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Lead Distribution and Mobility in a Soil Embankment Used as a Bullet Stop at a Shooting Range

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The distribution of lead in and below a soil embankment used as a stop butt for lead bullets at a sport shooting range for more than 30 years was investigated. A vertical profile, just behind the shooting target, was mapped by 54 soil samples characterized by contents of lead bullets, soil lead, and easily leachable lead as measured in a leaching test (L/S 2). At the target, the soil contained up to 40% metallic lead and 5 to 10% lead associated with the soil particles (<2 mm). The leaching test showed concentrations of dissolved lead in the range 5 to 20 mg/l. However, in the bottom of the stop butt (about 1 m lower than the target) soil lead was only slightly elevated, and no increase in lead was found below the stop butt in the original soil profile. In the lower part of the stop butt, pH was around 5, which is considered to favor lead migration, but in the soil samples with lead bullets present pH was between 6 and 7. The elevated pH values, probably caused by the corrosion of lead bullets, may have been a significant factor in limiting the migration of lead in the stop butt. The investigation showed that the lead in the stop butt did not affect the surroundings, but that the high lead content of the soil would require that this be treated as waste if the facility was abandoned.

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INTRODUCTION

LEAD is one of the most common anthropogenic soil contaminants of general concern and much has been done in recent years to limit the emissions of lead to the environment. Lead bullets and shots have been one of the significant emissions and lead bullets and shots are in many countries no longer allowed for hunting, but restricted in use to sport shooting ranges. At many shooting ranges, in particular at older facilities, the bullets are caught in stop butts, which are soil embankments constructed behind the targets. The bullets accumulate in the soil embankment, and usually the larger metallic chunks of agglomerated lead bullets are removed once in a while in order to avoid rebounding of the bullets. However, a substantial amount of metallic lead builds up in the soil (e.g., Tanskanen et al., 1991), and corrosion of the lead bullets may generate dissolved lead that could migrate out of the stop butt and into the surrounding environments (e.g., Jørgensen and Willems, 1987). Lead is usually strongly attenuated in soils (Alloway, 1995), but the combination of large concentrations of metallic lead and the long life time of the facilities (several decades) creates a concern for lead affecting adjacent groundwater and surface waters and for contaminating large quantities of soil that would require remediation if the facility was abandoned.

The purpose of this investigation was to map the distribution of lead bullets and soil lead in a stop butt at an old shooting range and to quantify the solubility of lead in order to evaluate the migration of lead. No information of this kind was found in the literature. By accomplishing the investigation at an old shooting range constituting a worst-case situation, field relevant information would be the basis for addressing the issue of lead migration in lead metal-contaminated soil.

MATERIALS AND METHODS

Shooting Range

Based on a preliminary survey of shooting ranges under the Danish Shooting Association, a site was selected in Northern Zealand about 50 km north of Copenhagen, Denmark. The site was 31 years old and, at least according to the records, had never been cleaned up nor had metallic chunks of agglomerated lead bullets removed from the stop butt. The shooting range (see Figure 1) is a 50 m range for small bore shooting and has six firing lines. The maximum shooting per line was estimated to about 230,000 shoots, which with an average of 2.6 g of lead per bullet compares to a maximum load per line of approximately 600 kg lead. The actual use of the range has been less than the maximum, but no records exist. A course estimate would be that 200 to 300 kg of lead bullets has been caught in the stop butt at the left line used in this investigation.

