

# Emission and Leaching Potential of Mercury from Flue Gas Desulfurization (FGD) By-products Amended Soil

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## Abstract

The release of mercury (Hg) into atmosphere, infiltration, and uptake by plants from flue gas desulfurization (FGD) material-amended soils was studied. The project was carried out in three phases: (1) laboratory study, (2) field study, and (3) life cycle calculation. In the laboratory study, factors controlling the release of Hg from FGD by-product amended soil were investigated under well-controlled greenhouse environment. The field investigation was carried out based on results obtained from laboratory greenhouse studies. Life cycle analysis was carried out to calculate the release of Hg from the FGD material-treated soils under a conceptive model to evaluate the environmental impacts associated with the additions the three FGD materials used in this study.

## Introduction

The production of flue gas desulfurization (FGD) by-products (especially synthesized gypsum) is expected to increase in response to the tightening emission regulations for coal-fired power industry<sup>1</sup>. With the declining demand of FGD gypsum for wallboard production, alternatives to landfill are needed. Soil amendment is a potential mass application for FGD gypsum<sup>2</sup>. It has been reported that FGD material amendment, although increased the tissue concentration of trace elements (e.g., arsenic, selenium, and boron), did increase the plants grown without significant adverse effect on soil and leachate quality<sup>2,3,4</sup>. In these studies, the environmental impact associated with the FGD land application was evaluated by the concentration levels of trace elements (e.g., arsenic, selenium, boron, and molybdenum) in the soil, soil lechate, and plant tissues. However, the information on the release of mercury from the FGD material-amended soil has been lacking. It is expected that more mercury or other trace elements will present in the FGD by-products under the implementation of federal and/or state

mercury emission regulations (e.g., Illinois Mercury Rule). Despite the need to increase the utilization potential of FGD gypsums for agricultural land utilization, the release of mercury and other trace elements has not been well-characterized. The objectives of this study were (1) To measure the emissions of Hg from FGD by-product amended soil as well as the release of Hg and other trace elements into infiltration; (2) To investigate the uptake of Hg and other trace elements by plants; (3) To correlate the emission/release of Hg and other trace elements to the types of amendments and land application conditions; (4) To elucidate the mechanisms controlling the emission and release of Hg; (5) To determine the Hg exchange between air and amended soil; and (6) To conduct life cycle calculation for environmental sustainability evaluation.

## Methodology

The project was carried out in three phases: (1) laboratory study, (2) field study, and (3) life cycle calculation. In the laboratory study, factors controlling the release of Hg from FGD by-product amended soil were investigated under well-controlled greenhouse environment (Figure 1). The field study was carried out based on results obtained from laboratory greenhouse studies. The progress of field study can be seen in Figure 2. Life cycle analysis was carried out to calculate the release of Hg from the FGD material-treated soils under a conceptive model to evaluate the environmental impacts associated with the additions the three FGD materials used in this study.

Three FGD materials obtained from two coal combustion power plants were used. The SNO FGD material was obtained from the S-plant, which used wet, limestone, natural oxidation FGD process for sulfur dioxide (SO<sub>2</sub>) emission control. Two FGD materials, (i.e., AFO Gypsum and AFO-CPS) were collected from the A plant, whose FGD process was operated under forced oxidation mode. The AFO Gypsum was collected from the vacuum belt of the FGD system and AFO-CPS was the de-watered sludge solid collected from the FGD wastewater treatment plant.

The emission of mercury (Hg) was measured using a purge and trap sampling approach, which incorporates an air sampling pump and mercury sampling traps developed by ICSET to capture multiple mercury species (total Hg) in air. Water samples, i.e., infiltration and surface runoff, were collected and preserved for mercury and other trace elements analysis. Soil and plant samples were also collected to evaluate the change of mercury and trace element concentrations.

