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Utilization of Water Utility Lime Sludge for Flue Gas Desulfurization in Coal-Fired Power Plants: Part II. Lime Sludge Characterization and Mercury Reemission

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Abstract

The feasibility of utilizing lime sludge in the flue gas desulfurization process of coal-fired power plants was evaluated through laboratory-scale studies. Eight lime sludge samples, collected from various water treatment plants, and a high-purity limestone sample were extensively characterized and tested for their ability to capture SO₂ from a simulated flue gas, while investigating the mercury reemission profiles during the scrubbing process. The reactivity of lime sludge samples for acid neutralization was significantly higher than the reactivity of the tested limestone sample. At doses less than that of the limestone sample, the lime sludge materials reduced the SO₂ concentration from 2,000 to <0.5 ppm. The residual lime, higher surface area, and more accessible pores in lime sludge samples were the major factors contributing to their higher reactivity. Concentrations of several elements including B, Mg, Mn, Fe, Cu, Zn, As, Sr, and Ba in some of the tested lime sludge samples were considerably higher than those elements in the limestone. However, no significant leaching of these elements into the scrubber solutions was observed. To investigate mercury reemission during the scrubbing process, ionic mercury was introduced into the simulated slurry and mercury reemission was monitored continuously. Results showed that compared with the limestone sample, the lime sludge samples tested had lower or similar cumulative mercury reemissions. However, different lime sludge samples showed different emission profiles. No conclusive correlation between the composition or trace element content of lime sludge samples and their mercury reemission could be identified. This result was likely due to the oxidative condition of the scrubbing process, which prohibited the reducing species from transforming the ionic mercury into elemental mercury.

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Notes

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Keywords

flue gas desulfurization; limestone; lime sludge; mercury reemission

1. INTRODUCTION

In Part I of this article, the main economic and environmental benefits of replacing limestone with lime-softening sludge in the flue gas desulfurization (FGD) process of coal-fired power plants were discussed.¹ This replacement may (1) eliminate the cost of high-quality limestone material and its transportation over long distances, (2) eliminate the energy-intensive grinding stage of limestone preparation, and (3) substantially reduce the overall CO₂ footprint and environmental impact.

Wet FGD systems are highly effective in removing SO₂ and other acidic gas species and can also remove oxidized mercury from the flue gas. Data collected from several coal-fired power plants equipped with wet FGD systems indicated that approximately 90% of the oxidized mercury in the flue gas is removed by FGD scrubbers.² However, FGD scrubbers cannot remove elemental mercury due to insolubility of Hg⁰ in water.

Some of the ionic mercury removed in FGD scrubbers can be reduced and released as elemental mercury. Mercury reemission in wet FGD scrubbers occurs when the elemental mercury concentration in the flue gas exiting the scrubber is higher than the inlet elemental mercury concentration. This phenomenon is a result of the reduction of Hg²⁺ to Hg⁰, mainly by sulfite ion in the scrubber solution:

A proposed mechanism suggests that elemental mercury reemission is controlled by the formation and decomposition of HgSO₃ and ClHgSO₃⁻ complexes.³ Various researchers (e.g., Refs. 3–5) have investigated the impact of flue gas and wet FGD slurry composition, oxidation condition, temperature, and pH on the emission of mercury. Sulfite–sulfate speciation is another important factor in controlling mercury reemission. Flue gases with a lower oxygen composition decrease sulfite oxidation to sulfate. A higher sulfite concentration increases the formation of mercury sulfite complexes, which are partially responsible for generating elemental mercury. Forced-air oxidation, to convert the sulfite ions to sulfate ions, is suggested as a practical option to maintain the mercury in the aqueous phase and minimize mercury reemission.⁶

Trace and major element contents of the FGD slurry are other factors that may affect the mercury reemission rate. Cations such as Ca²⁺ and Mg²⁺ may form sulfite compounds and, consequently, lower the formation of mercury sulfite species. Similarly, the addition of either fluoride or nitrate ions to the solution decreases mercury reemission, possibly because of the formation of stable compounds with Hg²⁺.⁴ Furthermore, other species, such as Fe²⁺, can be oxidized to their higher oxidation states (e.g., Fe²⁺ to Fe³⁺) in the scrubber solution and, consequently, reduce the ionic mercury to elemental mercury. Similar to Ca²⁺ and Mg²⁺,

Fe^{3+} reduces mercury emission because it sequesters sulfite anions by forming ferric sulfite.^{7,8}

A publication by Ochoa-Gonzalez and colleagues⁸ reports the impact of limestone characteristics on mercury reemission from simulated FGD slurries prepared from three Spanish limestone samples. Results of this work suggested that limestone samples with a smaller particle size, higher porosity, and lower magnesium content result in higher reactivity and enhance mercury capture in the scrubbers. Higher mercury reemission was observed when limestone samples with higher iron contents were tested using a simulated flue gas without oxygen.