The shooting range was established on former farm land and the stop butt was built on top of the original ground with top soil excavated from the area hosting

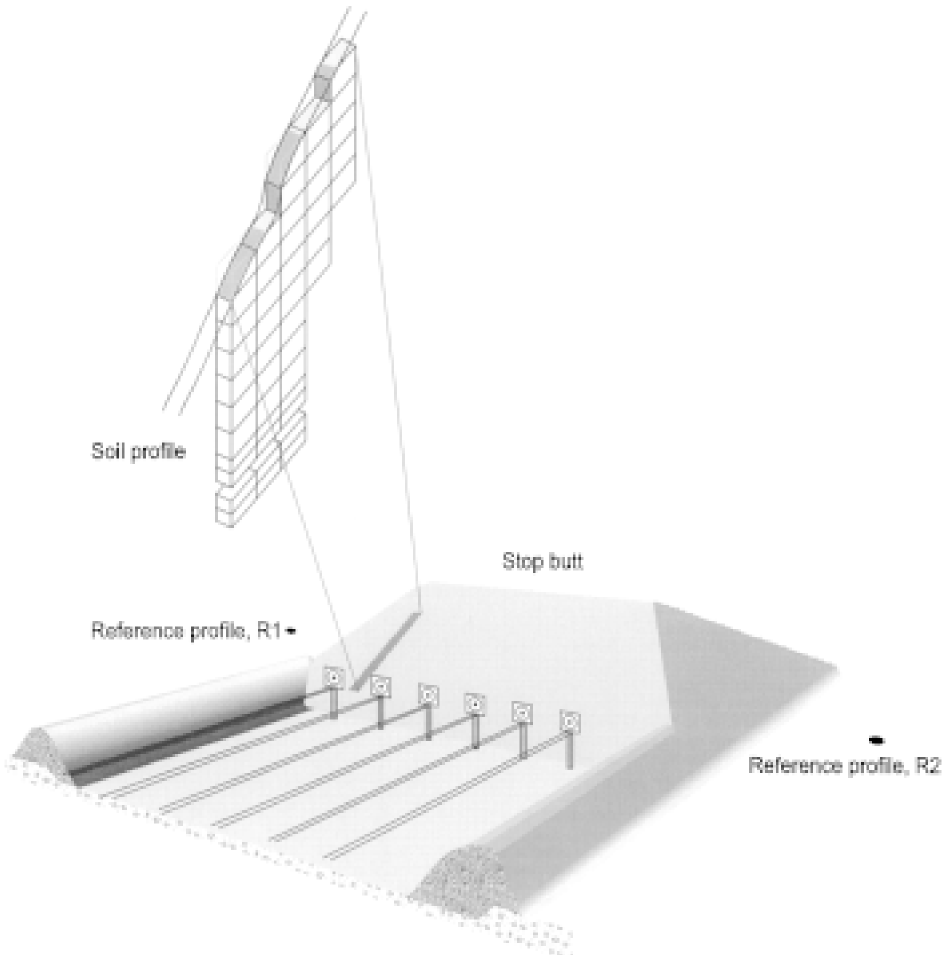

FIGURE 1

A sketch of the shooting range and the location of the soil profile investigated in the stop butt behind the left firing line. The location of the reference profiles (R1 and R2) also are shown.

the lines. The stop butt is at the bottom 12 m long and at the top 7 m long. The height is about 2.6 m and the bottom width is about 5 m. The vegetation is grass and shrubs.

Soil Sampling

A soil profile representing part of the cross section directly behind the target of the left line was dug out of the stop butt as indicated in Figure 1. The soil samples taken each represent a 0.2 m by 0.2 m by 0.2 m soil unit. The profile has a total depth of 2.5 m. Trenches were made on both sides of the soil profile in order to limit unintended mixing of the soil over the profile. The soil samples were collected in 10 l closed polyethylene containers. In total 54 soil samples were collected: 17 samples from the stop butt at and over the target, 17 samples from the stop butt below the target, 12 samples from the soil profile below the stop butt, and 8 reference samples (see later). The samples of the original soil profile below the stop butt and of the reference profiles were only 0.1 m deep and the lowest depth sampled was 0.4 to 0.5 m below ground surface. The top soil was about 0.4 m deep and the clayish till below was represented by the deepest soil samples.

Reference soil samples were sampled from the farm land about 3 m away from both ends of the stop butt (profile R1 and R2).

