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RESEARCH ARTICLE

Use of flue gas desulfurization gypsum to reduce dissolved phosphorus in runoff and leachate from two agricultural soils

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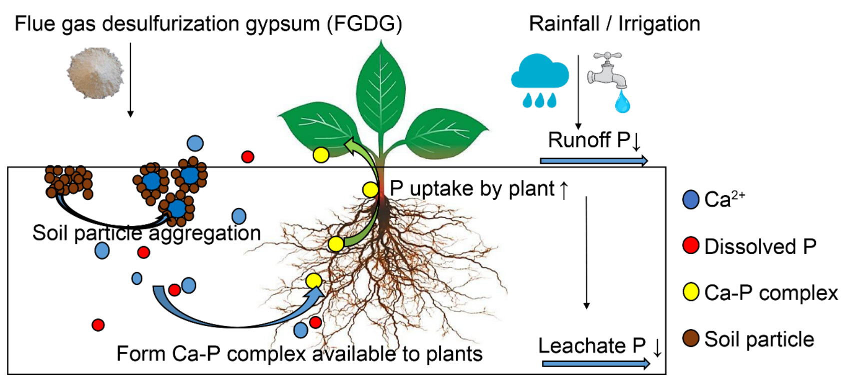
HIGHLIGHTS

• Gypsum can effectively decrease dissolved P loss via runoff and leachate from the areas with high soil P levels by increasing P uptake by ryegrass.
• Both surface application and mixing of gypsum into the topsoil reduced dissolved P losses.
• The effect of gypsum application method on dissolved P losses varied by soil texture.
• Flue gas desulfurization gypsum did not affect ryegrass biomass and also didn't increase the accumulation of trace elements in soil and ryegrass.

GRAPHICAL ABSTRACT



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ABSTRACT

Controlling dissolved phosphorus (DP) loss from high P soil to avoid water eutrophication is a worldwide high priority. A greenhouse study was conducted in which flue gas desulfurization gypsum (FGDG) was applied by using different application methods and rates to two agricultural soils. Phosphorus fertilizer was incorporated into the soils at 2.95 g kg^{-1} to simulate soil with high P levels. The FGDG was then applied at amounts of 0, 1.5, and 15 g kg^{-1} soil on either the soil surface or mixed throughout the soil samples to simulate no-tillage and tillage, respectively. Ryegrass was planted after treatment application. The study showed that FGDG reduced runoff DP loss by 33% and leachate DP loss 38% in silt loam soil, and runoff DP loss 46% and leachate DP loss 14% in clay loam soil, at the treatment of 15 g kg^{-1} FGDG. Mixing applied method (tillage) provided strong interaction with higher FGDG. To overall effect, the mixing-applied method performed better in controlling DP loss from silt loam soil, while surface-applied (no tillage) showed its advantage in controlling DP loss from clay loam soil. In practice it is necessary to optimize FGDG concentrations, application methods, and DP sources (runoff or leachate) to get maximized benefits of FGDG application. The FGDG application had no negative effects on the soil and ryegrass.

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1 Introduction

Phosphorus (P) applied to soil as fertilizer or manure in excess of crop requirements can lead to a buildup of available P (AP) in the soil. High levels of AP can lead to dissolved P (DP) loss from soil. Dissolved P is a primary cause of eutrophication in receiving water bodies (Sharpley and Wang, 2014). In 2011, a cyanobacteria bloom in Lake Erie, USA was mainly attributable to DP coming from agricultural lands (Michalak et al., 2013).

Reduction of P loss as DP from agricultural lands via runoff and leaching (Smith et al., 2015; Wang et al., 2018) is needed. Gypsum is an excellent source of Ca and agricultural grade gypsum can be mined from natural deposits in the earth. Flue gas desulfurization gypsum (FGDG) is created during a process that removes sulfur dioxide (SO_2) from the flue gases of coal-burning power plants and typically consists of 95.0%−99.6% $CaSO_4 \cdot 2H_2O$ (Dick and Chen, 2011; Watts and Dick, 2014). FGDG has many potential applications in agriculture (Wang and Yang, 2018; Koralegedara et al., 2019) and has been used to remediate saline-alkali soils and to reclaim surface coal mine lands (Chen et al., 2015; Mao et al., 2016). Flue gas desulfurization gypsum is also used as a source of essential plant nutrients such as Ca and S needed for optimum crop growth (Chen et al., 2011; Li et al., 2017), and to reduce DP loss from agricultural soil by precipitating Ca-P complex (Torbert and Watts, 2014; He and Li, 2019).