As discussed in Part I of this article, based on the reported physicochemical properties of lime sludge and limestone samples and reactivity data, lime sludge could be a suitable replacement for limestone in FGD systems.¹ Mercury reemission from FGD scrubbers needs to be investigated further if power plants switch from limestone to lime sludge for the scrubbing process. We were unable to identify any research investigating the impact of lime sludge use on mercury reemission from FGD scrubbers. The main objective of this work was to evaluate the performance of various lime sludge samples for SO_2 removal from a simulated flue gas while monitoring the mercury reemission profiles during the scrubbing reaction. A high-purity limestone sample was also included in the experimental matrix as a baseline for further comparison and analysis.

2. EXPERIMENTAL SECTION

2.1. Lime Sludge and Limestone Samples

Lime sludge samples were collected from various water treatment plants in the states of Illinois and Ohio in the United States. Different factors were considered in the selection of representative water treatment plants that generate lime sludge with different characteristics. These included the water treatment plant location, water source (i.e., groundwater, surface water, or a mixture of both), and treatment process (with or without the addition of activated carbon powder [PAC] in the treatment process). Table 1 provides more information on the lime sludge samples collected. A high-purity limestone sample used in the FGD unit of a local power plant, identified hereafter as “limestone,” was also included in the experimental matrix as a baseline material for comparison.

Collected wet lime sludge samples were transported in closed 5-gallon buckets and then dried at 80 °C to measure their moisture content, which was determined gravimetrically from 44 to 86%. Dried aggregated lime sludge samples and the limestone sample were ground and sieved through a #325 U.S. standard sieve to a particle size of <44 μm (the typical size for FGD applications) for further characterization and testing.

Synthetic gypsum was prepared from limestone and lime sludge samples for use in scrubbing experiments. Ideally, gypsum should have a trace element composition like its limestone or lime sludge precursor. Therefore, synthetic gypsum samples were prepared by reacting the same limestone or dried lime sludge powder with ACS-grade sulfuric acid:

For gypsum preparation, 100 g of dried lime sludge (or limestone) sample was added to 3 L of deionized water, then 55 mL (~98 g) of sulfuric acid was added slowly while the slurry was stirred. After completion of the acid–base reaction at a final pH of 6, the slurry was rinsed with deionized water, filtered, dried at 60 °C, ground to a fine powder, and stored in airtight containers.

2.2. Sample Characterization

The surface area and pore size distribution of samples were obtained from the adsorption isotherms of nitrogen at a relative pressure (P/P_0) of 0.01–0.99 at 77 K by using a Micromeritics Gemini VII instrument. Samples were degassed under vacuum for 1 h at 200 °C prior to nitrogen adsorption. Surface area was calculated from the linear range of the Brunauer–Emmett–Teller (BET) equation with a relative pressure ranging from 0.03 to 0.3. Total pore volume was calculated from the adsorbed volume of nitrogen near the saturation point ($P/P_0 = 0.99$).

The average particle size and particle size distribution (PSD) of the samples were measured by using a Horiba LA-950 laser-scattering PSD analyzer. Mean particle size was calculated based on the PSD data obtained from three replicate measurements.

X-ray diffraction (XRD) was used to identify the degree of crystallinity and the proportion of different minerals present in the tested samples. The XRD analysis was performed by the XRD Laboratory at the Illinois State Geological Survey. In addition to XRD, X-ray fluorescence (XRF) of fused samples was utilized to quantify the major and trace oxides. Inductively coupled plasma-mass spectroscopy (ICP-MS) was also used to measure the trace element content of the samples. Activation Laboratory (Ancaster, Canada) performed ICP-MS (using the UT-1 Aqua Regia method) and XRF (using the Code 4C method) characterizations.