Soil Preparation

The soil was mixed in the sample container and a 2-kg subsample was dried at about 25°C for at least 36 h. A 2-mm polyethylene mesh screen was used for sieving the samples, removing stones, gravel, and the lead bullets. The lead bullets were washed, dried, and their weight recorded for each sample. The less than 2-mm fraction was used for the further characterization (except pH measurements) and experiments.

Soil Characterization

The dry matter content of the soil samples were determined by drying at 105°C for 24 h. Soil pH was determined by a combination electrode after mixing 15 g of wet soil with 15 ml of 10^{-2} *M* $CaCl_2$ for 30 min. The lead content in the soil (without lead bullets present) was measured after digestion of approximately 1g of soil with 20 ml of 6 *M* HNO_3 for 30 min in an autoclave at 200 kPa (120°C). The texture was determined on two soil samples, identifying the soil as a loamy sand: 8% clay (<0.002 mm), 16% silt (0.002 to 0.02 mm), and 76% sand (0.02 to 2.0 mm).

Leaching Experiments

The leaching of lead was investigated by means of a modified CEN test (CEN, 1996). About 30 g of dry soil was mixed with 60 ml 10^{-3} *M* $CaCl_2$ in a 100 polyethylene bottle for 72 h, corresponding to a L/S ratio of 2 at a pH value determined by the soil. After equilibration, pH was measured in the suspension and soil and solute were separated by centrifugation. The supernatant was preserved at pH 2 by addition of HNO_3 until analysis. The measured amount of dissolved lead was named easily leachable Pb content and expressed as mass of Pb per kg of soil.

Lead Analysis

Soil digestions and solutes from the leaching experiments were analyzed for lead by atomic absorption spectroscopy: soil digestions by flame atomization (Perkin-Elmer 5000) and solutes by graphite furnace atomization (Perkin-Elmer 5000, HGA 400 graphite furnace, AS-1 automatic sample injection, deuterium background correction) after liquid-liquid extraction (1% Na-diethyldithiocarbamate,trihydrate in *4*-methylpentan-*2*-one)).

Results and Discussion

The results are presented and discussed in terms of distributions of lead bullets, soil pH, soil lead content, leachable lead content, and calculated distribution ratios (soil Pb concentration divided by solute Pb concentration).

Distribution of Lead Shot Content

Figure 2 shows the distribution of lead bullets in the transect of the stop butt and in the reference profiles in terms of g Pb/kg dry soil (including real soil and lead shots). Most of the lead bullets were found close to the target, where up to 40% of the stop butt material consisted of lead bullets. Lead bullets were found as deep as 1 m into the soil embankment, but very few bullets were found more than 0.5 m above or below the center of the target. The 0.2-m-wide profile located just behind the target contained about 60 to 70 kg of lead bullets, and as bullets have missed the target also to the sides, it is estimated that about 100 kg of lead bullets were present in the soil embankment behind the target of the investigated firing line. This is less than half the amount of lead estimated from the number of shots presumably fired at the left line over a period of 31 years.

The discrepancy between the presumed number of shots fired at the firing line and the weight of lead bullets found in the stop butt may be due to unrecorded removal of chunks of agglomerated lead bullets sometime in the history of the shooting range. However, the amount of lead bullets present in the stop butt, as identified by the measurements, was substantial (of the order of 100 kg) and considered to be high for a stop butt at a single firing line.

