought to be the contamination source, Pb concentrations in surface soil were measured systematically on-site using X-ray fluorescence (FP-XRF). Statistical comparison of measurements made at approximately equal distances from the recycling smelter indicated the contamination pathway. We discuss the identified decreasing Pb concentration with increasing distance and position of the roads relative to the Pb contamination source. In addition, the effectiveness of countermeasures—such as covering the surface of the roads and construction of shielding walls—was assessed by means of multiple-year Pb measurements (2011, 2013, and 2014).

Materials and methods

Location

We selected Dong Mai (hereafter DM) village in Van Lam district, Hung Yen province, Vietnam, as our study site and visited the village and its surroundings three times in January 2011, 2013, and 2014 [Fig. 1 and Fig. S1 (satellite image)]. DM village is a small used lead–acid battery (ULAB) recycling craft village with approximately 2300 residents [10]. Two smelters, S1 and S2, are located to the south of DM village (>500 m) and at its southern edge, respectively (Figs. 1 and S2). An older smelter site, S3, is closed and is located to the southwest of the village (>200 m). S1 was constructed recently and has been in operation since 2009. S2 was in operation for 9 years, from 2005 to 2013. At the same site as S2, workers dismantle large numbers of ULABs and discharge acid waste liquid from the ULABs into the ground at the site (Fig. S3). After closing smelting operations at S2, a new smelter was constructed adjacent to the site of S1 and has been in operation since 2013. Dismantling of ULABs at the site of S2 continues to date. Collection sites for ULABs and related materials (labeled 'C' in Fig. 1) are located at the southern edge of DM village.

Measurement of lead concentrations

On-site field portable X-ray fluorescence (FP-XRF) can be used to measure Pb concentrations in small surface soil areas (measurement window diameter, 2 cm). Previous studies have applied cost-effective [11] FP-XRF for on-site measurements of Pb concentration in surface soil [6, 12–19]. Pb concentrations in the surface soil were measured at >500 points at 5–50-m intervals in and around DM village using an FP-XRF device [Innov-X alpha (Innov-X System, Inc.)], which has been used in various recent on-site soil studies [6, 12, 15, 16, 18, 19]. The FP-XRF has a "soil-mode" setting for the measurement of Pb concentrations in soil samples [6, 15, 18, 19]. Measurement at each point required ~30 s. Care was taken to remove foreign objects at the measurement points, where possible, such as grasses and gravel. The FP-XRF detection window was cleaned before each measurement with a pre-moistened wipe to remove any residual from the previous sample. The average recovery ratio for the matrices was 81 %. The average relative standard deviation (RSD) was 6.0 % (range 2.9–16 %). Moisture content of the soil affects measurement value using FP-XRF [20, 21]. In addition,

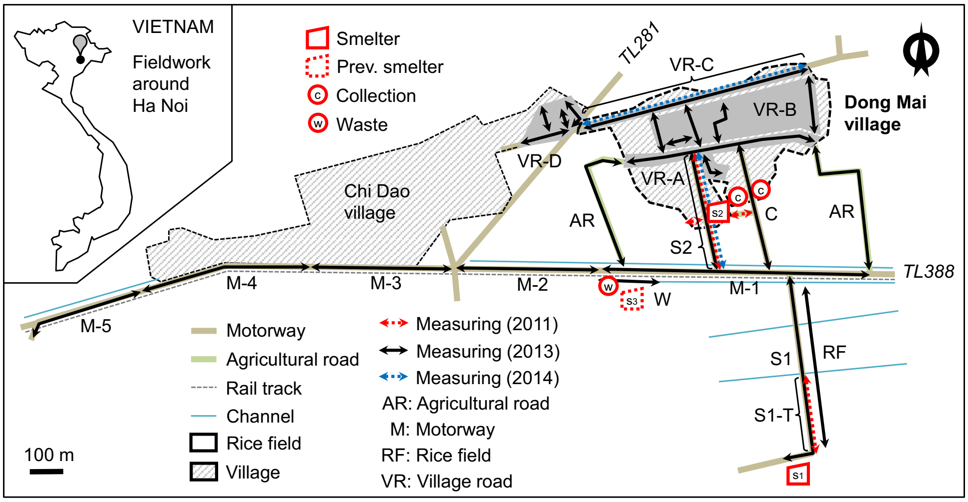

Fig. 1 Study area in and around Dong Mai (DM) village, Hung Yen province, Vietnam, from 2011 to 2014

large biases in the on-site FP-XRF measurements are caused by inadequate soil preparation and heterogeneous soils [20, 21]. To avoid such bias, we used statistically representative values with deviations of various subsets and subjected these values to statistical analysis [17, 18]. For example, we categorized the FP-XRF data from 2013 into 15 groups and then compared each Pb concentration statistically (Fig. 1). Data set was analyzed by the statistical method. Detailed analytical procedure is described in Electronic Supplementary Material.

