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# Mercury Concentration and Distribution in Soils Impacted by Long-Term Applied Broiler Litter

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**Abstract** Mercury (Hg) is a non-essential element for plants and animals nutrition. Its presence in agricultural systems is of concern due to its high potential toxicity. Mercury is persistent in the environment and has been listed as a pollutant by several environmental organizations. This work focuses on Hg concentrations and distributions, trends, and relationships with some properties of soils that have received repeated broiler litter application and currently under pasture. Results revealed significant increase in Hg concentration from 41 to 105 μg $kg^{-1}$ and downward transport in the fields due to repeated applications. Correlation analysis indicated that sulfur and soil bulk density significantly (0.626*** and −0.645*** at $p \leq 0.05$, respectively) influenced its accumulation and distribution in this soil.

**Keywords** Correlation · Concentration · Factor analysis · Multiple regression

Mercury has been listed by several international organizations as a priority pollutant due to its persistence in the environment and toxicity to organisms and humans (Jiang et al. 2006; Grigal 2002). Studies by (Tessier-Lavigne et al. 1985) showed that exposure to Hg causes neurological and vision defects in humans. Mercury has also been implicated to cause Alzheimer disease (Mutter et al. 2004). Methyl mercury is a serious developmental neurotoxicant in fetus as a result of pregnant women consuming Hg contaminated seafood (Trasande et al. 2005). Mercury toxicity also causes plant growth retardation and reduces enzymatic activities (Pena et al. 2008). Jamal et al. (2006) reported a significant reduction in shoots and roots growth of wheat varieties treated with various Hg concentrations.

Mercury contamination in the environment is mostly due to anthropogenic activities, as chemical, paper, electrical, pharmaceutical, coal, waste, medical, and agriculture industries (Mirlean et al. 2008; Bash and Miller 2007; Hylander et al. 2006). Over 50% of agricultural profits in the United States are generated from poultry industries (Nyakatawa et al. 2001). The State of Alabama is ranked third in the United States in broiler production accounting for over 971 million birds (Alabama Agriculture Statistics Service 1999). This industry generates approximately 1.5 million Mg of broiler litter which is applied to pasture fields (Kpomblekou et al. 2002) as cheap source of fertilizers. Unlike various elements (As, Co, Cu, Fe, Mn, Se, and Zn) that are added to poultry diets to enhance weight gain and disease prevention, there is no evidence indicating that Hg is added to feeds. Mercury found in broiler litter may result from birds consuming grains containing trace amounts of Hg. Broiler litter applied to agricultural lands enhance soil productivity, but repeated land applications may also result to detrimental buildup and downward movement of Hg. Little information is available on the impact of repeated broiler litter application to pasture fields on mercury levels and distributions. Investigating Hg status of agricultural soils is vital for agricultural products safety. The objective of this study was to investigate the concentrations and distributions of Hg in soils amended with broiler litter and provide information to foster best management practices using broiler litter.

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## Materials and Methods

Soil samples were collected in triplicate from 0–20, 20–40, and 40–60 cm depth of the Hartselle series (fine-loamy, siliceous, subactive, thermic, and typic Hapludults) on a 3%–8% slope in the Sand Mountain region of north Alabama. The soil is being managed for pasture and had received broiler litter for 0, 5, and 10 years at rates of 0, 2.27, and 2.27 Mg $ha^{-1}$ $year^{-1}$ respectively. The samples were air-dried, ground to pass a 100-mm mesh, and stored at 4°C until used. Similarly, broiler litter were collected in Ziploc bags from 23 broiler houses across three counties (Dekalb, Franklin, and Cullman) in Alabama and stored at 4°C. The physiochemical properties of the soil have been described (Tazisong et al. 2005). Total Hg in soil and broiler litter was determined based on EPA method 7473 using direct mercury analyzer (DMA-80) with a detection limit of 0.005 ng Hg. Matrix spiked (30 ng Hg) samples were used after every 10 samples in the absence of reference standard material for quality assurance or control purpose. The mean concentration of Hg recovered in the spiked samples was 35.01 ± 3.55 ng which was within the limit of ±20% of the spiked value recommended by EPA method 7473.