## Results and Discussion

### 1. Emission of Hg

Based on greenhouse studies, the Hg concentration in the soil, types of FGD material, and possibly moisture content of the soil were found to affect the emission of Hg from FGD material amended soil. Results from greenhouse studies showed that, for a given FGD material, the total mass of Hg emission increased as the amount of Hg in the soil increased. However, soils treated with different FGD materials showed different Hg

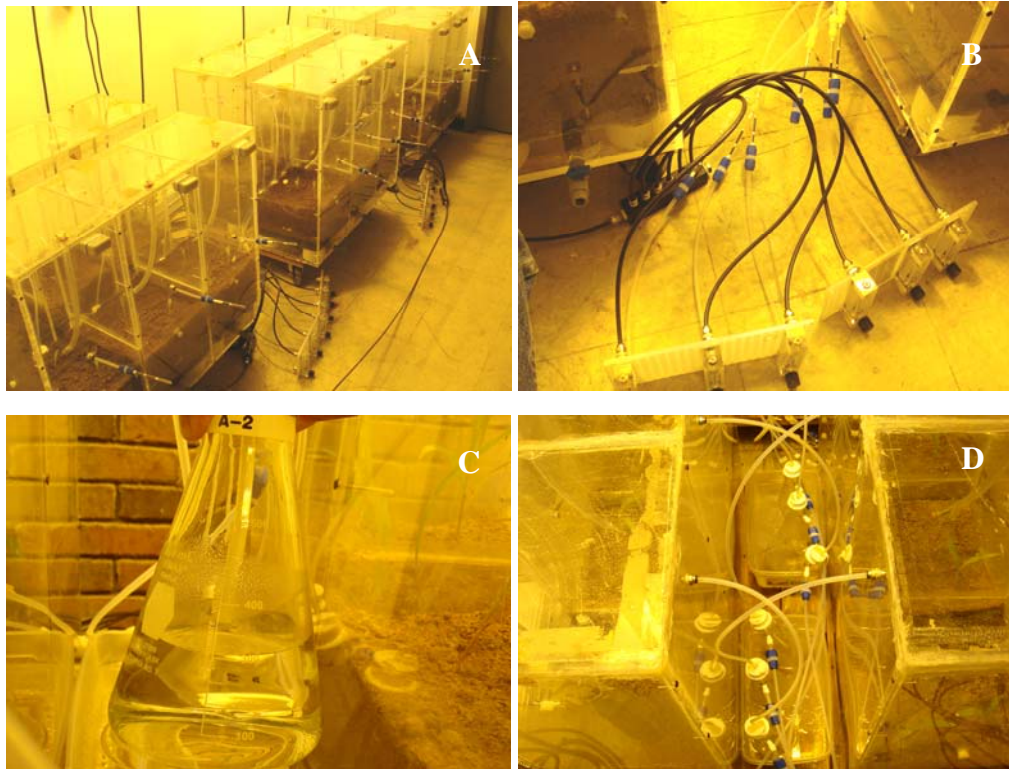


Figure 1. Laboratory greenhouse study

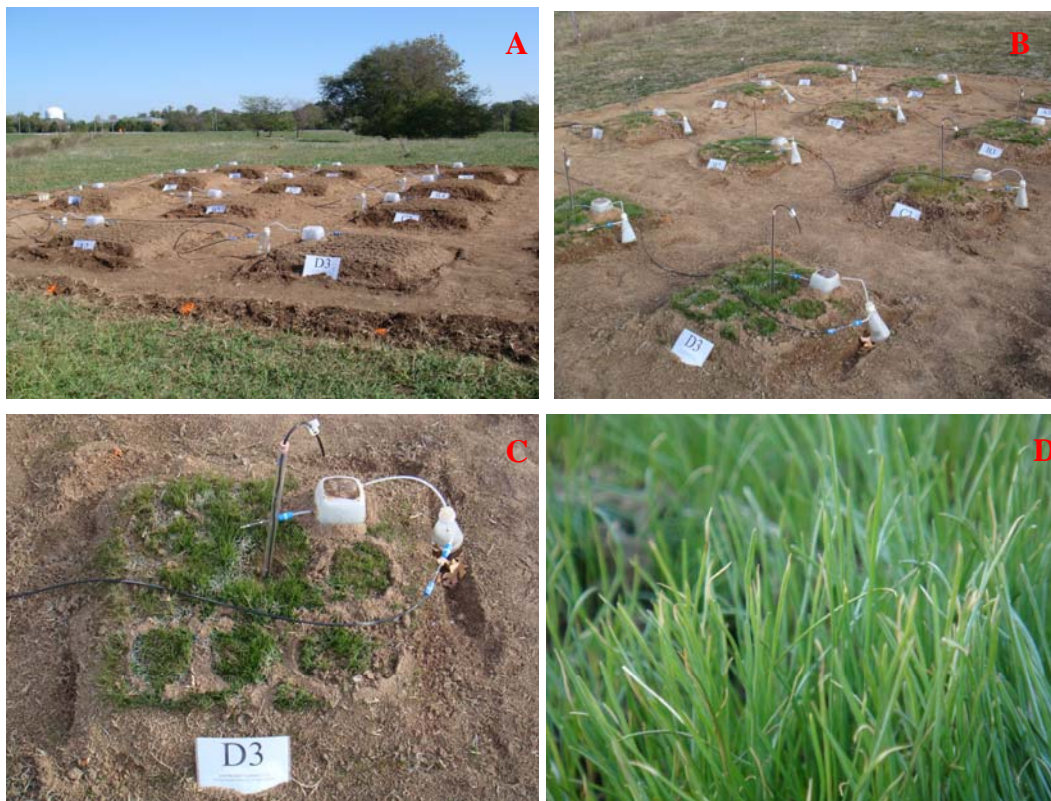


Figure 2. Progress of field study

emission behaviors (Figure 3). The SNO treatment had the highest Hg emission flux when compared to the other two FGD materials with the same amount of dosage. It was found that, with 1% SNO FGD material treatment, the emission of Hg increased about 340% when the moisture of the soil increased from 17 to about 28.5%. The moisture of the soil was calculated from the mass balance of irrigation water and infiltration.

The ratio of total Hg in the soil released into atmosphere was also calculated. Based on results from greenhouse studies, during the testing period, less than 1% of Hg was emitted into the atmosphere when the SNO FGD material was used with chicken litter as soil amendment. In the case of AFO-Gypsum and AFO-CPS, the mass of Hg emission was less than 0.8% and 0.02%, respectively, of total Hg in the soil.

Field study showed that nearly 6% of total Hg that was added into the soil as a result of SNO treatment was released into the gaseous phase. The ratio was found much higher than what was observed in the greenhouse studies. It was likely due to increase of contact between the carrier air and soil-FGD material mixture in the field study. In the greenhouse study, unlike the field study, less carrier air was able