Results and discussion

Village

In 2013, we measured surface soil Pb concentrations at 131 points in DM village to assess the Pb contamination level. Vietnamese standard soil Pb values for residential and industrial areas have been established at 120 and 300 mg/kg, respectively [22]. Of all measurement points in DM village, 99 and 98 % were greater than the residential and industrial maximum standards, respectively. The frequency distribution indicated the 95th percentile point at 500 mg/kg (Fig. S4) and 35th percentile point at 2000 mg/kg. The blood Pb levels of 93 residents (including 23 children) in the village were surveyed in January 2011 [10]. All residents displayed 14–122 (median, 34) µg/dL lead in blood, which exceeds the minimum toxic threshold (10 µg/dL blood) recommended by the Agency for Toxic Substances and Disease Registry [1]. These measurements showed an environmental pollution level of surface soil in DM village comparable to severe cases in the suburbs of Dakar, Senegal, in which deaths of children, involved in ULAB recycling, were reported [6].

Road pollution

In 2013, we measured surface soil Pb at 384 points in and around DM village. The data were categorized into 15 groups, according to the location and characteristics of the roads from which samples were taken. We found severe Pb contamination on the roads of S1 (to the smelter S1), S2 (across the smelter S2 site), and C (across the collection sites Cs). Median Pb concentrations of roads S1, S2, and C were 10,000 ($n = 40$), 7200 ($n = 26$), and 6400 ($n = 20$) mg/kg, respectively (Table S1). Road S1 displayed the highest Pb concentrations in all 15 categories [Fig. 2 ($p < 0.05$)]. Roads S2 and C displayed Pb concentrations similar to those of roads S1 and W (in front of the waste yard, Fig. S5), and significantly higher Pb concentrations than village roads (VRs), the agricultural road (AR), the rice field (RF), and the highways (Ms) ($p < 0.05$). Thus, the smelters (S1 and S2) and collection sites of ULABs and related materials had serious impacts on road surface soil Pb levels, because they connect the entrances of the plants.

The front of the waste yard (W) also showed high-level Pb contamination of the surface soil. The median Pb concentration in W, 3100 mg/kg ($n = 9$), was statistically comparable with the levels on roads S2 and C (Fig. 2). Figure 1 shows W, which was located near the inactive smelter S3 (photo in Fig. S2). Possible sources of surface soil Pb were waste from DM village and materials from smelter S3. The surface soil in front of the waste yard (W) displayed some of the highest Pb surface soil concentrations around DM village.

Surface soil Pb plays a critical role in elevating human blood lead levels [7], and fugitive dust Pb contributes to surrounding environmental matrices by re-suspension of surface soil Pb [8, 9, 23, 24]. Therefore, surface soils contaminated with the highest level of Pb on roads S1, S2,