Questions remain, however, as to the best strategy for applying gypsum to reduce DP losses from soil. If no-tillage crop production is practiced, it is impossible to mix the gypsum with the soil. Also, questions remain about the amount of gypsum needed to affect DP losses from the soil and whether there is an interaction with tillage (or lack thereof), soil texture, and application rate. The objectives of this study were to evaluate the effect of the application concentration and application method of FGDG on reducing DP losses via runoff and leachate from two contrasting agricultural soils. Results will be useful in informing producers and consultants about the most effective management strategies for applying gypsum to soil to reduce DP losses and improve soil and water quality.

2 Materials and methods

2.1 Experimental setup

Soil samples (0−20 cm depth) from two locations in Ohio, USA, Wooster and Hoytville, were collected using a flat bottom spade and brought to the greenhouse. The Wooster soil collected was a silt loam, Oxyaquic Fragiudalfs, and the Hoytville soil was a clay loam, Mollic Epiaqualfs (Campbell, 2014). Soil samples were air-dried for one week, crushed, and passed through a 2-mm sieve to remove root material. Selected properties of the two Ohio soil samples (Table 1) and the properties of FGDG used in this study (Table 2) are provided.

A greenhouse experiment was conducted by mixing 2.95 g kg^{-1} chemical-grade triple superphosphate ($Ca(H_2PO_4)_2 \cdot H_2O$) (P_2O_5 46%) (2.95 g kg^{-1} is approximately equivalent to 660 kg ha^{-1}) into each treatment to greatly increase available P (Table 1). The experimental design was a factorial randomized block design with three replications. Treatments included (1) two application concentrations of FGDG, G and GG (1.5 and 15 g kg^{-1} that were approximately equivalent to 336 and 3360 kg ha^{-1}, respectively) and (2) either surface-applying (S) or mixing (M) the gypsum into the entire soil volume to simulate no-tillage and tillage, respectively; a fifth treatment (0) was a control treatment without FGDG.

A amount of 17 kg soil with/without FGDG treatment was packed inside plastic boxes of size 38 cm (long) by 25.5 cm (wide) by 23.5 cm (high) (Fig. 1). Three holes (1.5 cm diameter) were drilled evenly across the bases of the boxes and an aluminum foil plate was placed below the boxes to collect leachate water. A modified syringe (1.5 cm diameter) at the downslope (3%) end of the runoff boxes diverted all

Table 1 Initial properties of two soil samples from Ohio.

Soil	Silt loam soil	Clay loam soil
pH	6.59	6.14
Available P (g kg^{-1})$^{\alpha}$	0.038	0.057
Total P (g kg^{-1})	0.41	0.70
Exchangeable Ca	1.12	3.00
Total N (%)	0.104	0.256
Total C (%)	0.88	2.25
Clay $^{\beta}$	15	40
CEC $^{\gamma}$(cmol kg^{-1})	7.30	21.8
Bulk density (g cm^{-3})	1.40	1.35

$^{\alpha}$ Available phosphorus as determined by the Bray-1 solution. $^{\beta}$ Data from Campbell et al. (2014). $^{\gamma}$ Cation exchange capacity.

Table 2 Concentrations of selected elements in FGDG, soils and the Standard in comparison.

Elements	FGDG	Clay loam soil	Silt loam soil	Standards$^{\gamma}$
Major elements				
Ca (g kg$^{-1\,\alpha}$)	198	–	–	–
Mg (g kg$^{-1\,\alpha}$)	2.19	–	–	–
S (g kg$^{-1\,\alpha}$)	159	–	–	–
P (g kg$^{-1\,\alpha}$)	0.100	–	–	–
Na (g kg$^{-1\,\alpha}$)	0.144	–	–	–
K (g kg$^{-1\,\alpha}$)	0.421	–	–	–
Fe (g kg$^{-1\,\alpha}$)	2.39	–	–	–
Trace elements		–		
Zn (mg kg^{-1})	93.7	93.7	69.3	2800
Cr (mg kg^{-1})	5.06	35.8	25.2	
Pb (mg kg^{-1})	1.43	13.2	17.6	300
Cd (mg kg^{-1})	0.73	1.90	1.47	39
As (mg kg^{-1})	4.50	4.82	8.89	41
Ni (mg kg^{-1})	4.21	28.1	19.9	420
Cu (mg kg^{-1})	<0.43 $^{\beta}$	23.8	7.16	1500
Hg (mg kg^{-1})	<5.58	<5.58	<5.58	17

$^{\alpha}$ Dry weight basis. $^{\beta}$ Below detectable limit (the number is the detectable limit). $^{\gamma}$ Standards for the use or disposal of sewage biosolids (US EPA, 1993).