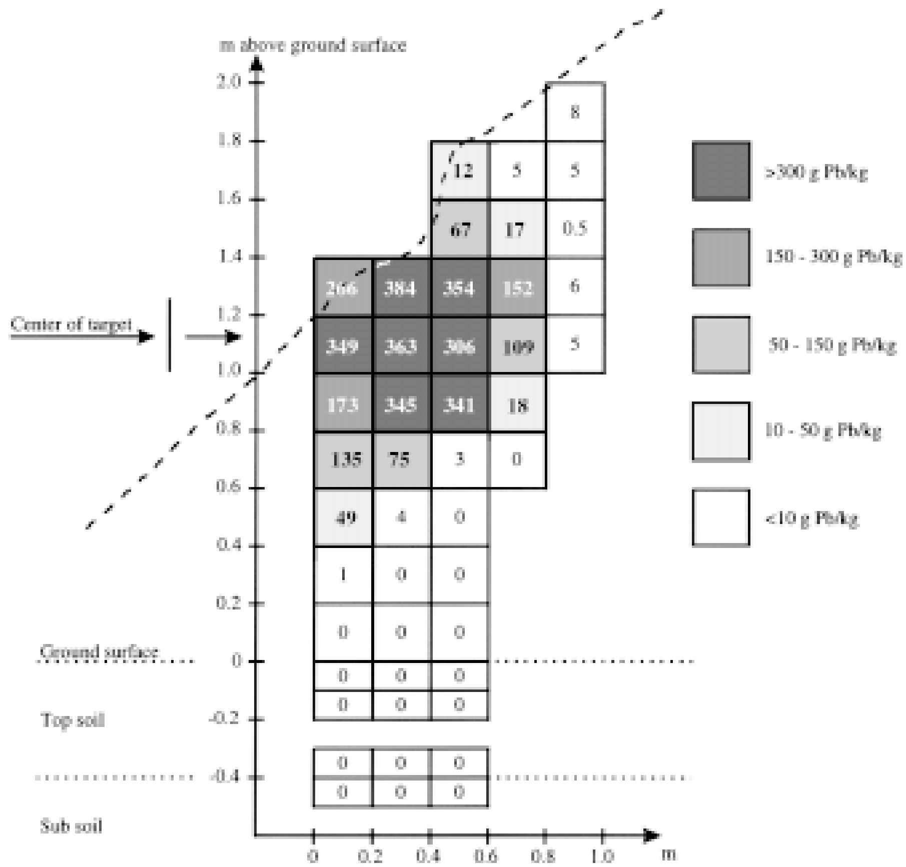

FIGURE 2

Contents of lead shots in the transect of the stop butt and in the reference soil profiles.

Distribution of Soil pH

Figure 3 shows the distribution of soil pH (measured in 10^{-2} *M* $CaCl_2$) in the transect of the stop butt and in the reference profiles. The internal parts of the stop butt seemed to be fairly low in pH, showing soil pH values around 5, while the original soil profile below the stop butt had pH values in the range 5.4 to 6.6. The upper parts of the reference profiles had slightly higher pH values than the original soil profile below the stop butt due to the agricultural use of limestone on the farm land. The soil-pH in the area with the high contents of lead bullets, however, was somewhat elevated compared with the internal parts of the stop butt: pH ranged in the soil samples containing lead bullets between 6.0 to 7.0, while soil samples from the internal parts of the stop butt ranged 4.7 to 5.4. The elevated soil pH values in