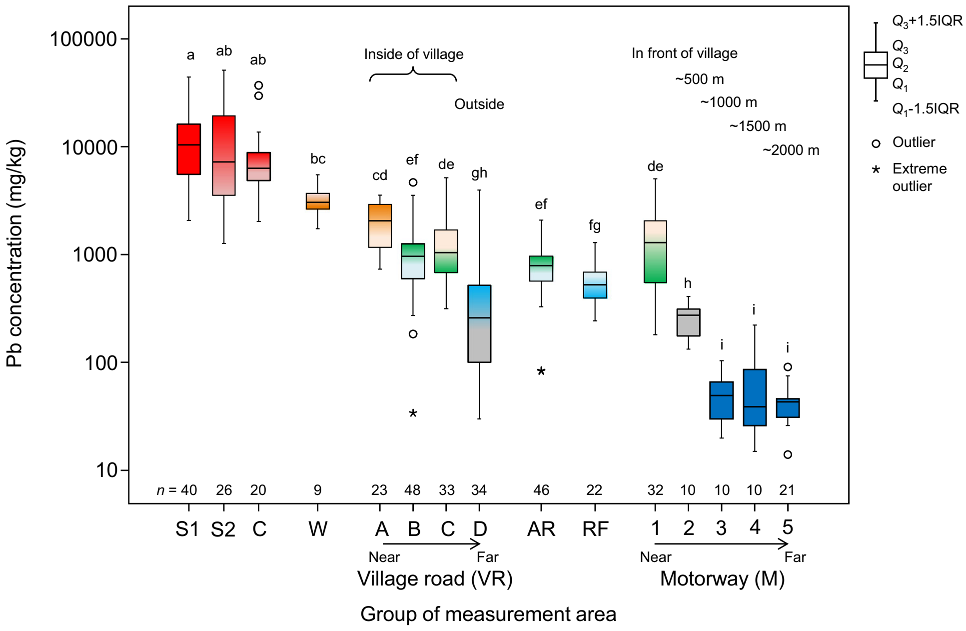

Fig. 2 Pb concentrations in surface soil in 2013; group names are defined by type of sampling location, *different letters* (*a*, *b*, …, *i*) at the top of each column represent significant differences ($p < 0.05$), *same letters* indicate not significant

C, and W were also likely to be sources of Pb contamination of the surrounding environment.

Distance and position

Figure 3a shows the average Pb concentrations with maximum and minimum values at 0–180 m from smelter S2. Linear regression shows that the Pb concentrations in surface soil on the road to TL388 (I) and on the road to the village (II) decreased significantly ($p < 0.01$) with distance from the entrance of smelter S2 (Fig. 3a). The half-distance of Pb concentration ($d_{1/2}$) on road I was 47 m (95 % CI 31–63 m). The $d_{1/2}$ on road II was 45 m (95 % CI 28–62 m). Statistical modeling showed a consistent trend of decreasing soil Pb with increasing distance along the road at the side of a ULAB site in the US [8]. In contrast, we found no significant trend of decreasing Pb concentration with distance in surface soils in the garden surrounded by a concrete wall (III) or on the agricultural ridge road (IV). Roads I and II were busy transportation routes for ULABs and materials produced by smelter S2. Pb contamination due to this activity was not observed in surface soils from III and IV, whereas decreasing surface soil Pb with increasing distance was evident on the transportation route at the nearby smelter.

Road S1 was divided into three subgroups of 180 m length, S1-A, -B, and -C. Median Pb concentrations in S1-A, -B, and -C were 15,000 ($n = 15$), 6500 ($n = 8$), and

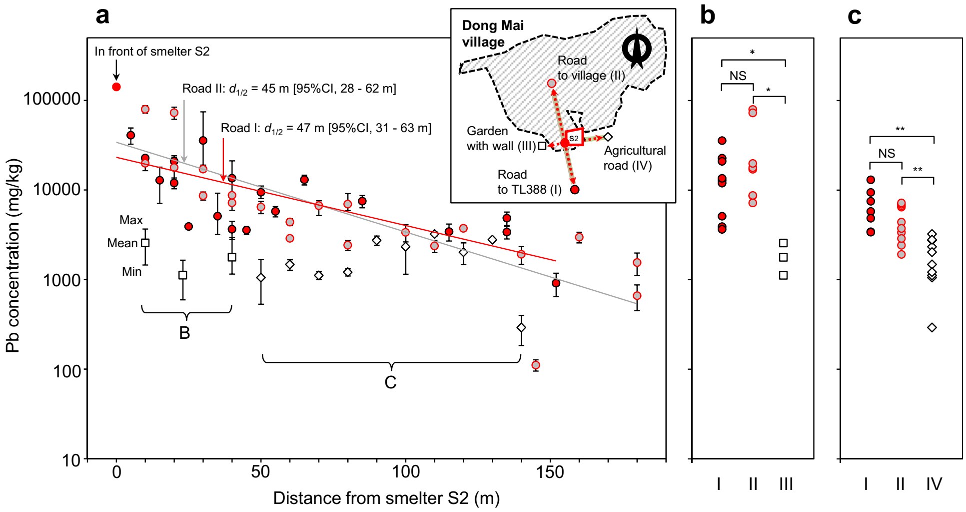

Fig. 3 Pb concentrations at nearside of smelter S2 in 2011; **a** Pb concentration as a function of distance in four directions from smelter S2, **b** comparison of Pb concentrations at equal distances in three directions (10–40 m, labeled as *B* in **a**), **c** comparison of Pb concentrations at equal distances in three directions (50–140 m, labeled as *C* in **a**), $*p < 0.05$, $**p < 0.01$, NS is not significant