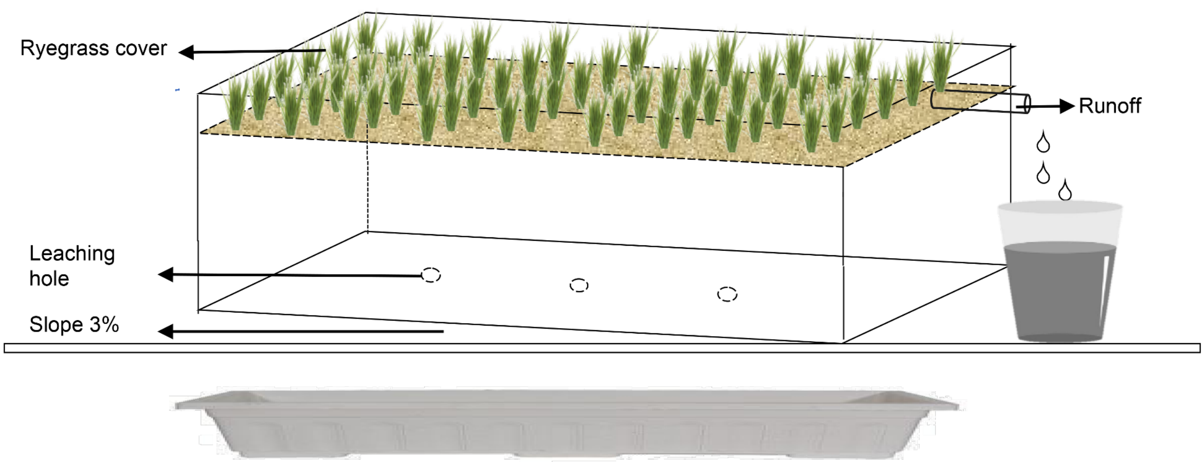

Fig. 1 Schematic of soil boxes used to collect runoff and leachate waters.

runoff water to a plastic collection vessel.

FGDG and phosphorus fertilizer were applied to the soils according to the experimental design, followed by 3000 mL deionized water to moisten the soil, which was then incubated for 24 hours. Two hundred ryegrass (*Lolium perenne*) seeds were planted in each box, covered with a 1.0 cm dry soil, and wetted with deionized water. Three weeks after planting, seven rainfall events were applied at two-week intervals to each box, a simple rainfall simulator with 1500 mL water placed 10 cm above soil surface of each box provided a 30-min rainfall event at 30 mm h^{-1}. Between simulated rainfall events, the watering frequency was based on the water requirement of plant growth and evaporation; the amount required was enough to keep ryegrass growing well, and all boxes received the same amount of water. The experiment lasted 15 weeks from sowing to the last rainfall event. The greenhouse was maintained at 28°C for daytime and 20°C for nighttime. Light was provided for 14 h, relative humidity was kept at 60%.

2.2 Sampling and analysis

The runoff and leachate water were collected after every rainfall event, and the flow volumes and DP concentrations in the runoff and leachate water were measured. Dissolved

P in water samples was determined colorimetrically by the molybdenum-blue method after filtering through a 0.45 μm membrane filter (Murphy and Riley, 1962). Dissolved P accumulation was calculated by multiplying DP concentrations by flow volumes per time, and then adding them up. Average DP concentration was calculated by DP accumulation divided by volume accumulation. Total DP accumulation in combined runoff plus leachate was calculated by summing runoff DP accumulation and leachate DP accumulation.

Soil samples and ryegrass were collected at the end of the experiment. Five soil cores (3 cm diameter) per box were collected and combined to create one sample. Soil samples were air-dried, crushed, passed through a 2-mm sieve and the concentrations of available P and total P were analyzed. All the ryegrass/box containing above-ground biomass and roots were collected, dried in an oven for 48 h at 60°C, and then yield was calculated on a dry-weight basis.