The pH of the lime sludge slurry was expected to be higher than the pH of the limestone slurry because of the presence of residual unreacted lime (CaO). To measure the pH of slurries, 1 g of each sample powder was placed into identical capped vials containing 65 mL of deionized water. The pH values of the samples were recorded after shaking samples for 24 h in ambient conditions.

The reactivity of dried lime sludge samples was evaluated and compared with the reactivity of the limestone sample. The reactivity of all samples was measured by the dissolution rate of the limestone or the lime sludge in an HCl solution following a method similar to ASTM C1318-95 for determination of the total neutralization capability of calcium and magnesium oxides. One gram of sample was added to 100 mL of deionized CO₂-free water. The slurry was then placed in a water bath at 50 °C, continuously stirred at ~200 rpm, and titrated at pH 6 with a standard 0.5 M HCl solution using a Mettler DL21 titrator for 60 min. Reactivity tests were conducted in triplicate.

2.3. Laboratory-Scale FGD Scrubber and Mercury Emission Monitoring

A laboratory setup that included a simulated flue gas preparation unit, a laboratory-scale wet scrubber, and a mercury vapor analyzer system was used to evaluate the performance of the limestone and eight lime sludge samples for SO₂ capture and mercury reemission from the simulated FGD slurries. All glassware and tubing was washed with 0.1 M HCl and deionized water, and then air-dried before beginning a new experiment. In a typical run, a simulated flue gas containing 5% O₂, 15% CO₂, and 2,000 ppm of SO₂, balanced with N₂ to give a total flow rate of 600 cc/min, was bubbled in a simulated wet FGD slurry containing 474 mL of deionized water, 25 g of synthetic gypsum, and 1.1 g of limestone or 0.9 g of dried lime sludge. Several tests indicated that the initial addition of 1.1 g of limestone or 0.9 g of lime sludge could maintain the pH at a range of 5.8–6.9 (which is close to the target pH value of 6) for the ~2 h scrubbing experiments.

The flask containing the slurry was placed in a water bath to maintain the slurry temperature at 50 ± 2 °C while stirring the slurry at ~300 rpm. Ionic mercury was added to the gypsum slurry at the beginning of each run by adding 0.1 mL of a 736 mg/L HgCl₂ solution (equal to 543 mg of Hg/L). This resulted in an initial mercury concentration of 114 µg/L (114 ppb), which is similar to the average concentration of mercury in power plant wet scrubbers (a range of 50–194 ppb for 22 surveyed power plants⁹). The emission of mercury was measured continuously during all experiments with a Tekran 2537X automated mercury analyzer. The simulated flue gas exiting the reaction flask was sent to a condenser to remove the condensed moisture, and then sent to the mercury analyzer. Solid and liquid samples were collected at the end of each experiment, and selected samples were analyzed by the UT-1 Aqua Regia ICP-MS method. Selected experiments were performed in triplicate to demonstrate the reproducibility of the results.

Gas-phase mercury concentration profiles were integrated to calculate the cumulative amount of mercury emitted during each 2 h run. The total amount of residual mercury in solid phase at the end of each experiment was measured based on the mercury analysis results of solid samples collected at the end of each experiment. The residual amount of mercury in the liquid phase at the end of each run was calculated by subtracting the cumulative amount of mercury emitted to the gas phase and the residual mercury in the solid phase from the total amount of mercury injected into the solution (i.e., 54.3 µg).

3. RESULTS AND DISCUSSION

3.1. Physicochemical Characteristics of Lime Sludge Samples

Both surface area and particle size are important parameters that affect the reactivity and dissolution rate of lime sludge particles in acidic solutions. The mean particle size of the majority of lime sludge samples varied from 16 to 34 µm, whereas the average particle size of the limestone sample was 31 µm (Table 2). The BET surface areas of the lime sludge samples varied from 3 to 12 m²/g, whereas the limestone sample had a significantly lower surface area of 0.4 m²/g. The BET surface area represents the total internal (surface area between grains) and external (geometric) surface areas. However, the internal porosity of

micrometer-sized samples had a dominant impact on the total surface area, compared with the external geometric surface area.

Results showed that the pH of lime sludge samples (in a range of 10.1–10.3) was significantly higher than the pH of the tested high-purity limestone sample (i.e., 9.40) (Table 2). This was likely due to the presence of residual unreacted lime resulting from using an excess amount of lime in the water-softening process. The residual lime likely enhanced the acid neutralization reactivity of the lime sludge samples. Samples SL4, SL5, SL6, and SL8 had higher pH values (i.e., 10.2–10.3) compared with other samples; therefore, their residual lime content was also expected to be higher.