5100 ($n = 8$) mg/kg, respectively (Table S1). Compared with the 0–180-m zone in S1-A, surface soil Pb displayed significantly lower Pb concentrations over the 180 m of S1-B and -C [Fig. 4b ($p < 0.05$)]. In contrast, in the rice field, located a similar distance from smelter S1, we found increasing surface soil Pb concentration with increasing distance (RF, photo in Fig. S5). Subgroups RF-A, -B, and -C had median Pb concentrations of 350 ($n = 8$), 530 ($n = 6$), and 690 ($n = 8$) mg/kg, respectively (Table S1). Road S1 functioned as a transportation route to smelter S1, while there was no transportation traffic on RF. An inverse correlation between Pb concentration and distance was also found on road S2, which was used as a transportation route. Thus, usage as routes for transportation from the ULAB recycling plant had a significant impact on the trend of decreasing surface soil Pb concentration with increasing distance from the plant.

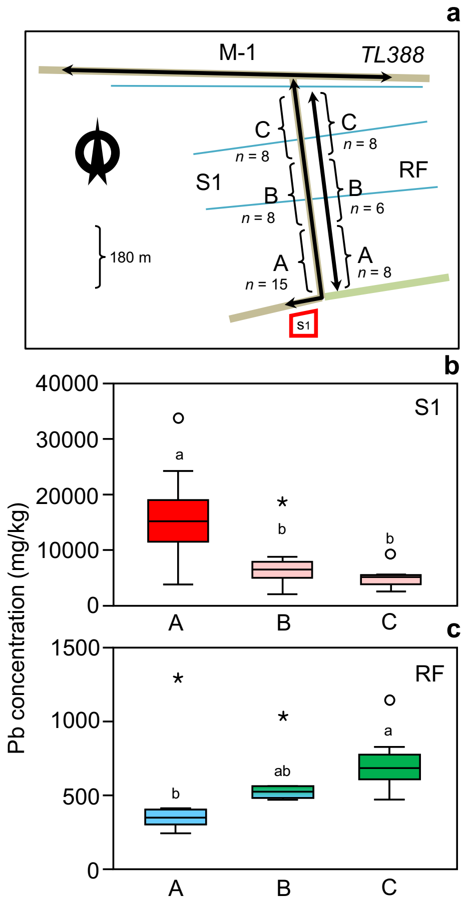

Fig. 4 Pb concentrations on the road to smelter S1 (S1) and rice field (RF) in 2013; **a** S1 and RF are divided into three 180-m intervals, *A*, *B,* and *C*, correspond to the proximity to smelter S1, **b** Pb concentrations on S1-A, -B, and -C, and **c** that in RF-A, -B, and -C, the *letters a* and *b* at the top of each column represent significant differences ($p < 0.05$)

Village roads in DM and neighboring villages were categorized into four groups, VR-A, -B, -C, and -D, according to their proximity to smelter S2 and collection sites, Cs (Fig. 1). The median Pb concentrations of VR-A, -B, -C, and -D in 2013 were 2100 ($n = 23$), 970 ($n = 48$), 1000 ($n = 33$), and 260 ($n = 34$) mg/kg, respectively (Table S1). VR-A showed significantly lower Pb concentrations than roads S2 and C, and significantly higher Pb concentrations than VR-B and -D ($p < 0.05$), although there was no significant difference between the Pb concentrations of VR-A and -C (Fig. 2). Surface soil Pb on the DM village road might be affected by its junction with the contaminated roads S2 and C. VR-D, where it crossed highway TL281 from DM village (Fig. 1), displayed minimal Pb concentrations compared with DM village roads VR-A, -B, and -C ($p < 0.05$). The median Pb concentration of VR-D was only ∼4 % of that on the contaminated roads S2 and C. Thus, it appears that the neighboring village was protecting the road VR-D from Pb contamination by keeping the road at a suitable distance from the recycling village. Although no ULAB recycling activities take place in this village, which is to the west of DM village, the median Pb concentration on VR-D exceeded the residential standard of 120 mg/kg [22]. Therefore, those who live in this neighboring village should pay attention to contamination of surface soil Pb instead of original recycling village.

