2019

Flue Gas Desulfurization Gypsum as Soil Amendment

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Panday, Dinesh; Ferguson, Richard; and Maharjan, Bijesh, "Flue Gas Desulfurization Gypsum as Soil Amendment" (2019). *Agronomy & Horticulture -- Faculty Publications*. 1245.  
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Flue Gas Desulfurization Gypsum
as Soil Amendment

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1 Flue Gas Desulfurization Gypsum

Flue gas desulfurization (FGD) gypsum is one of the by-products of a coal-fired power generation plant. Coal is the world’s most abundant and widely distributed fossil fuel. After natural gas, coal is the second primary source of energy to generate electricity globally (more than 25%) and remains a key component of the fuel mix for power generation to meet electricity demand in most of the developing countries. The U.S., China and India are the top coal producers and consumers (for production of electricity from coal sources) in the world (OECD/IEA, 2014; IEA, 2016). However, in the U.S., its contribution to power generation is declining in favor of natural gas and other energy sources due to low natural gas prices, renewable energy standards and environmental activism and regulations.

1.1 Sources of FGD Gypsum

Coal combustion in power plants generates about 120 million metric tons of coal combustion residues (CCR) annually. These by-products include fly ash, bottom ash, flue gas desulfurization (FGD) material and flue bed combustion ash. According to the American Coal Ash Association (ACAA, 2015), only 61.1 million metric tons of CCR were beneficially used. The 1990 U.S. Clean Air Act Amendments restrict sulfur dioxide (SO$_2$) emissions into the atmosphere from coal-fired facilities, if the coal contains considerable amounts of sulfur (S). To meet the SO$_2$ emission reduction requirements, most of the U.S. coal power plants use the FGD process, and in this process, the gypsum is produced which is known as FGD gypsum.

FGD gypsum is created by forced oxidation scrubbers in coal-fired power plants which remove SO$_2$ emission from the flue gas stream. There are three different scrubbing processes: wet, semi-dry and dry. However, SO$_2$ removal efficiencies are significantly higher in wet scrubbing process (90 to 98%) than semi-dry (80 to 90%) and dry (50 to 60%) processes for calcium-based sorbents (Schnelle and Brown, 2002). In general, a wet scrubbing process first exposes the flue gases to a slurry of hydrated lime, where it reacts with S in the gas to form calcium sulfite (CaSO$_3$). Forcing additional air into the system oxidizes the CaSO$_3$ and converts it into gypsum. The FGD gypsum is also known as recaptured gypsum, byproduct gypsum and synthetic gypsum.
The chemical formula for mined gypsum or FDG gypsum is the same, which is calcium sulfate dihydrate (CaSO$_4$$\cdot$2H$_2$O). By weight, it is 79% calcium sulfate and 21% water. It contains 23% calcium (Ca) and 18% sulfur (S). However, the amount and types of trace materials and unreacted sorbents found in the gypsum can vary among power plants and among mines. FGD gypsum contains 90 to 99% of purity concentration compared to 66 to 98% concentration in mined gypsum. Production of FDG gypsum has gradually increased in the past several years. According to the ACAA, approximately 33 million metric tons of FGD gypsum was produced in 2015 in the U.S., of which 53% (17.5 million metric tons) was used in building industry and road construction. Less than 2% of the total FGD gypsum production was used in agriculture.

1.2 Properties of FGD Gypsum

Compared to mined gypsum, FGD gypsum has more desirable spreading characteristics, which allows for easy application (Dontsova et al., 2004). It is a direct source of macronutrients, supplying readily available calcium (Ca$^{2+}$) and sulfate (SO$_4^{2-}$) ions for plants. It is considered moderately soluble in soil and has a solubility 200 times greater than lime or calcium carbonate (CaCO$_3$), thereby slowly releasing S over multiple years. It may also contain sodium chloride (NaCl), magnesium oxide (MgO), calcium chloride (CaCl$_2$), phosphoric oxide (P$_2$O$_5$), CaCO$_3$, silicon dioxide (SiO$_2$) and other by-products such as fluorine (fluoride compounds). Moreover, it generally has finer, more uniform particles than mined gypsum sources.