Available P (AP) concentrations were determined using a Bray-1 extracting solution and the molybdenum-blue method (Kuo, 1996). Total P (TP) concentrations were measured after perchloric/nitric acid digestion using inductively coupled plasma emission spectrometry (ICP-ES) (Hossner, 1996). Total C and N were determined after dry combustion using Carbon-Nitrogen analyzer. Also, selected element (Pb, Al, Cu, As, Cd, Cr, Ni) concentrations in the soil and ryegrass were determined after digestion with $HClO_4/HNO_3$ using inductively coupled plasma emission spectrometry (ICP-ES) (Hossner, 1996). In addition, Hg was measured after microwave digestion using cold vapor atomic fluorescence spectroscopy (CVAFS). Soil pH was determined by a glass electrode of a 1:5 ratio of soil to the water mixture. Soil bulk density was determined by the ring-knife method. Cation exchange capacity and exchangeable Ca were measured using ICP-AES after ammonium acetate extraction (Ross and Ketterings, 2010).

Bioassay was conducted based on plant biomass and P uptake. Ecological risk assessment was focused on heavy metal accumulation in soil and plant.

2.3 Statistical analyses

Means and standard errors were given for measured data (*n*=3). Data were subjected to analysis of variance (ANOVA) using the SPSS 17.0 statistics program to determine significance among treatment, and all test statistics were evaluated at *P*=0.05. When the analysis generated a significant F value (*P*<0.05) for treatments, means were compared by the Tukey honestly significant difference test.

3 Results

3.1 Soil and FGD gypsum characterization

The Wooster silt loam soil had lower clay and total C (i.e., organic C) than the Hoytville clay loam soil (Table 1). The clay loam soil, as a result, also had 70.7% and 50.0% TP and AP greater than silt loam soil, respectively. The CEC in clay loam soil was also about three times greater than that in silt loam soil.

Analysis of the FGDG ($CaSO_4 \cdot 2H_2O$) material used in this study indicated the concentrations of S and Ca were 198 and 159 g kg^{-1}, respectively. Flue gas desulfurization gypsum contained lower concentrations of trace elements than were observed for the soil background values and were also below the standard limits of Standards for the use or disposal of sewage biosolids (US EPA, 1993) (Table 2).

3.2 DP in runoff and leachate

3.2.1 The effects on volumes

Based on our designed rainfall events for the Control treatment, silt loam soil produced 77.90% runoff and 22.10% leachate, clay loam soil 86.53% runoff and 13.47% leachate in volume. For runoff, there were no significant differences (*P*<0.05) in runoff volumes between the two contrasting soils and application methods. However, runoff volumes were reduced by FGDG, up to 11% at FGDG 15 g kg^{-1} in both soils.

For leachate, volumes were increased with FGDG, up to 28.82%−54.33% at FGDG 15 g kg^{-1}, leachate from silt loam soil being 1.6−1.8 times greater than clay loam soil (*P*< 0.05). Mix (tillage) treatments seemed to enhance the percolation process especially in clay loam soil. There were no significant differences in leachate volumes between application methods of FGDG.

Total volumes of runoff and leachate between treatments were almost the same, while total volume of silt loam soil was a little higher than that of clay loam soil without significant difference statistically (Table 3).

3.2.2 The effects on DP

Concentration distributions of DP in runoff and leachate were very different in the two soils. At the control treatment, silt loam soil lost 10.98% DP from runoff and 89.02% DP from leachate, while clay loam soil lost 47.89% DP from runoff and 52.11% DP from leachate.

For runoff, the concentrations of DP from clay loam soil were an order of magnitude more than DP from silt loam soil in all treatments. DP was reduced up to 33% in silt loam soil and 43% in clay loam soil at treatment concentration of FGDG 15 g kg^{-1}.

For leachate, the concentrations of DP from both soils were on the same order of magnitude comparing to runoff. Stronger interaction between FGDG and mix treatment was observed in treatment of GGM (FGDG 15 g kg^{-1} and Mix-applied), DP reducing up to 51% in silt loam soil and 43% in clay loam soil (Table 3).

Table 3 Cumulative flow volumes, concentrations of dissolved P (mg L^{-1}), and cumulative dissolved P loss amounts (mg) $^{\alpha}$ from combined runoff and leachate after rainfall from two Ohio soils treated with FGDG at different application concentrations and by different application methods $^{\beta}$.