Agricultural road (AR), which is less densely populated, connects DM village road VR-A and highway M-1 (Fig. 1 and ref. photo in Fig. S5). The median Pb concentration on AR (790 mg/kg, $n = 46$ in Table S1) was greater than the soil industrial standard (300 mg/kg [22]) and statistically comparable to those of VR-B, -C, and M-1 (Fig. 2). Surface soil Pb displayed high concentrations near the junction with VR-A and M-1, indicating that AR might be affected by the high Pb concentration in the surface soil at the junction.

Highway M-1, which connects with the contaminated roads S1, S2, and C, displayed statistically identical Pb concentrations to DM village roads VR-A, -B, and -C (Figs. 1, 2). Similarly to VR-A, the junction with these contaminated roads might be involved in contamination of surface soil Pb on M-1. Median Pb concentrations of M-1, -2, -3, -4, and -5 were 1300 ($n = 32$), 270 ($n = 10$), 50 ($n = 10$), 39 ($n = 10$), and 43 ($n = 21$) mg/kg, respectively (Table S1). The sequence of Pb concentration followed M-1 > M-2 > M-3 = M-4 = M-5, in which > indicates significantly greater than, and = indicates not significant ($p < 0.05$). The lowest Pb concentrations were found in surface soil at distances greater than 1000 m from DM village (i.e., highways M-3, -4, and -5) [Fig. 2 ($p < 0.05$)]. Hence, Pb contamination in surface soil might be reduced to sufficiently low levels at sites >1000 m from the ULAB recycling area along the highway. This result substantiates the finding of an inverse correlation between distance from source and Pb concentration.

Year

Median Pb concentrations at S1-T in 2011 and 2013 were 1300 ($n = 20$) and 13,000 ($n = 20$) mg/kg, respectively (Table S1). Surface soil Pb in 2013 on S1-T increased tenfold in 2 years [Fig. 5a ($p < 0.01$)]. The lower surface soil Pb concentration at S1-T was likely because smelter S1 began operating in 2009, and prior to this, there was no transportation traffic on the road. After the smelter began operations, road S1 was used primarily as a transportation route. Hence, we suggest that this change in use of road S1 was a factor in the increase in surface soil Pb on S1-T over the 4-year period from the start of operation of smelter S1.

Surface soil Pb on road S2 was measured three times between 2011 and 2014. The median Pb concentrations in 2011, 2013, and 2014 were 6600 ($n = 42$), 7200 ($n = 25$), and 9700 ($n = 42$) mg/kg, respectively (Table S1). Statistical comparison indicated that surface soil Pb did not change during the 3-year period (Fig. 5b). Smelter S2 has operated since 2005. In 2011, road S2 was being used as a transportation route and 6 years had passed from the start of operation of smelter S2. Pb concentrations on road S2 for 2011–2014 were statistically comparable to those on road S1(-T) in 2013. Therefore, surface soil Pb concentration on transportation routes to and from the nearby ULAB recycling sites increased gradually, reaching unacceptably high values by the end of the 4-year period.

Village road VR-C was covered with concrete in June 2013. We measured surface soil Pb on VR-C before and after covering to assess its impact. Median Pb concentrations in 2013 and 2014 on VR-C were 1000 ($n = 33$) and 350 ($n = 28$) mg/kg, respectively (Table S1). Surface soil Pb on VR-C decreased significantly from 2013 to 2014 [Fig. 5c ($p < 0.01$)]. Covering the road with concrete decreased surface soil Pb, although the trend in Pb concentration over time on VR-C should be monitored.

Pathway

Figure 6a shows three pathways of Pb contamination of the surface soil: re-suspension, adherence, and deposition. Re-suspension and adherence occur primarily on the transportation route and are caused by differences in traffic type and wind. Surface soil is re-suspended by traffic and then scattered to the surrounding soil. Pb moves from the original surface soil to other areas through re-suspension. Surface soil Pb directly or indirectly adheres to various parts of vehicles as well as to residents. Adhered soil dust is transported by lifting and deposition onto other soil surfaces. Transfer of Pb to the atmosphere originates from the stack gas emissions of the ULAB recycling smelter.