The XRD results suggest that samples SL1, SL2, SL4, SL7, and SL8 had higher quartz or clay impurities and sample SL6 had the highest calcite content (Table 3). Detection limits (e.g., 1% for XRD) should be considered in comparisons and analyses of the data. Note that the XRF results presented are based on an XRF-fusion analysis in which minerals (e.g., CaCO_3 and MgCO_3) are converted to their oxide forms (e.g., CaO and MgO) prior to the XRF analysis. The XRF, which has a higher detection limit and thus greater accuracy than XRD, indicated >50% CaO purity for limestone and for samples SL2, SL3, and SL6, whereas sample SL7 had the lowest CaO content. The other samples had lower CaO contents and higher impurities (compared with the tested limestone and lime sludge samples SL2, SL3, and SL6), as indicated by the higher SiO_2 , Al_2O_3 , and Fe_2O_3 values (Table 3). The selected limestone is a high-purity limestone with a high calcite content and was intentionally included in the experimental matrix as the baseline limestone used in wet FGD scrubbers. The Ca and Mg composition results obtained by ICP-MS are consistent with the XRF data.

Concentrations of several elements in all or some of the tested lime sludge samples were considerably higher than those elements in the limestone (Table 4). These elements included B, Mg, Mn, Fe, Cu, Zn, As, Sr, and Ba. If some of these elements (i.e., B, Sr, and Ba, which were present in high concentrations) were to leach from the lime sludge into the scrubber solution during the scrubbing process, they could create additional wastewater treatment problems. We have shown, based on further analysis of the scrubber solutions (presented in the next section), that concentrations of the elements of concern are similar in scrubber solutions collected from lime sludge experiments or the baseline limestone test.

Manganese, Fe, and Cu, which were found in higher concentrations in lime sludge samples compared with limestone, could potentially affect the catalytic performance of the lime sludge samples. The concentrations of other tested elements, including Hg and Se, were similar in the lime sludge and limestone samples.

The Fe and Al contents of sample SL7 were up to several times higher than those in the other lime sludge samples (Table 4). This may have been due to the impact of residual coagulants or higher concentrations of Fe and Al in the water source. Concentrations of other trace elements in different samples varied depending on the sample source (Table 4).

The hydrochloric acid uptake by the lime sludge and limestone slurries versus time at the typical conditions of FGD scrubbers (i.e., pH 6 and temperature of 50 °C) was used as an

indicator of sample reactivity. The reactivity of samples to acid neutralization can be evaluated by the total amount of HCl that reacts with the sample at any given time and can be manifested by the initial rates of reaction (e.g., at $t < 1$ min), the initial slopes of HCl uptake plots, or the total HCl uptake at time t (Figure 1). All the tested lime sludge samples had a higher overall reactivity, including higher initial slopes and higher HCl uptakes, compared with the baseline limestone sample. The higher reactivity of lime sludge samples may be attributed to their higher surface area, the presence of unreacted lime (i.e., the amount of residual unreacted lime in the lime sludge), or the difference in grain structure that might facilitate the access to and reaction of HCl with the solid surface. The majority of the tested lime sludge samples (i.e., SL1, SL3, SL6, SL7, SL8) had a similar reactivity, whereas two samples (SL4 and SL5) had a significantly higher reactivity, and one sample (SL2) had a significantly lower reactivity (Figure 1). The observed differences in the reactivity of all lime sludge samples cannot be clearly explained based on only their surface area (Table 2) or chemical characteristics (Table 3). The lower reactivity of sample SL2 might be explained by its relatively large particle size (i.e., 34 μm), which is similar to the particle size of limestone but considerably higher than the mean particle size of other lime sludge samples (Table 2). Particles with larger sizes are expected to have lower reactivity, compared with smaller particles with the same properties, mainly because of the lower reactive surface area per mass of particles. The higher reactivity of samples SL4 and SL5 was likely due to the impact of a combination of several parameters, including the particle size, surface area, residual lime, and other characteristics, such as grain structure, which was not evaluated in this work.