Pearson linear correlation was used to evaluate the relationships between soil properties and Hg content. One way ANOVA was used to examine differences in Hg concentration among years. Principal component analysis (PCA) was applied to group correlated soil properties. The PCs generated were subjected to linear standard and stepwise multiple regression analysis to assess the relative or unique contribution of the PCs in predicting Hg concentration in the soil. All statistical analyses were performed with SAS Version 8.1 (SAS 1999).

## Results and Discussion

The background concentration of Hg in the 23 litter samples ranged from 0.001–0.041 ± 0.011 mg $kg^{-1}$ dry matter. The Hg concentration in these litters is by far lower than Pb, Cd, Ni, and Cr concentrations, also considered potential environmental toxicants (Table not shown). Mercury concentration in litter was in agreement with published data of other fertilizers (Shaffer 2001). Information on Hg concentration in broiler litter is limited thus we could not compare our results. Soil Hg concentration significantly increased after years of repeated broiler litter applications (Fig. 1). Similar findings were reported by Zheng et al. (2008) in three agricultural experiment sites with long term fertilization in China. Such build up of Hg is of great environmental concern because of possible food and health risks. It is noteworthy that Hg concentration in the control (0-year) soil is significant. This may be due to the parent materials or long-term atmospheric deposits from other regions. Atmospheric deposition flux at an estimated range of 0.74–2.97 μg $m^{-2}$ $year^{-1}$ did increase Hg levels in snowpits across the central Tibetan plateau (Loewen et al. 2007). Figure 2, shows Hg distribution along the soil profile. Mercury concentration tends to increase with depth irrespective of year of applied litter. The greatest downward movement is observed with 10 year of application. Such downward movement may have been influenced by such factors as parent materials, organic matter, iron and aluminum (hydr) oxides, clay minerals, and soil microbes (Barkay and Wagner-Dobler 2005; Roulet et al. 1998). The downward movement is also an indication of its mobility and should be of great concern because of potential ground water contaminations.

Correlation analysis (Table 1) showed highly positive significant correlation between Hg and sulfur. This relationship is noteworthy because of the affinity of Hg for