to contact with the deeper layer of the soil-FGD material matrix. The sampling chamber used in the field study was inserted approximate 2cm into the soil to make sure no ambient air slipped into the chamber from the contacting point between the edge of the chamber and soil. As a result, it was likely that some air was pulled through the soil matrix and had carried more Hg from the matrix. However, the effect did not apply to the other two FGD materials, about 0.3 and 0.01% of the total Hg was released into the atmosphere for the 1% AFO-CPS and AFO-Gypsum treatments.

The observation implied that the emission of Hg from the SNO FGD material maybe mass transport-controlled. In the case of the other two materials, chemical process might be more important. However, the mechanisms that controlled the release of Hg from the FGD material need further investigation.

## 2. Uptake of Hg by Plants

According to the results obtained from this study, the concentration of Hg in the soil and types of FGD materials are likely the most important factors that controlled the uptake of Hg by plant. With same amount of FGD material, the highest Hg concentration in the plant was observed in the SNO FGD material treatment (Figure 4). Although the addition of AFO-Gypsum and AFO-CPS resulted in higher Hg concentrations in the soil, the uptake of Hg by plant was not as fast as what was observed in the SNO treatment. Less than 0.01% of the total Hg in the soil was uptake by the plants in the SNO treatment. For AFO-CPS and AFO-Gypsum FGD materials, the uptake of Hg by the plants was less than 0.0002 and 0.005%, respectively.

Results obtained from field investigation showed that about 0.5% of the total Hg in the soil was uptake by the grass when SNO FGD material was used, which is about 50 times higher than what was observed in the greenhouse study where corn was used.