3.2. Mercury Reemission and Fate During SO₂ Capture

In all scrubber experiments with either the limestone or lime sludge slurry, the SO₂ concentration was reduced from the inlet value of 2,000 ppm to <0.5 ppm, which is the detection limit of our SO₂ analyzer. A smaller amount of lime sludge addition (i.e., 0.9 g of lime sludge compared with 1.1 g of limestone for each run) was sufficient to capture SO₂ from the simulated flue gas and reduce the SO₂ concentration to <0.5 ppm, confirming the higher reactivity of lime sludge samples for SO₂ capture. The addition of lime sludge or limestone samples at the beginning of each run controlled the pH of the slurries in the range of 5.8–6.9 during the experiments.

A general trend observed was that the Hg concentration in the gas stream initially increased sharply after addition of the HgCl₂ solution at the beginning of the experiments, and then rapidly decreased, followed by longer less-variable phase of Hg emission (Figure 2). However, different lime sludge samples showed different emission levels and profiles. Generally, the amounts of mercury emitted peaked within 17 min after the addition of HgCl₂, and the peak values ranged from approximately 77.2 $\mu\text{g}/\text{m}^3$ (for sample SL4) to 200 $\mu\text{g}/\text{m}^3$ (when limestone was used) (Figure 2). The initial mercury peak for all samples, except SL1 and SL2, appeared 5 min to 10 min after the addition of HgCl₂. SL1 and SL2 samples had a delayed mercury peak at ~17 min. Furthermore, SL2 showed a tail with higher mercury concentration than other samples. Observed variation in mercury release profiles of different samples remains to be further investigated.

The cumulative amount of mercury emitted during the 2 h tests for lime sludge samples ranged from 1.42 μg for sample SL4 to 3.16 μg for sample SL2, whereas the cumulative mercury emission for the limestone sample was 2.74 μg (see Table 5). The calculated relative standard error percentage for these measurements ranged from 0.1% for SL1 to 9% for SL8. These results illustrate that the cumulative mercury emission of the majority of tested lime sludge samples was similar to that of the baseline limestone. For samples SL1, SL2, SL5, SL7, and SL8, the cumulative mercury emission varied from 2.36 to 3.16 μg , whereas the cumulative mercury emission for the tested baseline limestone sample was 2.74 μg . Furthermore, the cumulative mercury emission for three lime sludge samples (SL3, SL4, SL6) varied from 1.42 to 1.87 μg , which was considerably lower than the value observed for the tested baseline limestone sample. These results clearly illustrate that compared with the tested high-purity limestone sample; the lime sludge samples had (1) superior performance for SO_2 capture, mainly because of their higher reactivity, and (2) lower or similar mercury reemissions. Future studies may further confirm these results by testing other limestone and lime sludge samples with different physicochemical characteristics.

Results of the gas–solid–liquid distribution of mercury showed that the amount of mercury emitted to the gas phase during each 2 h test was less than 6% of the total mercury initially added to the scrubber (Table 5). Except for samples SL1 and SL7, the majority of the injected mercury (i.e., 57–90%) stayed in the liquid phase. For samples SL1 and SL7, more than 80% of the mercury was adsorbed onto the solid phase. This was likely due to the existence of residual PAC in sample SL1 or possibly higher concentrations of natural organic matter in sample SL7, which adsorbed mercury or formed more stable mercury complexes.

Ochoa-Gonzalez and colleagues⁸ reported higher mercury reemission when limestone samples with higher Fe contents were tested using a simulated flue gas without oxygen. Various trace elements or impurities in the limestone or the lime sludge slurries may cause different catalytic or oxidation–reduction reaction pathways for mercury transformation, resulting in different mercury reemission profiles. Species such as Fe^{2+} can be oxidized to their higher oxidation states (e.g., Fe^{2+} to Fe^{3+}) in the scrubber solution and, consequently, reduce ionic mercury to elemental mercury under in the absence of oxygen. Similar to the presence of Ca^{2+} and Mg^{2+} , the presence of Fe^{3+} reduces mercury emission^{7,8} because sulfite anions are sequestered by the formation of ferric sulfite.

In this work, no clear correlation between the composition or trace element content of lime sludge samples and their mercury reemission was observed. We observed variation in the Fe and Mn contents and other chemical characteristics of the tested lime sludge samples, but the oxidative condition of the scrubbing process may have limited the impact of the reducing effect of some species, such as Fe^{2+} , as reported by Ochoa-Gonzalez and colleagues⁸ for reducing the ionic mercury to elemental mercury in the absence of oxygen in the flue gas.