Soil	Sample type	Volume (L)				
		0	GS	GM	GGS	GGM
Silt loam soil	Runoff	8.07±0.36a	7.81±0.36a	7.83±0.27a	7.27±0.2b	7.33±0.29b
Clay loam soil		8.16±0.07a	8.08±0.23a	8.12±0.10a	7.71±0.24b	7.27±0.22b
Silt loam soil	Leachate	2.29±0.01b A $^{\gamma}$	2.23±0.25b	2.36±0.02b	2.95±0.14a	2.91±0.08a
Clay loam soil		1.27±0.10b B	1.22±0.14b	1.41±0.11b	1.82±0.22a	1.96±0.23a
Silt loam soil	Total	10.4±0.32 A	10.0±0.19	10.2±0.28	10.2±0.25	10.2±0.33
Clay loam soil		9.43±0.30 A	9.30±0.24	9.53±0.42	9.54±0.34	9.23±0.38
Soil	Sample type	Average dissolved P concentrations $^{\delta}$ (mg L^{-1})				
		0	GS	GM	GGS	GGM
Silt loam soil	Runoff	0.60±0.02a B	0.56±0.06a	0.49±0.06ab	0.47±0.01b	0.44±0.02b
Clay loam soil		2.24±0.22a A	1.90±0.12a	2.13±0.23a	1.27±0.16b	1.56±0.07b
Silt loam soil	Leachate	17.0±0.44a	14.6±0.55b	11.6±1.61c	10.4±0.42c	8.27±0.30d
Clay loam soil		15.6±1.14a	12.4±1.15b	11.0±1.08b	9.35±0.80c	8.91±0.64c
Soil	Sample type	Cumulative dissolved P amounts (mg)				
		0	GS	GM	GGS	GGM
Silt loam soil	Runoff	4.81±0.18a B	4.39±0.58a	3.84±0.41ab	3.43±0.09b	3.22±0.12b
Clay loam soil		18.2±1.39a A	15.3±0.75a	17.3±2.34a	9.80±1.89b	11.4±2.75b
Silt loam soil	Leachate	39.0±2.58a A	32.4±2.09b	27.2±3.56b	30.7±0.25b	24.1±2.71c
Clay loam soil		19.7±0.48a B	15.3±2.95b	15.4±1.08b	16.9±0.67b	17.4±1.33b
Silt loam soil	Total	43.8±2.67a	36.7±2.30b	31.1±3.75bc	34.2±0.23b	27.3±2.80c
Clay loam soil		38.0±1.47a	30.6±2.68b	32.7±3.35b	26.8±1.25c	28.8±1.90bc

$^{\alpha}$ Values after the treatment means are means and standard errors within a row followed by the same letter or no letters are not significantly different (P <0.05) by Tukey's honest least significant difference test. $^{\beta}$ Treatments are 0 (no FGDG applied to soil), GS and GM (FGDG applied to the soil surface or mixed into the soil at the 1.5 g kg^{-1} soil rate, respectively) and GGS and GGM (FGDG applied to the soil surface or mixed into the soil at the 15 g kg^{-1} soil rate, respectively). $^{\gamma}$ Values after the soil means are means and standard errors within a column followed by the same or no capital letter in a column for each sample type are not significantly different (P <0.05) by Tukey's honest least significant difference test. $^{\delta}$ Average dissolved P concentration (mg L^{-1}) is calculated by the total amount of dissolved P accumulated divided by the total amount of water accumulated in runoff and leachate.

3.3 P in soil and ryegrass

Table 4 shows the changes in the soil pH, soil TP concentrations, and P uptake and biomass yield of ryegrass on the two agricultural soils between treatments. Soil pHs decreased at higher FGDG amounts comparing to other treatments, lower by 0.14 from pH 6.57 down to 6.43 in silt loam soil and by 0.20 from pH 6.26 down to 6.04 in clay loam soil at FGDG 15 g kg^{-1}. Application methods did not make any significant contribution to soil pH changes.

Soil TP contents in both soils were doubled after the P amendment at the beginning of the experiment (Tables 1 and 4). Compared with the Control (FGDG 0 g kg^{-1}), soil TP loss via runoff and leachate in clay loam was occurred at the treatment of FGDG 15 g kg^{-1}, while soil TP loss in silt loam soil did not clearly show at a level of g kg^{-1}, but about 30%−38% of DP loss from runoff and leachate at a level of mg kg^{-1} (Table 3).

Plant bioassay showed that biomass yield was not affected by FGDG, but biomass P was (Table 4). Biomass P of ryegrass was increased in all treatments-except in both soils. In this study surface application (no-tillage) did not bring significant benefits to biomass yield and P uptake, while mixing-applied (tillage) treatments improved the growing environment, especially at higher FGDG concentration.

4 Discussion

4.1 Controlling DP loss via runoff and leachate

4.1.1 DP loss via runoff and leachate

Runoff and leachate volumes in both soils was found by addition of FGDG. Runoff volume was reduced up to 11% and leachate volume increased up to 26%−35% at FGDG 15 g kg^{-1}. In addition, FGDG decreased runoff at lower DP

Table 4 The pH and total P (TP) in soil and biomass yield and biomass P in ryegrass as affected by different application rates and application methods of FGDG [α].