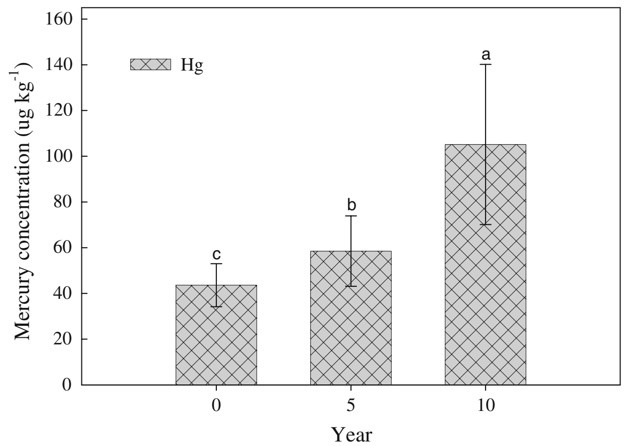


**Fig. 1** Mercury accumulation in soils with varying years of broiler litter application

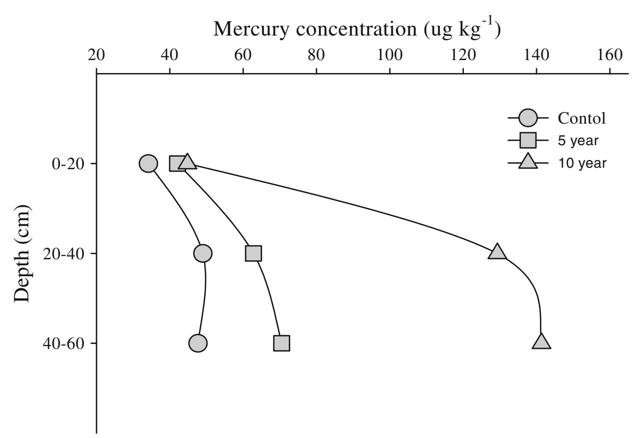


**Fig. 2** Depth distribution of mercury in soils with varying year of repeated broiler litter application

**Table 1** Pearson correlation matrix of soil properties and mercury

| | pH | Ec | C | N | S | Bd | $NH_4$ | $NO_3$ | P | Mg | Ca | Na | K |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Hg | −0.059ns | 0.104ns | −0.332* | −0.318* | 0.626*** | −0.645*** | −0.252ns | −0.373* | −0.412** | 0.314* | 0.115ns | 0.433** | 0.147ns |
| pH | | 0.262ns | 0.406** | 0.423** | −0.087ns | −0.080ns | 0.086ns | 0.235ns | 0.377** | 0.794*** | 0.750*** | 0.221ns | 0.610** |
| Ec | | | 0.123ns | 0.165ns | 0.006ns | −0.136ns | 0.161ns | 0.067ns | 0.235ns | 0.377** | 0.261ns | 0.108ns | 0.389** |
| C | | | | 0.985*** | 0.236ns | −0.217ns | 0.640*** | 0.774*** | 0.759*** | 0.490*** | 0.707** | 0.174ns | 0.173ns |
| N | | | | | 0.253ns | −0.229ns | 0.625*** | 0.791*** | 0.778*** | 0.520*** | 0.716** | 0.195ns | 0.215ns |
| S | | | | | | −0.742*** | 0.156ns | 0.150ns | 0.062ns | 0.353* | 0.429** | 0.516*** | −0.061ns |
| Bd | | | | | | | −0.143ns | −0.069ns | 0.063ns | −0.480*** | −0.459** | −0.560*** | −0.120ns |
| $NH_4$ | | | | | | | | 0.835*** | 0.554*** | 0.164ns | 0.370* | 0.466*** | 0.127ns |
| $NO_3$ | | | | | | | | | 0.774*** | 0.239ns | 0.495** | 0.373** | 0.168ns |
| P | | | | | | | | | | 0.374** | 0.606** | 0.146ns | 0.233ns |
| Mg | | | | | | | | | | | 0.860** | 0.346* | 0.595*** |
| Ca | | | | | | | | | | | | 0.436** | 0.403** |
| Na | | | | | | | | | | | | | 0.201ns |

*ns* not significant, *Bd* bulk density

*, **, *** significant at $p \le 0.05$, 0.01, 0.001, respectively

sulfur resulting in the formation of mercury ore [mercury sulfide (HgS)]. The association of mercury with sulfur is very important because it prevents the transformation of inorganic Hg to methyl Hg which is the most toxic form of mercury. Negative correlation existed between Hg and C, N, $NH_4^+$, $NO_3^-$, P, (Table 1) and Zn, Pb, Fe, Mn, Cu, and Al (Table not shown). These relationships may indicate its lack of affinity for these variables. There was no significant correlation between soil pH and Hg suggesting that Hg dynamics in this soil is not influenced by acidity. Principal component analysis retained five PCs (eigenvalues > 1), and accounted for >83% of the total variability of the measured data (Table 2). The first PC explained 40.66 of the variance, had very high positive loadings for Zn, $NO_3$–N, Cu, N, C, P, $NH_4$–N, Fe, and Mn. This shows a common physiochemical relationship. The second PC explained 19.19% of variance, had high positive loading on S, Hg, and Na and high negative loading on bulk density and Pb. The negative loading on bulk density is expected because decrease in bulk density increases soil porosity and thus less Hg accumulation. The high positive loading on S, Hg, and Na demonstrate the high affinity of Hg for S and Na.