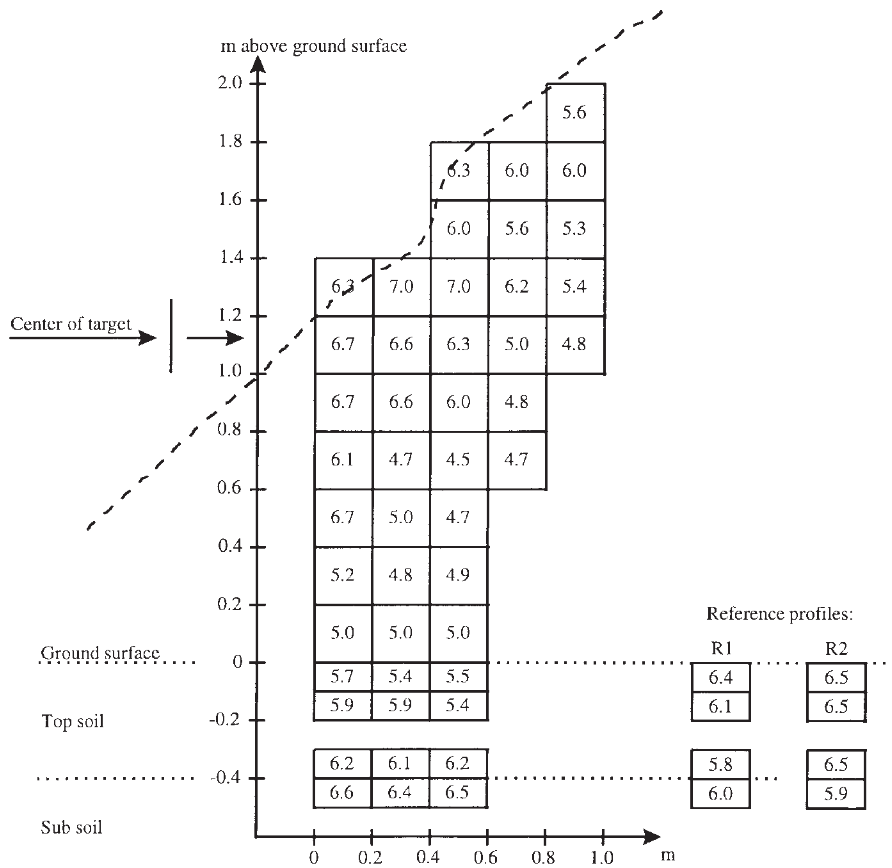

FIGURE 3

Soil pH in the transect of the stop butt and in the reference soil profiles.

the soil with high contents of lead bullets may be related to the corrosion of the lead bullets, as no amendments have been made to increase pH in this area.

Distribution of Soil Pb Content

Figure 4 shows the distribution of soil Pb content (determined after removal of lead bullets) in the transect of the stop butt and in the reference profiles. The Pb content was determined on an acid digestion of a dried soil sample after removal of lead bullets by sieving though a 2-mm screen, indicating that the term soil Pb content here includes Pb associated with the soil particles as well as fragments of metallic lead bullets smaller than 2 mm.