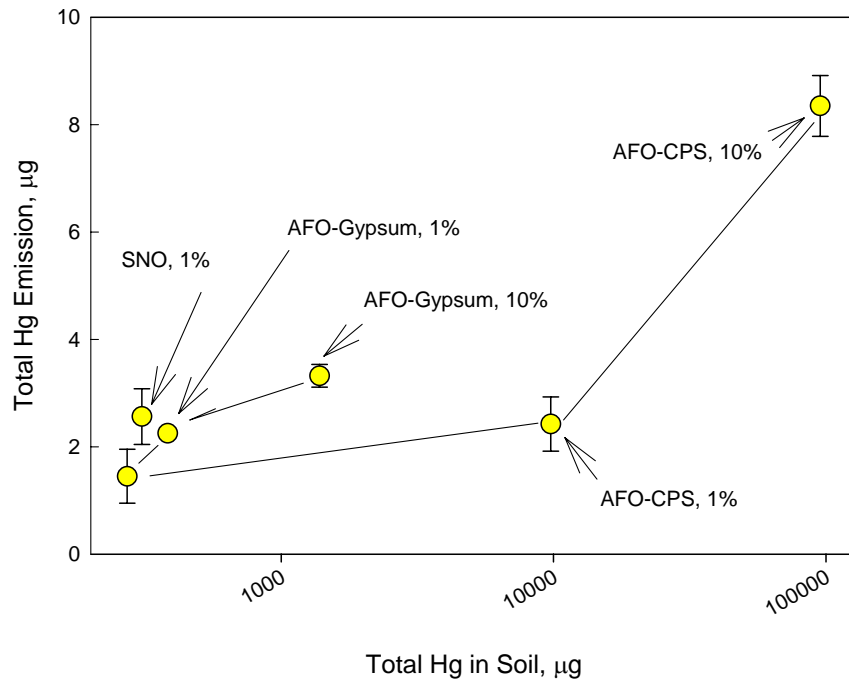


Figure 3. Correlation between Hg emission and total mass of Hg in soil treated with various dosage of three FGD materials and 1% of chicken litter

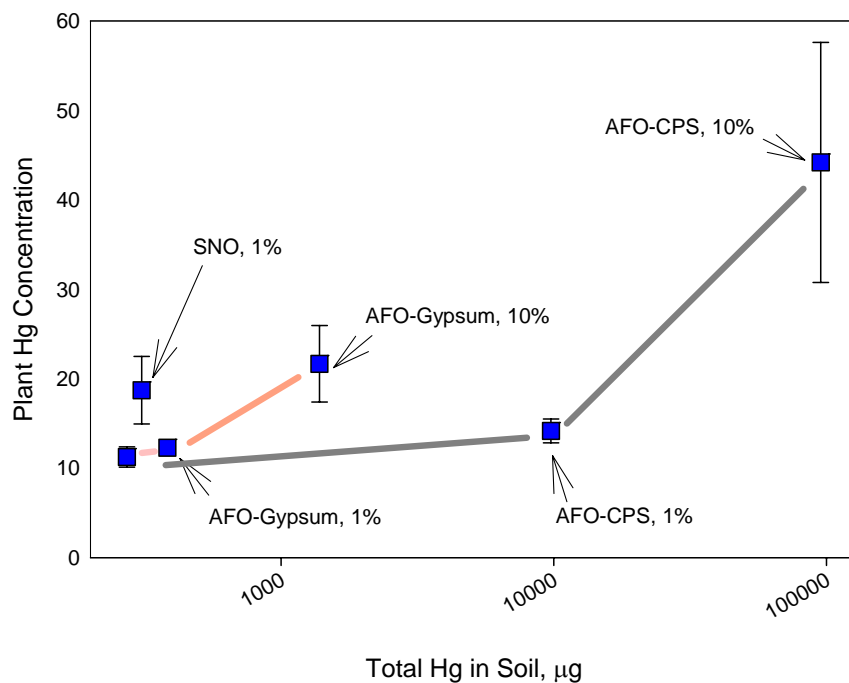


Figure 4. Correlation between plant Hg concentration and total mass of Hg in soil treated with various dosages of three FGD materials and 1% of chicken litter

It is also true for AFO-CPS and AFO-Gypsum treatment. The uptake ratio was about 0.09 and 0.2%, respectively, for AFO-CPS and AFO-gypsum, respectively. Although the experimental conditions were quite different between the field and greenhouse, results implied that different plants might have different abilities to uptake Hg from soil.