Soil type	Sample type	FGDG Treatment [β]				
		0	GS	GM	GGS	GGM
Silt loam soil	Soil pH	6.57±0.10 A	6.58±0.10	6.54±0.12	6.43±0.13	6.44±0.10
Clay loam soil		6.26±0.06 B	6.23±0.06	6.20±0.04	6.02±0.20	6.05±0.19
Silt loam soil	Soil TP(g kg^{-1})	0.95±0.01 A[γ]	0.97±0.02	0.96±0.02	0.96±0.02	0.96±0.03
Clay loam soil		1.36±0.07 B	1.37±0.05	1.36±0.05	1.34±0.06	1.34±0.04
Silt loam soil	Biomass Yield (g m^{-2}) [δ]	410±12.1 A	416±18.0	417±8.30	402±20.0	432±10.0
Clay loam soil		456±11.0 B	463±1.10	473±21.0	453±15.0	478±12.1
Silt loam soil	Biomass P (g kg^{-1}) [ε]	5.02±0.13c A	5.41±0.03b	5.53±0.08b	5.40±0.06b	5.78±0.13a
Clay loam soil		6.05±0.11b B	6.63±0.07a	6.68±0.07a	6.45±0.14a	6.71±0.11a

[α] Treatments are 0 (no FGDG applied to soil), GS and GM (FGDG applied to the soil surface or mixed into the soil at the 1.5 g kg^{-1} soil rate, respectively) and GGS and GGM (FGDG applied to the soil surface or mixed into the soil at the 15 g kg^{-1} soil rate, respectively). [β] Values after the treatment means are standard errors and means within a row followed by the same letter or no letters are not significantly different (P <0.05) by Tukey's honest least significant difference test. [γ] Values after the soil means are standard errors and means within a column followed by the same or no capital letter in a column for each sample type are not significantly different (P <0.05) by Tukey's honest least significant difference test. [δ] Ryegrass dry matter yield. [ε] 'Biomass P' is P concentration of ryegrass uptake.

concentration, increased leachate with lower DP concentration (Table 3). The results suggest that, in practice, it is beneficial to employ a balanced strategy of FGDG application. For instance, in clay loam soil DP concentration in runoff was much higher than that from silt loam soil, so in practice， control runoff from clay loam soil should be considered.

DP loss in leachate is another interesting issue for discussion. DP loss was more prominent from silt loam soil than from clay loam soil, because the clay loam soil had a larger P retention capacity (Chardon and Schoumans, 2017). The contrasting textures between the two soils affected the specific pathways by which DP was lost. Leachate was more dominant pathway for DP loss from silt loam soil than from clay loam soil.

Although FGDG increased leachate volume, FGDG application decreased DP loss via runoff and leachate. DP moves and diffuses slowly in soil, there was sufficient time for Ca^{2+} and DP to react. FGDG continuously provided dissolved Ca^{2+} to react with DP in the soil solution with a pH range of 5.5 to 7.0 to form calcium phosphate dihydrate ($CaHPO_4 \cdot H_2O$) or dicalcium phosphate ($CaHPO_4$), which are absorbed by the plant in the percolation process to reduce DP loss (Ryden and Syers, 1975; Recillasa et al., 2012).This was confirmed by our study. Biomass P of ryegrass was increased in all FGDG treatments in both soils (Table 4). Some studies have also shown that FGDG also can reduce phosphorus loss from an alkaline soil. In alkaline soils (pH ＞ 7.0), P and Ca^{2+} form insoluble calcium-phosphorus complex (Lopez and Garcia, 1997; Murphy et al., 2010; Watts and Torbert, 2016; He, 2016). This results in reduced amounts of DP in runoff and leachate from agricultural soils to surrounding water bodies (Li et al., 2018).

The higher concentration of FGDG application (i.e., 15 g kg^{-1}) was more effective at decreasing leachate DP concentration and total DP loss than the 1.5 g kg^{-1} application concentration. Most water leaving fields in northwest Ohio occurs via tile drainage (i.e., leachate) (Smith, 2015). FGDG was shown to be an effective treatment for controlling DP loss (King et al., 2016).