**Table 2** Varimax rotated principal components (PCs) and total communality estimates (CE) of soil properties

| Parameter | PC1 | PC2 | PC3 | PC4 | PC5 | CE |
|---|---|---|---|---|---|---|
| Zn | 0.934 | 0.069 | 0.0060 | 0.028 | 0.167 | 0.871 |
| $NO_3$–N | 0.929 | 0.019 | 0.075 | −0.086 | −0.188 | 0.936 |
| Cu | 0.912 | 0.057 | 0.118 | 0.045 | 0.229 | 0.930 |
| N | 0.873 | 0.124 | 0.281 | 0.240 | 0.124 | 0.840 |
| C | 0.859 | 0.114 | 0.264 | 0.325 | 0.097 | 0.676 |
| P | 0.848 | −0.103 | 0.221 | 0.013 | 0.275 | 0.829 |
| $NH_4$–N | 0.814 | 0.098 | 0.0054 | −0.058 | −0.467 | 0.894 |
| Fe | 0.801 | −0.019 | 0.123 | 0.184 | 0.479 | 0.911 |
| Mn | 0.800 | −0.168 | 0.110 | 0.282 | −0.117 | 0.905 |
| S | 0.159 | 0.898 | −0.049 | 0.022 | 0.075 | 0.755 |
| Bulk density | −0.047 | −0.887 | −0.167 | −0.085 | 0.065 | 0.594 |
| Hg | −0.461 | 0.785 | 0.154 | −0.100 | −0.016 | 0.921 |
| Na | 0.281 | 0.639 | 0.188 | −0.284 | −0.350 | 0.774 |
| Pb | 0.103 | −0.546 | 0.224 | 0.480 | −0.408 | 0.903 |
| pH | 0.196 | −0.047 | 0.903 | −0.041 | 0.115 | 0.756 |
| Mg | 0.223 | 0.376 | 0.844 | 0.066 | 0.133 | 0.862 |
| K | 0.061 | −0.063 | 0.800 | −0.081 | −0.148 | 0.855 |
| Ca | 0.517 | 0.395 | 0.673 | 0.016 | 0.186 | 0.925 |
| C:N | 0.191 | 0.060 | −0.015 | 0.796 | −0.034 | 0.911 |
| Al | 0.156 | −0.322 | −0.491 | 0.622 | −0.028 | 0.725 |
| Cd | 0.371 | −0.054 | 0.132 | −0.148 | 0.643 | 0.676 |
| Eigenvalue | 8.54 | 4.03 | 2.19 | 1.48 | 1.21 | |
| Variance (%) | 40.66 | 19.19 | 10.44 | 7.06 | 5.75 | |
| Cumulative (%) | 40.66 | 59.85 | 70.29 | 77.35 | 83.10 | |

PC 3 shows a variance of about 10.44, where pH, Mg, K, and Ca had strong positive loading. The fourth and fifth PCs explained 7.06 and 5.75 of the variance with positive loading on C:N, Al, and Cd respectively. Standard linear regression performed with the five PCs generated, resulted to an $R^2$ value of 0.862 and the overall model highly significant at $p < 0.001$. Stepwise multiple regression analysis showed that PC2 was the most significant, with an $R^2$ value of 61.6%. When PC1 was added to the model, $R^2$ value increased by 21.2% resulting in a significant $F$ change ($p < 0.001$) and when PC3 was included, the $R^2$ value increased by 2.4% with a significant $F$ change of $p < 0.014$. The three PCs account for 85.2% of the total variation, and the resulting regression model equation expressed as $Y = 29.5_{PC2} - 17.3_{PC1} + 5.78_{PC3} + 61.97$.

Mercury levels in the 23 broiler litter samples were below the EPA maximum allowable total concentration (0.3 mg $kg^{-1}$) of most fertilizers, but repeated broiler litter applications over years to agricultural lands would influence to some extend the Hg concentrations in soils with lower background levels. Regression analysis showed S and Na as the most important soil variables for predicting Hg concentration in this soil. We recommend the continuous monitoring of total Hg build up in broiler litter applied soils in the United States. It is also imperative to address its speciation and bioavailability in soils for assessing food and health risks.

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