According to the characterization data, concentrations of several elements (i.e., B, Mg, Mn, Fe, Cu, Zn, As, Sr, and Ba) in some of the tested lime sludge samples were considerably higher than those elements in the limestone (Table 4). To further investigate the possibility of these elements leaching into the scrubber solution and to evaluate their potential impact on mercury reemission, liquid samples were collected at the end of scrubbing experiments

performed with selected lime sludge samples and the limestone sample. The analytical results indicated that the majority of tested elements in the solutions collected from the lime sludge experiments had concentrations similar to those of the baseline limestone experiment (Table 6). For example, B concentration in the SL1, SL4, and SL8 lime sludge samples ranged from 62–142 mg/kg, compared with <1 mg/kg B content of the limestone sample (Table 4). However, concentrations of B in all liquid samples from the scrubbing experiments with lime sludge samples or the limestone sample varied from 2.1 to 2.6 mg/L. Similarly, Ba, As, Zn, Cu, and Fe concentrations of scrubber solutions from tested lime sludge and limestone experiments were at low and similar levels (Table 6). Strontium content of the SL1, SL4, and SL8 lime sludge samples was 548–1,110 mg/kg versus 89 mg/kg for the limestone sample. However, the measured Sr concentrations of scrubber solutions were only 1.2–4.0 mg/L for the lime sludge experiments compared with 0.4 mg/L for the limestone test. Concentrations of Mn and Fe in the scrubber solutions of the lime sludge experiments were also similar to or lower than those in the limestone experiment.

4. CONCLUSIONS

The performance of various lime sludge samples and a high-purity limestone sample for SO₂ removal from a simulated flue gas was evaluated, and mercury reemission was monitored continuously. Compared with the baseline limestone, all lime sludge samples exhibited superior reactivity to both HCl neutralization in reactivity tests and SO₂ capture from a simulated flue gas. The cumulative amount of mercury emitted during the 2 h tests for lime sludge samples ranged from 1.42 to 3.16 µg, whereas the cumulative mercury emission for the limestone sample was 2.74 µg. This corresponds to the reemission of 2.6–5.6% of total ionic mercury for lime sludge samples, compared with 5.0% reemission for the limestone sample. These results illustrate that the cumulative mercury emission of the tested lime sludge samples was similar to or lower than that of the baseline limestone sample. Future studies may further confirm the findings of this work by testing other limestone and lime sludge samples with different physicochemical characteristics.

Concentrations of B, Mg, Mn, Fe, Cu, Zn, As, Sr, and Ba in some of the lime sludge samples were considerably higher than those elements in the limestone. Leaching of these elements into the scrubber solution during the scrubbing process could create additional wastewater treatment problems. However, results indicated that concentrations of these elements in the solutions collected from the lime sludge experiments were similar to those of the baseline limestone experiment.

Overall, lime sludge can be used as a potential replacement to limestone for the removal of acid gases from power plant flue gas. Based on the results presented in Part I of this article, the reuse of lime sludge generated at water utilities to capture acid gases at power utilities can be a sustainable and cost-effective solution benefiting both the water and energy sectors.¹ The laboratory-scale results presented in this work confirmed the high reactivity of various tested lime sludge samples without negatively affecting mercury reemission or undesirable elements leaching into the scrubber slurry. More testing at a pilot scale is needed to further evaluate possible operational issues before consideration for potential large-scale implementation.

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Figure 1. Reactivity of limestone and lime sludge samples when reacting with a 0.5 M HCl solution at pH 6 and a temperature of 50 °C. Error bars show the reproducibility of results based on triplicate experiments.

Figure 2. Mercury reemission profiles from a laboratory scrubber using limestone or lime sludge samples with their synthetic gypsum pair at 50 °C in the pH range of 5.8–6.9. Error bars show the reproducibility of results based on triplicate experiments of samples SL1 and SL8.

Figure 3.

Cumulative amount of mercury emitted during 2 h tests based on the mercury reemission profiles shown in Figure 2. Error bars show the reproducibility of results based on triplicate experiments for samples SL1, SL7, SL8, and SL6.