The FGDG treatment may be especially beneficial for controlling DP loss from soils managed by no-tillage farming practices (when the FGDG is surface-applied). That is because surface-applied P fertilizer and P in plant residues in no-tillage system are largely retained at the soil surface and in the upper soil layer. Mixing the FGDG into the soil will also decrease DP losses from tilled soils. Increasing P absorption by ryegrass can reduce the export of DP via runoff and leachate from a field and protect receiving bodies of water from the harmful effects of the DP.

4.1.2 DP loss via application methods

FGDG mixing into silt loam soil (tillage) was more effective in reducing DP loss in silt loam soil because DP loss in this soil was mainly via leachate. Mixing FGDG into the soil provides more interaction with soil DP and is intercepted and absorbed by the plant's roots before it leaches off the soil. Surface-applied FGDG to soil (no tillage) could postpone or limit clay dispersion and surface seal formation, maintained porosity, and thus increased infiltration (Yu et al., 2003). But in our study, surface-applied (no tillage) only showed its advantage in controlling runoff DP (Fig. 2 B).

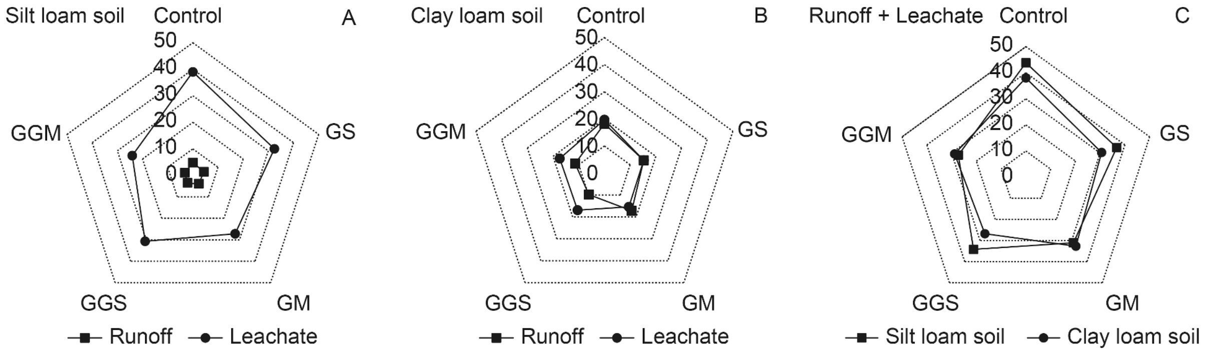

Fig. 2 The evaluation results of single treatment applied on two agricultural soils by cumulative DP. Treatments are 0 (no FGDG applied to soil), GS and GM (FGDG applied to the soil surface or mixed into the soil at the 1.5 g kg^{-1} soil rate, respectively) and GGS and GGM (FGDG applied to the soil surface or mixed into the soil at the 15 g kg^{-1} soil rate, respectively).

4.1.3 The evaluation results by cumulative DP

Table 3 gives cumulative DPs from runoffs and leachates of two soils in all treatments, and the evaluation results of single treatment applied on two agricultural soils by cumulative DP are expressed through a radar graph in Figure 2. For silt loam soil, DP loss from leachate was much higher than that from runoff. Mixing higher concentration of FGDG (in our experiment FGDG 15 g kg^{-1}) into soil was able to control DP loss effectively (Fig. 2A). Leachate DP reduction efficiency was raised to 20% at FGDG 1.5 g kg^{-1}, and 27% at FGDG 15 g kg^{-1} by mixing-applied (tillage). For clay loam soil, no treatment had obvious advantage in controlling DP loss from soils, although higher FGDG concentrations had some effects on DP loss from runoff (Fig. 2B).

As mentioned in section 4.1.1, FGDG decreased runoff and increased leachate at same time. Overall, mixed-applied performed better in controlling DP loss from silt loam soil, while surface-applied showed its advantage in controlling DP loss from clay loam soil (Fig. 2C).

4.2 Other impacts

4.2.1 Biomass yield and biomass P

In our study, biomass yield and biomass P of ryegrass were increased in all treatments except GGS treatments in both soils (Table 4). The Ca and S in FGDG can act as a plant nutrient and soil conditioner for agricultural production. A higher concentration of FGDG releases more Ca^{2+} into the soil solution to react with more DP to form a calcium-phosphorus complex, which is more easily absorbed by ryegrass. But no effects of surface-applied FGDG at 15 g kg^{-1} (treatment GGS) were detected for plant yields and P uptake. Surface-applied (no tillage) did not show advantage to plants during the experimental period; as has been the case for other short-term studies which have yielded negative results (Watts and Dick, 2014). This is a common phenomenon and the benefits will not always be obtained in the first, or even second, year after FGDG application.