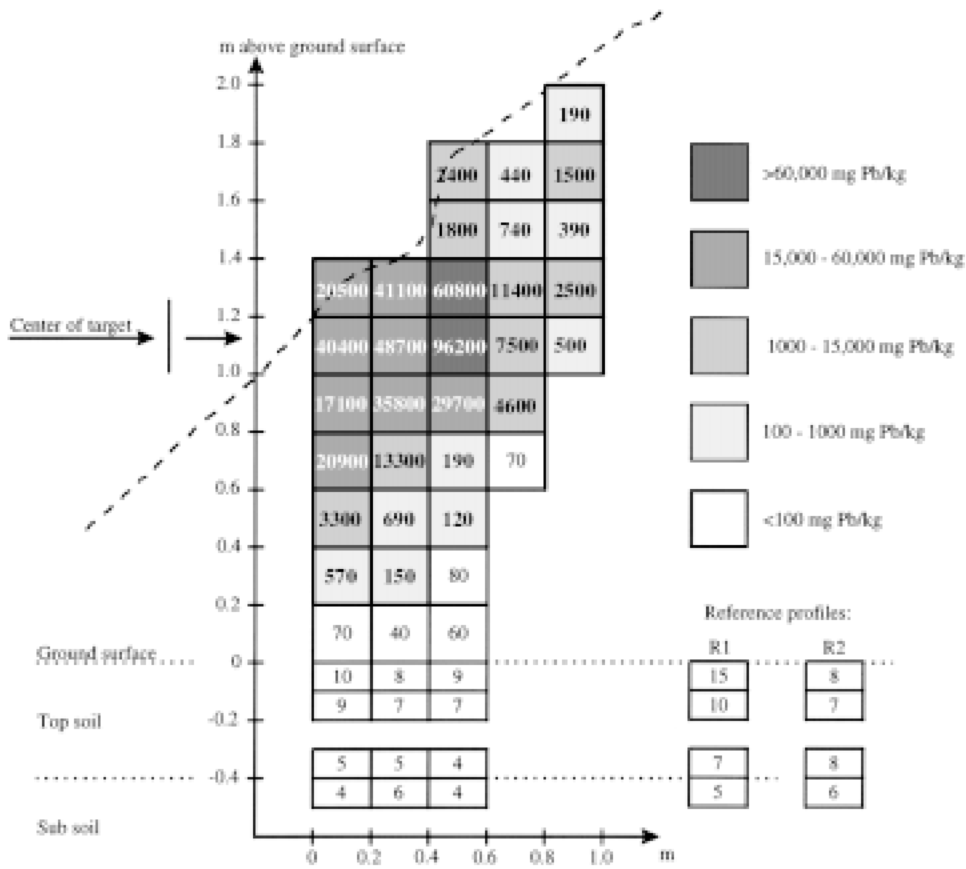

FIGURE 4

Contents of soil Pb (after removal of lead shots) in the transect of the stop butt and in the reference soil profiles.

In the target area the soil contained 5 to 10% lead not identifiable as lead bullets (removed by sieving). Substantially elevated Pb concentrations were present in all the soil samples from the stop butt. At the bottom of the soil embankment, the Pb concentrations ranged 40 to 80 mg Pb/kg soil, which is comparable to what can be found in many garden soils (Alloway, 1995). Pb concentrations in the soil profile below the stop butt ranged 4 to 10 mg Pb/kg, which is identical to the Pb concentrations found in the reference soil profile (5 to 15 mg Pb/kg) and below the average for Pb in Danish agricultural soils (Tjell and Hovmand, 1978).

The high Pb contents associated with the soil in the stop butt suggest that even after removing the lead bullets by sieving, the soil is strongly contaminated with Pb and would require special handling if the embankment should be removed.

The total amount of soil Pb was close to 10 kg and was too small (corresponding to about 10 to 15% of the amount of lead bullets found) to make up for the discrepancy, as discussed above, between presumed number of shoots fired at the line and the amount of bullets found in the stop butt.

Distribution of Leachable Pb

Figure 5 shows the distribution of leachable soil Pb determined as the amount of Pb per kg of soil that was dissolved during the leaching test. The concentrations of Pb found in the solution after equilibrating with the soil at L/S 2 corresponds to the leachable amounts divided by 2 l/kg.