Since road S1 and the rice field (RF) are located at almost equal distance from smelter S1, the contribution of atmospheric deposition to the soil surface is expected to be comparable at each site. However, we found a significant difference in Pb concentration between road S1 and RF [Fig. 2 ($p < 0.05$)]. Compared with median Pb concentrations (Table S1), subgroups S1-A, -B, and -C displayed 43-, 12- and 8-fold higher Pb concentrations than RF-A, -B and -C, respectively [Fig. 4b ($p < 0.01$)]. Atmospheric deposition alone does not explain this significant difference, and it can be concluded that other pollution pathways are involved.

In 2011, smelting was conducted at the site. Surface soil Pb concentrations close to smelter S2 (10–40 m) along three transects in different directions were compared statistically (Fig. 3). Although these surface soils had equal potential for Pb contamination via atmospheric deposition, surface soils on transportation routes, such as along the road to TL388 (I) and to the village (II), had significantly

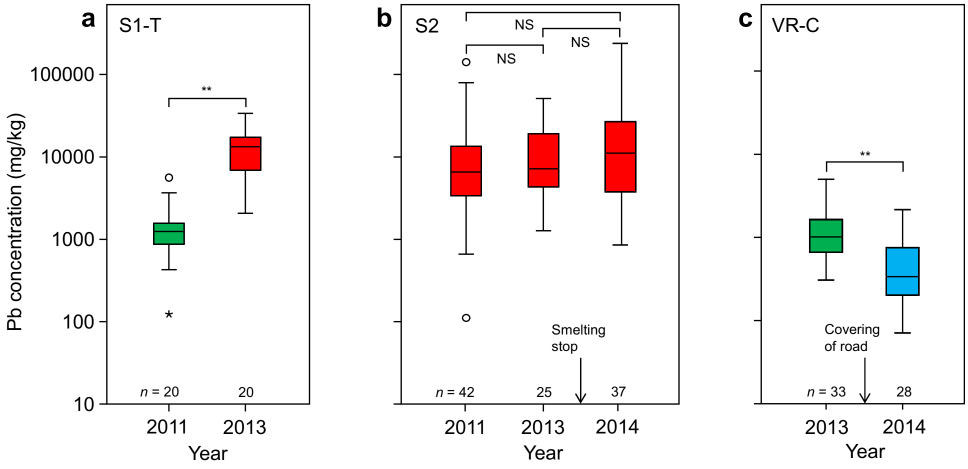

Fig. 5 Time trends of Pb concentrations in surface soil at three roads, **a** S1-T, **b** S2, and **c** VR-C; the three roads used in the comparison are denoted by *curly brackets* in Fig. 1, $^{**}p < 0.01$, NS is not significant

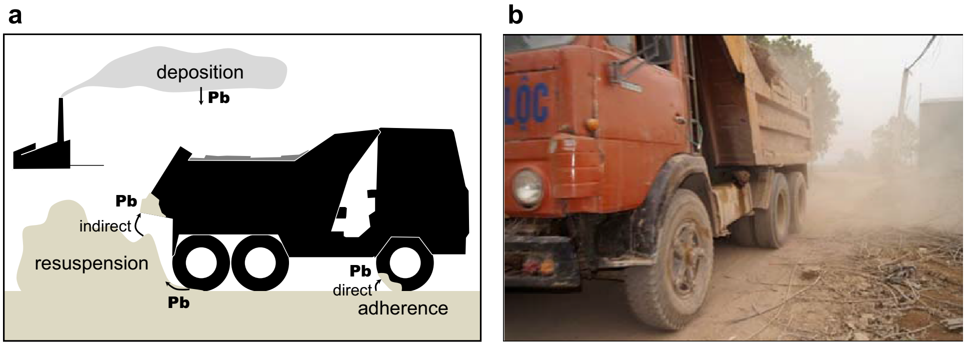

Fig. 6 Conceptualization of lead contamination pathways to surface soil in and around ULAB recycling sites; **a** concept panel of re-suspension, adherence, and deposition, and **b** a representative photo of re-suspension by heavy truck