2 FGD Gypsum In Agriculture

2.1 Source of Plant Nutrients

Mined gypsum has been applied to agricultural soils for more than 250 years in those crops which have high Ca requirements, or to areas that have Ca poor soils since it is an excellent source of soluble Ca and S. Root and field crops such as peanut (Arachis hypogaea), potato (Solanum tuberosum), tomato (Solanum lycopersicum), corn (Zea mays L.), wheat (Triticum aestivum), etc. seem to especially respond to Ca,
and application of FDG gypsum can improve both yield and quality of products. Similarly, S fertilization is required for many crops, such as alfalfa (Medicago sativa), soybean (Glycine max L.), canola (Brassica napus L.), etc., and application of FGD gypsum can be an effective source of S (Wang and Yang, 2017). In general, crops grown on soil with low organic matter and coarse-textured can respond to S application. In addition to Ca and S, FDG gypsum also provides some essential micronutrients to plants. Not all FGD gypsum will be acceptable for agricultural use because of high chloride content and potential issues associated with heavy metals.

In the U.S., an imposition of SO₂ emissions standards on power plants has reduced atmospheric S deposition on soil, thereby reducing S levels in soils. The typical row crops such as corn and soybean result in a net removal of nutrients from the soils, if there are no supplemental nutrients added to soils. Modern agricultural practices are increasing crop yields, but at the cost of soil depletion in nutrients. Harvest removes plant material rich in nutrients supplied by soil. This annual removal of nutrients can be compensated for by applications of inorganic fertilizers which can be very costly. Soil amendments, such as FGD gypsum, are not substituted for sources of macro- and micro-nutrients but can supply certain nutrients as well as improve soil properties and processes whereby they sustain soil productivity.

### 2.2 Soil Improvement

In addition to supplying Ca and S for plant nutrition, many researchers have shown that FGD gypsum can be used as a soil conditioner to improve physical and chemical properties by promoting better aggregation, increasing water infiltration rate and movement through the profile, reclaiming sodic soils, mitigating subsoil acidity and aluminum (Al) toxicity and reducing soil and soluble phosphorus (P) loss from agricultural fields (Watts and Dick, 2014).

#### 2.2.1 Physical Properties

Sodic soils are characterized by the occurrence of excess sodium (Na) to levels that can adversely affect soil structure and availability of some nutrients.
Sodium (Na) dominated soils are called sodic soils, also known as very poor agricultural soils. The presence of excessive Na ions adversely affects soil structure and disturbs the availability of some nutrients in the soil for plants (Qadir et al., 2001). FDG gypsum is helpful as an effective product used in the remediation of such types of sodic soils, and soils having crusting and other structural problems. Gypsum itself is more readily soluble in water than limestone and therefore may move throughout the soil profile more easily. The Ca ions present in gypsum can exchange with Na ions on clay particles and reduce the dispersion of soil particles by promoting clay particles to bind together (flocculate).

Many studies have shown that tillage after FDG gypsum application increases subsoil exchangeable Ca ion concentration, and allows roots to penetrate subsoils. Many soils from semiarid to humid regions have an unstable structure and are susceptible to erosion. The application of FGD gypsum promotes flocculation, reduces dispersion of soils and slows the rate of surface drying, which is a necessary condition for the formation and stabilization of soil structure. It helps to reduce soil crust formation, which improves seed emergence and plant establishment. It makes it easy to manage unstable structure, which can increase potentially available water and percolation, thus reducing soil erosion and improving water quality (Figure 1) (Chen and Dick, 2011).