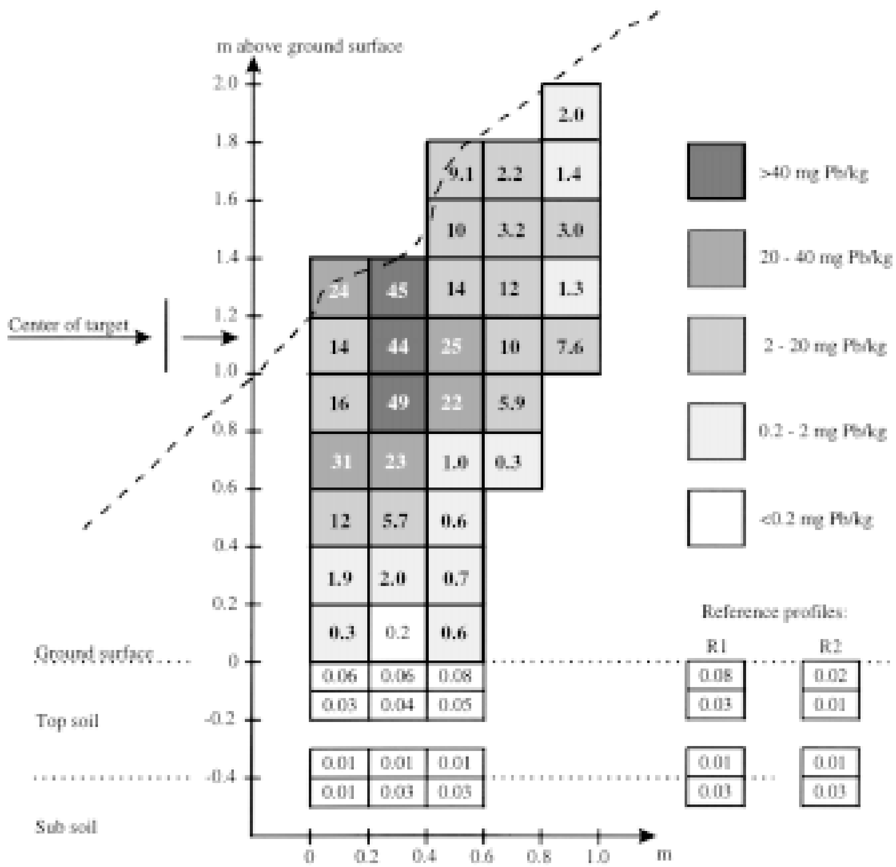

FIGURE 5

Contents of leachable soil Pb (at L/S = 2 after removal of lead shots) in the transect of the stop butt and in the reference soil profiles.

The highest amounts of easily leachable Pb was found just behind the target in the area with the highest contents of lead bullets: 20 to 50 mg Pb/kg corresponding to solute concentrations of 10 to 25 mg Pb/l. In the bottom of the stop butt the leachable Pb content was hundred times lower, but still about one order of magnitude higher than in the soil profile below the stop butt and in the reference profiles. In the reference profiles the leachable Pb content was 0.01 to 0.08 mg Pb/kg corresponding to solute concentrations of <5 to 40 μg/l The mapping of the easily leachable Pb showed that the presence of the stop butt did not change the content of easily leachable Pb in the original soil below the strongly contaminated section of the stop butt.

Distribution ratios (R_d) of Pb have been calculated for all soil samples by dividing the soil Pb concentration (excluding lead bullets) by the dissolved Pb concentration determined in the leaching experiment. A plot of the distribution ratio (as log R_d) vs. pH measured in the leaching experiments, see Figure 6, showed very little correlation (r^2), suggesting that the equilibrium between solid and solute was not governed by sorption processes (metal distribution governed by sorption usually shows a strong correlation with pH, see, for example, Anderson and Christensen, 1988). Small fragments of lead bullets (<2 mm) present in the soil may have contributed to the total content of Pb in the soil (metallic lead is dissolved by the digestion procedure used) but may not have had enough time to dissolve

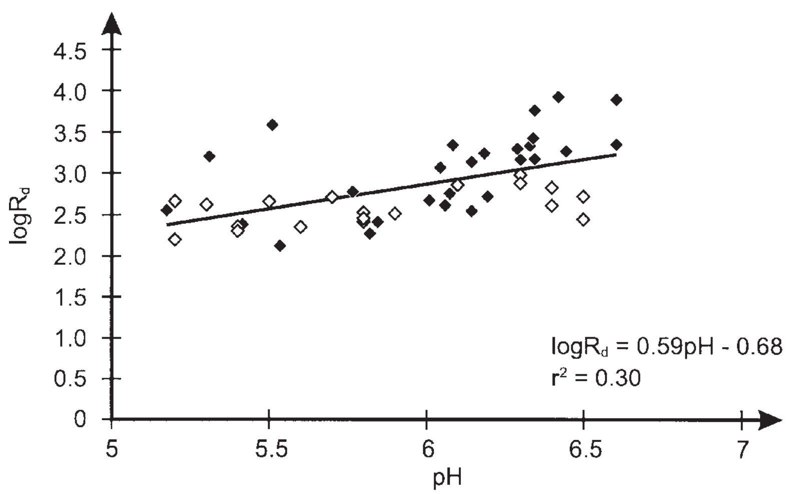

FIGURE 6

The Pb distribution ratios (Log R_d) for 54 soil samples as a function of the pH (measured in the experiments used to determine R_d). The hollow diamonds indicate soil samples with no identifiable lead bullets present.

significantly during the 72 h equilibration period. Precipitation processes may also have contributed to the distribution of Pb between the solid and solute phase. This excludes that the observed distribution ratios can be used directly as a mean to estimate the migration velocity of Pb in the soil embankment.