Table 1

List of Collected Lime Sludge Samples and the Relevant Feed Water or Water Treatment Information.

sample name	location	feed water source	water source hardness, mg/L (CaCO ₃)	powdered activated carbon treatment
LS1	Illinois	Both ground water and surface water (river)	250–374	Yes
LS2	Illinois	Both ground water and surface water (river)	250–374	Yes
LS3	Ohio	Groundwater	260–312	No
LS4	Illinois	Groundwater	150–500	No
LS5	Ohio	Groundwater	120–140	No
LS6	Ohio	Groundwater	350–700	No
LS7	Illinois	Surface water (river)	150–373	No
LS8	Illinois	Groundwater	420–450	No

Table 2

Physical Characteristics of Samples.

	mean particle size (μm)	BET surface area (m^2/g)	total pore volume (cm^3/g)	slurry pH
SL1	21	9.29	0.052	10.1
SL2	34	5.48	0.031	10.1
SL3	16	3.08	0.018	10.2
SL4	19	8.78	0.06	10.2
SL5	19	8.17	0.052	10.3
SL6	16	4.20	0.027	10.3
SL7	29	8.30	0.054	10.1
SL8	18	12.27	0.091	10.3
Limestone	31	0.40	0.004	9.40

Table 3

Mineralogical Characteristics of Samples Determined by X-Ray Diffraction (XRD) and X-Ray Fluorescence of Fused Samples (FUS-XRF). All Units Are in wt %.

unit	detection limit	limestone	SL1	SL2	SL3	SL4	SL5	SL6	SL7	SL8
Na wt %	0.001	0.021	0.065	0.082	0.045	0.065	0.050	0.042	0.036	0.067
Mg wt %	0.01	0.55	3.34	1.87	1.07	2.99	2.79	1.49	3.61	3.45
Al wt %	0.01	0.02	0.07	0.03	0.09	0.04	0.05	0.05	0.62	0.09
K wt %	0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.13	<0.01
Ca wt %	0.01	33.6	31.3	34.5	34.8	35.1	32.6	31.6	26.0	30.6
Fe wt %	0.01	0.05	0.53	0.40	0.05	0.34	0.03	0.06	1.34	0.28
P wt %	0.001	0.006	0.030	0.012	0.010	0.024	0.009	<0.001	0.046	0.016
B mg/kg	1	<1	62	17	5	142	35	9	27	76
Mn mg/kg	1	235	139	59	568	54	623	364	293	150
Cu mg/kg	0.01	0.42	5.74	4.25	1.93	1.81	4.28	2.20	11.80	1.72
Zn mg/kg	0.1	8.7	13.7	6.7	8.1	9.3	1.3	0.6	34.4	9.0
As mg/kg	0.1	<0.1	1.2	<0.1	<0.1	3.8	0.6	<0.1	2.8	5.9
Rb mg/kg	0.1	0.5	0.7	0.1	<0.1	<0.1	<0.1	<0.1	11.4	<0.1
Co mg/kg	0.1	<0.1	0.4	<0.1	0.5	<0.1	0.6	0.2	2.3	<0.1
Sr mg/kg	0.5	89	1110	1320	876	614	529	406	153	548
Y mg/kg	0.01	13.60	1.40	0.73	0.61	1.13	0.40	0.31	2.90	1.03
Zr mg/kg	0.1	0.8	1.2	1.2	2.2	1.8	1.6	1.0	0.6	1.8
Sn mg/kg	0.05	<0.05	0.22	0.11	<0.05	0.09	0.09	<0.05	0.58	<0.05
Ba mg/kg	0.5	12	205	198	206	355	187	205	92	172
La mg/kg	0.5	4.7	1.0	<0.5	0.6	0.6	<0.5	<0.5	3.3	0.5
Ce mg/kg	0.01	4.25	1.91	0.77	1.43	1.01	0.71	0.66	6.95	0.87
Pb mg/kg	0.01	1.25	2.41	0.79	0.38	0.85	0.39	0.44	5.13	0.62
Ni mg/kg	0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	8.3	<0.1
Se mg/kg	0.1	<0.1	0.3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Hg µg/kg	10	20	30	30	20	<10	20	30	50	10

Table 4

Chemical Characteristics of Lime Sludge and Limestone Samples Determined by Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS).