2.2.2 Chemical Properties

2.2.2.1 Improving Acidic Soils

Acidic soils hold a higher concentration of hydrogen (H) and Al ions, which could be due to their formation from parent materials or from the application of ammonium (NH$_4^+$) based fertilizers. In soil with pH ≤ 5.5, Al$^{3+}$ toxicity in plants is observed, mainly in roots. Damages in the upper parts due to Al$^{3+}$ toxicity may also be possible (Meriño-Gergichevich et al., 2010). Unlike Al$^{3+}$, manganese (Mn$^{2+}$) is an essential plant micronutrient, but it also is a metal and could become toxic in very acidic soils. All these problems with acidic soils reduce the availability of other essential nutrients and lead to poor plant growth.

The application of lime, as well as FGD gypsum that has a considerable amount of CaCO$_3$ would be highly beneficial to soils containing acidic subsoils. Amelioration of acidic subsoils is harder than topsoils. Since FGD gypsum promotes downward movement of Ca$^{2+}$ in the soil profile, it can help reduce acidity in subsoil. However, the downward movement of Ca$^{2+}$ is dependent on tillage depth, soil texture and rate of the FGD gypsum applied. Sumner (1993) described a mechanism involved in subsoil acidity amelioration using FGD gypsum as (i) the self-liming effect; (ii) precipitation of solid phases; (iii) co-sorption of (SO$_4^{2-}$) and Al$^{3+}$; and (iv) ion pair formation. Overall, by applying FGD gypsum, it enhances the soil pH value and reduces the exchangeable Al ion concentration.

2.2.2.2 Improving Nutrient Availability

The application of FGD gypsum on weathered soils increases the sorption activity of Ca$^{2+}$ and (SO$_4^{2-}$) by plants, and results in improvement of nitrogen (N) uptake. This may reduce the need to apply more N to the plants, as well as diminish the potential for nitrate (NO$_3^-$) contamination of surface and ground waters. Chen et al. (2008) conducted field experiments to study the interaction effects of N and S fertilization on corn growth and yield. They found that application of FGD gypsum (33 kg S ha$^{-1}$) with N (0-233 kg N ha$^{-1}$) promoted corn growth and uptake of N in a silt loam soil in Ohio, U.S. However, the addition of gypsum in young soils, which weather readily and release
electrolytes, will have fewer effects. The excess of gypsum in sandy soils may cause a tie-up of magnesium (Mg) and potassium (K) (Levy and Sumner, 1998).

It is important to note that all forms of gypsum are not a liming agent and do not affect the pH of the soils. However, FGD gypsum can ameliorate the phytotoxic conditions arising from excess soluble Al in acid soils. It reacts with $\text{Al}^{3+}$ and removes it from the soil solution and thus greatly reduces the toxic effects (Smyth and Cravo, 1992). This can effectively increase the supply of water and nutrients to the crops due to the improvement of a deep rooting system (Chen and Dick, 2011). Additionally, FGD gypsum can be used as a substitute for agricultural limestone to solid waste stream and utilized to restore degraded landscapes. Chen et al. (2013) carried out a 16-year long-term study to investigate the use of FGD gypsum for reclamation of an abandoned surface coal-mined land in Ohio, and found that the use of FGD gypsum for remediating acidic surface coal-mined sites could provide effective long-term reclamation.

2.2.3 Reducing Soil and Nutrient Loss

Soil-applied FGD gypsum releases electrolytes that prevent soil surface sealing, thereby preventing a leading cause of soil erosion (Baligar et al., 2011). It increases ionic strength and $\text{Ca}^{2+}$ concentration in the soil solution. Thus, adsorption of phosphate ($\text{PO}_4^{3-}$) becomes stronger, and it reduces the dispersion of soil particles by promoting flocculation and aggregation of clay particles. Converting readily soluble reactive P into insoluble Ca phosphate complex, FGD gypsum can reduce nutrient runoff, mainly P, into receiving adjacent streams, lakes or groundwater (USEPA, 2008).