Mobility of Pb in the Stop Butt

The patterns of Pb in the stop butt showed that the dominant part of the Pb was found as lead bullets. The metallic lead bullets (>2 mm) supposedly constituted 85 to 90% of the lead in the stop butt. However, the fine fragments of lead bullets (<2 mm) and the Pb associated with soil particles still amounted to a very high concentration of Pb in the solid as well as the solute phase in the stop butt around the target. No lead bullets (>2 mm) were found in the bottom part of the stop butt (0 to 0.4 m above ground surface), but soil Pb as well as easily leachable Pb was significantly above the background levels as represented by the reference soil profiles and the profiles of the original soil below the stop butt. This could potentially be due to vertical migration of Pb from the higher and much more contaminated parts of the stop butt.

If migration of dissolved lead had increased the lead content of the bottom part of the stop butt, Pb should have migrated in average at least 0.6 m during approximately 30 years corresponding to 2 mm per year. The net infiltration of water in a soil embankment in this part of Denmark is of the order of 100 to 200 mm annually. Assuming that dissolved Pb in low concentrations is attenuated by sorption during migration down through the soil embankment, the annual migration velocity of Pb, V_{Pb} (mm/y) can be related to the annual net infiltration, N (mm water/y), dry bulk density of the soil, ρ (kg dry soil/l of porous media), and the sorption distribution coefficient for Pb, K_d (mg Pb per kg soil/mg Pb per l solute) by the equation:

$$V_{Pb} = N\,(\rho K_d)^{-1}$$

This equation suggests, assuming that ρ is approximately 1.6 kg/l, that K_d should not be any higher than 60 l/kg to allow Pb to migrate 2 mm annually with an infiltration of 100 mm annually. However, the distribution ratios (R_d) in the bottom part of the stop butt, which at low solute concentrations may approach real distribution coefficients, K_d as used in the above equation, show values in the range 200 to 500 l/kg, corresponding to a much lower migration velocity for Pb in the soil embankment. Migration of dissolved Pb has taken place in the soil embankment, but the above considerations suggest that also other factors may have contributed to the elevated concentrations of Pb in the bottom of the soil embankment: (1) penetration of grass roots and the activity of soil fauna may have contributed to the mixing in the soil embankment, (2) occasional repair of the soil embankment may have contributed to mixing as well, and (3) the distribution coefficients in the bottom part of the soil embankment having low pH values (around 5) are actually around a 200 to 300 l/kg, but dissolved organic carbon present in the leachate formed complexes with Pb enhancing its mobility. The significance of dissolved organic carbon complexes may have been underestimated in the leaching test, due to the strong dilution used in the experiment. The leaching experiment had a L/S ratio of 2, while in the unsaturated soil embankment, the L/S ratio is of the order of 0.1 to 0.2. The significance of these contributing factors cannot be further evaluated, but it is possible that all three factors have been involved.

Conclusion

The detailed mapping of the distribution of lead bullets, soil lead, and easily leachable lead in a more than 30-year-old earthen stop butt, just behind the target of a sport shooting line, showed that the stop butt was heavily polluted with lead, but that migration of lead out of the stop butt had not taken place. The stop butt investigated is considered to constitute a worst-case, because lead bullets had accumulated over very long time (supposedly 30 years) and the low soil pH values around 5 in the stop butt supposedly would increase lead mobility. The investigation shows the lead in the stop butt does not constitute a significant risk to the surrounding environment, but that a major part of the soil in the embankment is so polluted with lead that it should be handled as a waste, if removed from the facility or if the facility is abandoned.

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