	unit	detection limit	limestone	SL1	SL2	SL3	SL4	SL5	SL6	SL7	SL8
Hg	µg/kg	10	20	30	30	20	<10	20	30	50	10
S	wt %	1	<1	<1	<1	<1	<1	<1	<1	<1	<1
B	mg/kg	1	<1	62	17	5	142	35	9	27	76
Na	wt %	0.001	0.021	0.065	0.082	0.045	0.065	0.05	0.042	0.036	0.067
Mg	wt %	0.01	0.55	3.34	1.87	1.07	2.99	2.79	1.49	3.61	3.45
Al	wt %	0.01	0.02	0.07	0.03	0.09	0.04	0.05	0.05	0.62	0.09
K	wt %	0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.13	<0.01
Ca	wt %	0.01	33.6	31.3	34.5	34.8	35.1	32.6	31.6	26	30.6
Mn	mg/kg	1	235	139	59	568	54	623	364	293	150
Fe	wt %	0.01	0.05	0.53	0.4	0.05	0.34	0.03	0.06	1.34	0.28
P	wt %	0.001	0.006	0.03	0.012	0.01	0.024	0.009	<0.001	0.046	0.016
Cu	mg/kg	0.01	0.42	5.74	4.25	1.93	1.81	4.28	2.2	11.8	1.72
Zn	mg/kg	0.1	8.7	13.7	6.7	8.1	9.3	1.3	0.6	34.4	9
As	mg/kg	0.1	<0.1	1.2	<0.1	<0.1	3.8	0.6	<0.1	2.8	5.9
Rb	mg/kg	0.1	0.5	0.7	0.1	<0.1	<0.1	<0.1	<0.1	11.4	<0.1
Co	mg/kg	0.1	<0.1	0.4	<0.1	0.5	<0.1	0.6	0.2	2.3	<0.1
Sr	mg/kg	0.5	89.4	1110	1,320	876	614	529	406	153	548
Y	mg/kg	0.01	13.6	1.4	0.73	0.61	1.13	0.4	0.31	2.9	1.03
Zr	mg/kg	0.1	0.8	1.2	1.2	2.2	1.8	1.6	1	0.6	1.8
Sn	mg/kg	0.05	<0.05	0.22	0.11	<0.05	0.09	0.09	<0.05	0.58	<0.05
Ba	mg/kg	0.5	12.2	205	198	206	355	187	205	91.5	172
La	mg/kg	0.5	4.7	1	<0.5	0.6	0.6	<0.5	<0.5	3.3	0.5
Ce	mg/kg	0.01	4.25	1.91	0.77	1.43	1.01	0.71	0.66	6.95	0.87
Pb	mg/kg	0.01	1.25	2.41	0.79	0.38	0.85	0.39	0.44	5.13	0.62
Ni	mg/kg	0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	8.3	<0.1
Se	mg/kg	0.1	<0.1	0.3	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1

Table 5

Reemission and Liquid–Solid–Gas Distribution of Mercury During 2 h Tests.

	Hg reemission peak concentration ($\mu\text{g}/\text{m}^3$)	cumulative Hg reemitted in 2 h (μg)	total Hg in solids	total Hg in liquid (wt %)	Hg in gas	Hg in solids	Hg in liquid
Limestone	200	2.74	11.2	41.1	5.0	20.4	74.6
SL1	149	2.88	47.7	4.52	5.2	86.6	8.2
SL2	89.8	3.16	20.5	31.4	5.7	37.2	57.1
SL3	165	1.86	6.22	47.0	3.4	11.3	85.3
SL4	77.2	1.42	7.51	46.1	2.6	13.6	83.8
SL5	177	2.36	3.11	49.6	4.3	5.6	90.1
SL6	179	1.87	4.92	48.3	3.4	8.9	87.7
SL7	147	2.85	44.8	7.40	5.2	81.4	13.4
SL8	114	2.83	15.8	36.4	5.1	28.7	66.2

Table 6

Concentrations of Various Elements in the Solutions Collected at the End of Scrubbing Experiments with the Selected Lime Sludge Samples and Limestone Sample. Concentration unit is mg/L.

	limestone	SL1	SL4	SL8
B	2.1	2.3	2.1	2.6
Mg	121	>200	77	157
Al	0.14	0.28	0.24	0.34
Mn	>5	0.83	0.15	0.76
Fe	0.11	0.20	0.11	0.12
Cu	0.013	0.014	0.007	0.011
Zn	0.063	0.008	0.008	0.007
As	0.0011	0.0017	0.0042	0.0049
Sr	0.37	4.0	1.2	2.1
Ba	0.025	0.04	0.036	0.033