Norton and Rhoton (2007) reported that FGD gypsum application reduced water runoff by 17%, soil loss by 60% and P losses by 67% when compared to the control. Jaakkola et al. (2012) found that FGD gypsum reduced total P losses by 44% into the field scale simulation model. Excess of P in runoff leads to water quality problems, including algal blooms and eutrophication of water resources. The use of FDG gypsum as a soil amendment would be helpful to reduce the eutrophication in surface waters by reducing the runoff losses of P, N and carbon (C) through increased infiltration. Additionally, Alan et al. (1998) conducted a leaching column experiment to investigate
the effect on the transport of NO$_3^-$ and NH$_4^+$ using FDG gypsum in a Candler fine sand soils, and found that FDG gypsum at the rate of 4.5 Mg ha$^{-1}$ decreased the leaching from ammonium nitrate (NH$_4$NO$_3$) by 22% as compared to the control.

**2.3 FDG Gypsum Use in Agriculture: A Case Study**

*(Adapted from Maharjan et al., in preparation)*

A study was conducted to evaluate the potential use of FGD gypsum in improving soil properties and crop production on irrigated cropland in Adams County in Nebraska, U.S. in 2014 and 2015 (*Figure 2*). The selected site had variable soil properties, with some areas of eroded topsoil, and it was planted to corn both years. A randomized complete block design with four replications of field length treatment strips (60 ft or 18.3 m wide) was implemented. The main treatment was FGD gypsum rate and was applied at four rates of 0, 1000, 4000 and 8000 lbs acre$^{-1}$ (equivalent to 0, 1.1, 4.5, and 9.0 Mg ha$^{-1}$).

*Figure 2.* Study site in Adams County, Nebraska, U.S. (Courtesy of Dr. Bijesh Maharjan.)
The source of FGD gypsum was a local coal-powered power generation plant, and 50% of this FGD gypsum was CaCO$_3$ by content. The FGD gypsum treatment was applied using a 1844 Terragator tractor with flotation tires that pulled a 1034G4 New Leader spreader box with Raven controller mounted on a trailer which also had floatation tires. The system allowed the operator to control the rate, swath width, configuration settings and other aspects of application. Prior to FGD gypsum application, both fields were mapped with a VERIS MSP-3R instrument for soil apparent electrical conductivity ($\text{EC}_a$), organic matter content (OM) and pH. Based on these maps and apparent topography variability, locations of treatment strips were determined.

Each year, grain and stover were hand-sampled from geo-referenced sampling locations (GSL) based on OM and pH maps and apparent topography variability in each treatment strip. Manual harvest was followed by combine harvest by the cooperator in a few weeks. After the fields were cleared following combine harvest, soil samples were collected at each GSL points at depth increments of 0–20 cm, 20–40 cm and 40–60 cm. In the following spring, soil physical properties such as bulk density, porosity, penetration resistance and sorptivity were measured at GSL points where hand harvest and soil samples were collected the previous fall.

All grain and stover samples and upper 20 cm soil samples were analyzed for arsenic (As), selenium (Se), cadmium (Cd), chromium (Cr), lead (Pb) and mercury (Hg). All grain and stover samples were also analyzed for total C and N contents while the upper 20 cm soil samples were analyzed for agronomic chemical properties of pH, OM, CEC, P, S, K, Ca, Mg, Zn and nitrate-N. Soil samples from depths of 20–40 cm and 40–60 cm were analyzed for nitrate-N.

Grain yield data collected from combine harvest following FGD gypsum application was segregated by management zones (MZ) based on soil organic matter or soil pH and analyzed separately. The experiment was initiated with assumptions that FGD gypsum may positively affect sub regions of fields with lower organic matter content (due to decreased availability of S from organic matter and S supply from FGD gypsum) and with low pH (due to the presence of unreacted CaCO$_3$ in FGD gypsum and subsequent liming effect). Therefore, soil OM and pH were two variables used to delineate MZ to take spatial variability into account while determining FGD gypsum effect on grain yield.
There was no detrimental effect of FGD gypsum application on soil or crop production. There was no metal contamination in soil, grain or stover or reduction on grain yield following FGD gypsum application. As far as spatial variation in the field is concerned, grain yield was not affected by different soil pH ranges but by different OM content in soil. Greater grain yield was observed at sub regions of the field with higher OM (Figure 3). There was neither main effect of FGD gypsum rate nor interaction effect of FGD gypsum rate and MZ based on pH or OM. This suggests that FGD gypsum did not have any significant positive effects on grain production either through liming or S fertilization effects in the given two years. However, there was a trend for FGD gypsum rate to increase yield ($P = 0.06$). Grain yield was numerically greater in FGD gypsum treatments compared to the control treatment, especially in MZ with higher OM content. This observation is apparently conflicting to an initial assumption that FGD gypsum may positively affect sub regions with lower soil OM. However, there could be interacting effects of FGD gypsum treatment with different soil conditions, such as water-holding capacity, aggregation, etc. due to varying soil OM content. There is a consensus that it takes multiple years after application of amendment before measurable benefits are observed. Therefore, monitoring the site for a longer period could help identify factors that mediate the contrasting responses to FGD gypsum application across MZ.