### 3. Release of Hg into Infiltration

No detectable Hg concentration level was found in all infiltration samples collected from the three batches of greenhouse studies. All the Hg concentration levels were less than the detection limit of 0.1 ppb. By taking the amount of infiltration collected during each experiment into account, less than 50ng of Hg was released into the infiltration.

The lysimeter system applied in the field study did not collect infiltration effectively. Only infiltration samples from A (blank) and one of the C (1% AFO-CPS) batches were available. Same as laboratory study, no detectable Hg level was observed in these samples.

### 4. Effect of FGD Material Additions on the Yield of Plants

According to the results from Batch Two greenhouse study, the addition of SNO FGD material showed a negative effect on the growth of plant. This conclusion was also valid from the results of Batch Three and field studies.

In the case of AFO-Gypsum, the total mass of corn did not show observable increased with 1% of AFO-Gypsum treatment in both Batch Three and field studies. But with 10% treatment, the total mass of the plants was significantly higher than the blank soil. Although not many data points demonstrated the trend, the addition of AFO-gypsum likely had positive effect on the growth of plants.

Same as the SNO FGD material, the AFO-CPS material showed negative effects on the growth of both corn and grass. It is likely due to higher uptake of Hg and other trace element from the AFO-CPS treated soil.

### Conclusion

The release of Hg using FGD material as soil amendments was also calculated based on the overall material balance of Hg in the two plants where the FGD materials were obtained from. It was assumed that 100% of the FGD materials produced from the processes were used as soil amendments. As shown in Table 1, the SNO FGD material released the most Hg during the testing period. The AFO-Gypsum treatment had the least release of Hg.

Table 1. Life Cycle Calculation of Release of Hg from Soil Amended Using FGD Material

Treatment	FGD Material to Soil Ratio <sup>1</sup>	Soil Hg <sup>2</sup>	Emission	Hg uptake	Total
		0-15 <sup>3</sup>			
		% (wt/wt)	kg of Hg/1000kg of Coal Burned		
		ng/g			
B-Field	SNO 1.0	39±8	2.26E-06	1.95E-07	2.46E-06
C-Field	AFO-CPS 1.0	4790±30	1.33E-07	1.08E-06	1.22E-06
D-Field	AFO-Gypsum 1.0	71±18	2.79E-07	1.58E-07	4.37E-07

<sup>1</sup> the ratio is in as received basis

<sup>2</sup> the results are from analysis after experiment

<sup>3</sup> section between surface and 15cm deep into ground

Results obtained from the calculation suggested that the AFO-Gypsum would release the lowest amount of Hg into the environments when all three FGD materials were compared under same operation assumption. However, all the experiments were carried out within a limited time frame. Long-term experiments are much needed to further investigate the effect of aging on the release of Hg into the environments.

#### Reference

- <sup>1</sup> Miller, C.E., eeley T.J., Aljoe, W.W., Lani, B.W., Schroeder K, Kairies, C, McNemar, A., Jones, A.P., and Murphy, J., Mercury capture and fate using wet FGD at coal-fired Power Plants, DOE/NETL Mercury and Wet FGD R&D, August 2006
- <sup>2</sup> Chen, L., Dick, W., Nelson, S., Flue Gas Desulfurization by-product additions to acid soil: Alfalfa productivity and environmental quality. Environmental Pollution. 114: 161-168. 2001
- <sup>3</sup> Clark, R.B., Ritchey, K.D., and Baligar V.C., Benefits and Constraints for use of FGD Products on Agricultural Land.” Fuel, 80, 80:821-828. 1999.
- <sup>4</sup> Punshon, T., Adriano, D.C., and Weber, J.T., Effect of Flue Gas Desulfurization Residue on Plant Establishment and Soil and Leachate Quality, Journal of Environmental Quality, 2001, 30, 1071-1080.