higher Pb concentrations than surface soils in the garden surrounded by concrete wall (III) [Fig. 3b ($p < 0.05$)]. Median Pb concentrations in I, II, and III at 10–40 m were 13,000 ($n = 9$), 17,000 ($n = 8$), and 1800 ($n = 3$) mg/kg, respectively (Table S1). Pb concentrations in surface soils I and II were seven-to-nine-fold higher than in surface soil III. We also analyzed surface soils along three transects in different directions over distances of 50–140 m from smelter S2. Atmospheric deposition displayed a similar influence on these surface soils. However, surface soils from transportation routes I and II showed significantly higher Pb concentrations than soils from the agricultural ridge road (IV) [Fig. 3c ($p < 0.01$)]. Median Pb concentrations in I, II, and IV at 50–140 m were 5800 ($n = 7$), 4100 ($n = 10$), and 1700 ($n = 10$) mg/kg, respectively (Table S1). Pb concentrations in surface soils I and II were two-to-three-fold higher than that in surface soil IV. These statistical analyses corroborated the importance of re-suspension and adherence as pathways of Pb contamination along the transportation routes.

Similar Pb concentrations on road S2 before and after the closure of smelting operations suggest that atmospheric deposition from stack gas emissions is not an important contamination pathway (Fig. 5b). The dismantling of the smelter was a significant source of Pb contamination of surface soil. Sulfuric acid contaminated with lead from the ULABs was discharged directly into soil (Fig. S3a). We also observed dispersion of Pb contamination on road S2 due to adherence to truck tires of liquid containing Pb discharged from ULABs (Fig. S3b).

Heavy trucks passed along roads S1 and S2 to and from smelters S1 and S2, respectively. Although roads in urban areas have asphalt surfaces, many roads in DM village were unpaved. Roads S1 and S2 were dusty, because they are unsealed by asphalt. Thus, we observed considerable re-suspension and adherence caused by heavy trucks on roads S1 and S2 (Fig. 6a, b). The quantity of Pb re-suspended per vehicle passage increased as a function of vehicle speed, and was greater for heavy trucks than passenger cars [25]. Lough et al. reported that increased emission rates of surface matrices by re-suspension were related to a higher proportion of heavy trucks [26]. Therefore, we hypothesized that re-suspension and adherence would have important effects on contamination pathways. Data from transportation routes and emission stacks were subjected to statistical analysis. The effects on Pb concentrations of re-suspension and adherence should be assessed in future studies.

Conclusions

Increasing distance from the source results in a significant reduction in Pb surface soil concentration. The half-distance of Pb concentration ($d_{1/2}$) along the transportation route was 45–47 m within a 180-m radius from the ULAB recycling plant S2 (Fig. 3a). Surface soil Pb along the transportation route decreased significantly at >180 m from smelter S1 [Fig. 4b ($p < 0.05$)]. Measurements along the highway (TL388) (Fig. 2) revealed that surface soil Pb concentration 500–1000 m from DM village (M-2) was one-fifth of the concentration on the highway in front of DM village (M-1) ($p < 0.05$). Surface soil Pb concentration at >1000 m from DM village (M-3, -4, and -5) was one-fifth to one-seventh of that on M-2 ($p < 0.05$). We suggest that residential areas should be situated at least 1 km distant from ULAB recycling plants.

Village road VR-C displayed a significant decrease in surface soil Pb after the road was covered [Fig. 5c ($p < 0.01$)], with a reduction in Pb concentration of two-thirds compared to that on the uncovered road in January 2014, less than 1 year after the road was covered. It is possible, however, that the surface soil Pb on the covered road might increase in the future, as occurred on road S1-T (Fig. 5a). Figure 3b shows that garden soil surrounded by a concrete shielding wall (III) exhibited one-seventh to one-tenth lower Pb concentrations than surface soil on parts of transportation route S2 (I and II) ($p < 0.05$). Because the wall was greater than 170 cm in height, it was difficult for soil dust to admix into garden soil by re-suspension and adherence. We advocate the building of walls as a simple and effective measure to prevent surface soil Pb contamination.

Roads S1, S2, and C were the most contaminated transportation routes. Village road VR-A and highway M-1 connected directly with these roads (Fig. 1). Surface soils in VR-A and M-1 contained Pb derived from secondary contamination (Fig. 2). Thus, a junction with a highly contaminated road had the potential to spread surface soil Pb to the roads connecting to the junction. The construction of housing along transportation routes (i.e., roads S1, S2, and C) should be avoided; moreover, roads directly connected with transportation routes are unfavorable for dwellings. The significantly lower Pb concentrations on the agricultural ridge road (IV) compared to points along transportation track S2 (I and II) ($p < 0.01$, Fig. 3c) provided additional evidence to support the avoidance of positioning housing along the transportation route.

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