**Figure 3.** Strong trend ($P = 0.06$) of the positive effect of FGD gypsum rate treatment on mean (standard error) corn grain yield, especially in management zones 3 and 4 based on soil OM.
3 FGD Gypsum Management

3.1 Risk Associated with FGD Gypsum

As a by-product from coal-fired power plants, one of the great concerns about FGD gypsum is that it contains heavy metals, such as Hg, Cd, As, Cr, Pb or Tl (thallium) introduced either from the coal used as a fuel for power generation or from the limestone used for desulfurization (Chen et al., 2015). The introduction of heavy metals with the FGD gypsum application into the sodic soils may give rise to ecological hazards in the soil environment because of accumulation of heavy metals in the soil, and exposure to heavy metals is generally chronic.

Hao et al. (2016) found a slight increase in heavy metal concentrations when FGD gypsum was used for amelioration of alkali soils in China; however, those metals concentration were far below the background values stipulated by the Environmental Quality Standard for Soils (GB15618-1995). Typical trace constituents in FDG gypsum are 0.01–1.4 ppm for Hg, 0.02–1.2 ppm for Cd, 0.6–4 ppm for As, 8.7–30.5 ppm for Cr, 0.6–2 ppm for Tl and 0.8–12 ppm for Pb (Maloney, 2013). Sanchez et al. (2008) found that B, Cd, Mo, Se and Tl may be released from FGD gypsum at levels exceeding either a maximum contaminant level or drinking water equivalent level under some conditions under exposure to water. There are growing concerns regarding the environmental risks associated with FGD gypsum applied soils. However, more information is needed to explore the risks associated with the introduction of heavy metals.

3.2 Determining the Appropriate Application Rate

The general consideration for amending soil properties with gypsum products given by Natural Resources Conservation Service, U.S. Department of Agriculture (NRCS, 2015) includes:

1. Gypsum should not be applied in watersheds where sulfate additions are restricted.
2. If soil pH is less than 5, the application of products with high sulfite content may be harmful to plants that are present at the time of application.
3. Long-term use of gypsum or using rates higher than given in the criteria can have adverse impacts on soil or plant systems. This can include:

- Where gypsum derived products are alkaline due to impurities, raising the soil pH to a level that is detrimental to plant growth or nutrient balance.
- Creating a Ca imbalance with other mineral nutrients such as Mg and K.

FGD gypsum is not suitable for all soil types, soil conditions or crops. Appropriate application rates should be determined by soil analysis, especially for electrical conductivity (EC) and sodium adsorption ratio (SAR) measurement. It also depends upon the specific purposes of soil amendments or to supply fertilizer minerals like Ca, S and B to plants. An over application rate of FGD gypsum may result in seedling damage and nutrient imbalance.

The US EPA states, “In general, application rates of up to 2 tons acre$^{-1}$ (equivalent to 4.94 tons ha$^{-1}$) should be sufficient to accomplish most agronomic and horticultural objectives.” Mixing of at least 1 ton acre$^{-1}$ (equivalent to 2.47 tons ha$^{-1}$) of FGD gypsum with manure prior to application is recommended to improve surface water quality by reducing dissolved P concentrations in surface runoff. Rates of FGD gypsum as high as 10–30 tons acre$^{-1}$ (equivalent to 24.71-74.13 tons ha$^{-1}$) have been used as soil amendments. Chen and Dick (2011) summarized recommended rates, time of application and method of gypsum application for various functions as given in Table 1. However, management practices for specific uses of FGD gypsum also need to be developed across a range of soils, cropping systems and climate regimes.