Although our experiment obtained good results in terms of plant growth, time is required for the benefits of FGDG application to be realized (EPRI, 2011−2014).

4.2.2 Trace elements in soil and ryegrass

The content of heavy metals in both FGDG self and experimental soil is the most concern for ecological safety. Table 2 shows the comparison of some trace elements among FGDG, soils and US EPA standard, which indicated that the concentrations of selected trace metals in FGDG was lower than those in soils and EPA Standard. Table 5 gives selected trace elements in soil and ryegrass before and after FGDG treatments. As shown in the table, FGDG application at the highest level of 15 g kg^{-1} caused a little fluctuation of heavy metal contents which were below soil backgrounds and EPA's level of concern, and did not cause any accumulations in the soils.

In our experiment plant enrichment of heavy metals did not occur in ryegrass growth (Table 5). Some studies have also showed that FGDG did not cause an accumulation of trace elements in the edible parts of various crop products (Wang et al., 2015, 2018). FGDG did not pose any adverse threat to soil, plant, or environment.

5 Conclusion

Flue gas desulfurization gypsum (FGDG) can effectively control DP loss via runoff and leachate, as observed in the two contrasting soils, silt loam soil (Wooster) and clay loam soil (Hoytville), in Ohio, USA.

Our greenhouse study showed that FGDG decreased runoff volume and increased leachate volume in both soils. FGDG reduced runoff DP loss by 33% and leachate DP loss by 50% in silt loam soil, and runoff DP loss by 37% and leachate DP loss by 43% in clay loam soil, at the treatment of 15 g kg^{-1} FGDG rate and mixed-applied (tillage).

Overall, mixing-applied method (tillage) provided strong interaction with higher FGDG, enhancing DP loss reduction in both soils, especially in leachate from silt loam soil.

Table 5 Trace elements concentrations in soil and ryegrass.

Soil	Treatment [α]	Soil (mg kg^{-1})						
		Pb	Al	Cu	As	Cd	Cr	Ni
Silt loam soil	0	16.9±0.59	17.2±0.11	7.09±0.31	8.08±0.25	1.90±0.25	25.2±0.95	19.1±0.09
	FGDG	17.4±0.70	17.6±0.32	7.29±0.49	8.51±0.63	1.80±0.07	25.6±0.73	19.6±0.37
Clay loam soil	0	14.4±0.55	30.5±1.23	25.8±1.19	3.53±0.57	2.72±0.15	40.7±1.70	30.3±0.62
	FGDG	14.1±0.73	30.2±0.68	25.6±0.60	3.57±0.49	2.63±0.09	40.3±0.96	30.3±0.71
Soil	Treatment [α]	Ryegrass (mg kg^{-1}) [β]						
		Pb	Al	Cu	As	Cd	Cr	Ni
Silt loam soil	0	<0.863 [γ]	0.34±0.13	4.93±0.27	<1.931	0.10±0.01	0.98±0.26	1.17±0.08
	FGDG	<0.863	0.51±0.31	4.95±0.46	<1.931	0.13±0.03	1.17±0.10	1.38±0.25
Clay loam soil	0	<0.863	2.36±0.49	8.31±0.49	<1.931	0.36±0.04	2.89±0.29	3.91±0.35
	FGDG	<0.863	2.06±0.49	7.13±0.60	<1.931	0.39±0.09	2.64±0.18	3.62±0.12

[α] Treatments are 0 (no FGDG applied to soil) and with FGDG applied to the soil. FGDG means were calculated by averaging the means of the 15 g kg^{-1} FGDG application rates. [β] Values after the treatment means are standard errors and means within a row followed by the same letter or no letters are not significantly different (P <0.05) by Tukey's honest least significant difference test. [γ] Means preceded by a less than sign (<) represent concentrations below the detection limit, which is given by the number in the table.

Surface-applied (no-tillage) showed its advantage in controlling DP loss from clay loam soil, but did not bring significant benefits to plants during the experimental period.

FGDG did not pose any adverse threat to soil pH and TP, plant biomass and P uptake, and ecological safety relating to heavy metals.

Applying FGDG to soil has great potential in reducing DP losses to the environment via runoff and leachate from areas receiving excessive P fertilizer or manure as a nutrient source. In practice, it is necessary to consider FGDG concentrations, methods of application, DP sources (runoff or leachate), and their optimization combination to get maximized benefits of FGDG application. Ecological risk assessment is also needed for the effects of some trace elements which may accumulate in soils and plants, even at low concentrations of FGDG.

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