### 3.3 Economic Consideration for FGD Gypsum Use

Although FGD gypsum has been widely used in many developed countries, there is less successful adoption of FGD gypsum in developing countries. There might be many reasons for it, primarily the associated costs of SO$_2$ scrubbing, transportation of FGD gypsum and land application. However, the FGD technology is promising in terms of transforming waste product into a beneficial product that increases
crop production and improves soil quality in an economically and environmentally sustainable manner as many developing countries would benefit from.

Economic consideration should include the cost-benefit analysis. A study on tomato yield and value in Mississippi increased 9% from 2 tons acre⁻¹ (equivalent to 4.94 tons ha⁻¹) when compared to the control (Sumner and Larrimore, 2006). Typically, application rates are 1 to 2 tons acre⁻¹ in every one to two years, and it costs $30 to $50 acre⁻¹ in the U.S. Chen and Dick (2011) reported that dewatering of FGD gypsum can reduce the transportation costs. The spreading costs for FGD gypsum would be similar to that for lime.

Table 1. Rate, Time, and Method of Application of Gypsum for Various Functions

<table>
<thead>
<tr>
<th>Function</th>
<th>Suggested Rates of Application (tons acre⁻¹)</th>
<th>Suggested Time of Application</th>
<th>Suggested Application Method</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Function</strong></td>
<td><strong>Low</strong></td>
<td><strong>Normal</strong></td>
<td><strong>High</strong></td>
</tr>
<tr>
<td>Sulfur fertilizer to enhance crop production</td>
<td>0.05</td>
<td>0.15</td>
<td>0.25</td>
</tr>
<tr>
<td>Calcium fertilizer to enhance crop production</td>
<td>1</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>(especially root crops, e.g., peanuts)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Soil amendment to remediate subsoil acidity</td>
<td>2</td>
<td>3</td>
<td>5</td>
</tr>
<tr>
<td>Soil amendment to remediate sodic or sodium-affected soils</td>
<td>1</td>
<td>5</td>
<td>10</td>
</tr>
<tr>
<td>Soil amendment to improve water quality</td>
<td>1</td>
<td>3</td>
<td>5</td>
</tr>
<tr>
<td>(e.g., by reducing phosphorus concentrations in surface water runoff)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Soil amendment to improve soil physical properties</td>
<td>1</td>
<td>2</td>
<td>5</td>
</tr>
<tr>
<td>and water infiltration and percolation</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>As a lawn care product and sport field application</td>
<td>2</td>
<td>4</td>
<td>7</td>
</tr>
</tbody>
</table>

4 Conclusion

Mined gypsum has been used as a plant nutrient source and soil conditioner in agricultural production for a long time. At present, gypsum for agricultural use is derived from both mined and synthetic sources. Industrial by-product such as FGD gypsum can potentially be a more economic source of gypsum as well as provide additional agricultural and environmental benefits by supplying nutrients (Ca and S) for plants, ameliorating sodic and acidic soils, improving soil physicochemical properties and reducing soil and nutrient (P) losses.

The annual production of FGD gypsum will increase continuously, since more coal-fired power plants may come online for power generation, and those power plants’ facilities are upgraded to meet the SO$_2$ emissions requirement. There are significant areas of degraded soils, which could benefit from FGD applications. There is no report of negative effects due to FGD gypsum application, but a good understanding of its composition and properties are very essential to know and avert the possible environmental risks. FGD gypsum may not be suitable for all soil types, soil conditions or crops, and hence management practices for specific uses also need to be developed across a range of soils, cropping systems and climate regimes.

References


Maharjan, B. et al. (in preparation) Evaluation of flue gas desulfurization gypsum in irrigated cropping systems.


