VEGETATION AND OVERBURDEN COVER ON PHOSPHOGYPSUM: EFFECTS ON RADON EMISSION, RUNOFF WATER QUALITY, AND PLANT UPTAKE OF FLUORIDE AND RADIUM [1]

by

Steven G. Richardson [2]

Abstract. Phosphogypsum is a byproduct of phosphate fertilizer production, and more than 700 million metric tons have accumulated on 2,500 ha in Florida. Field research was conducted to compare the benefits of capping phosphogypsum with overburden (up to 15 cm in depth) from mined sites versus treatment of the phosphogypsum with minimal amendments. After four growing seasons, vegetation cover was excellent (no bare ground) on plots amended with dolomitic limestone or capped with overburden. However, more species became established with an overburden cap. Fluoride uptake by bermudagrass (*Cynodon dactylon*) was high when grown directly on phosphogypsum (895 mg kg^{-1} in leaf tissue) and was reduced slightly by a 15 cm overburden cap (670 mg kg^{-1}). Unexpectedly, radium (^{226}Ra) uptake in bermudagrass grown directly on phosphogypsum (0.6 pCi g^{-1}) was less than when grown on the overburden cap (1.8 pCi g^{-1}). The presence of grass cut the radon (^{222}Rn) efflux from phosphogypsum in half (from 24 pCi m^{-2} s^{-1} to 11 pCi m^{-2} s^{-1}), while 15 cm of overburden, in addition to grass cover, halved it again (down to 5 pCi m^{-2} s^{-1}). Vegetation cover on phosphogypsum resulted in a 30-fold decrease in electrical conductivity and a 5-fold decrease in the fluoride concentration of surface runoff water. Runoff water quality from vegetated plots was equally good with or without a 15 cm overburden cap on top of the phosphogypsum.

Introduction

Phosphogypsum is a by-product of phosphoric acid production. For each kg of P_2O_5 produced, approximately five kg of phosphogypsum must be stockpiled or disposed. To date, more than 700 million metric tons have accumulated in Florida on 2500 ha, and phosphogypsum is continuing to accumulate at the rate of about 30 million metric tons per year. After phosphate rock has been reacted with sulfuric acid to produce phosphoric acid, the precipitated gypsum is removed on filters and is pumped in slurry form to an impoundment where it is allowed to settle. As the gypsum accumulates, a small dragline removes some of the gypsum for raising the height of the dikes. By this process the gypsum settling impoundment, or "stack" as it is often called, increases in elevation. As a stack grows in height (up to 65 m) the area of the settling impoundment decreases, until a point is reached where the pond capacity becomes too small and the pumping height requires too much energy. When phosphogypsum stacks reach the end of their useful lives, they must be closed and reclaimed. Reclamation includes a cover of vegetation to improve aesthetic qualities, to reduce erosion and improve the quality of surface runoff water.

There are several problems associated with establishing vegetative cover on phosphogypsum, including (1) the residual acidity, (2) nutrient deficiencies and low nutrient-holding capacity, and (3) the tendency for caking and crust formation. In addition, there are other potential environmental problems. Phosphogypsum has elevated levels of fluoride and radionuclides, such as Radium-226 (^{226}Ra). In Florida there is concern about the quality of runoff water and leachate from these waste piles.

The primary problem for vegetation establishment is the acidity of the phosphogypsum. May and Sweeney (1980) observed pH values usually in the range of 2 to 3 in numerous samples, although samples from older, more weathered stacks had slightly higher pH values near 4. Bromwell and Carrier, Inc. (1989) reported a pH of 5.1 in a sample of phosphogypsum from an older, inactive stack. Phosphogypsum is comprised primarily of the slightly soluble neutral salt, calcium sulfate dihydrate. The

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[2]Steven G. Richardson is Director of Reclamation Research at the Florida Institute of Phosphate Research, 1855 West Main Street, Bartow, Florida 33830

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acidity is due mainly to residual phosphoric and sulfuric acids, although hydrofluoric and fluosilicic acids are also formed during processing (Smith and Wrench 1984, Wrench 1987).

Laboratory, greenhouse and field studies, plus field observations, have indicated that plants could grow on phosphogypsum if the pH was raised to 4.0 or greater and if N, K, and Mg nutrients were supplied (Richardson 1993). Dolomitic limestone (also an Mg source) and phosphatic clay (also a K and Mg source) were effective neutralizing agents, while leaching with water was also an effective mechanism for reducing acidity. Leaching with rainwater is probably responsible for the higher pH values (4.7 to 5.3) found at the surface of older, inactive phosphogypsum stacks compared to pH values near 2 or 3 at active, acid-laced phosphogypsum stacks (Richardson et al. 1995).

Fluoride concentrations in phosphogypsum produced from Florida phosphate rock often range from 4 to 7 g kg^{-1} (Rechcigl et al. 1992, Carter and Scheiner 1992, Oates and Caldwell 1985). Radium-226 commonly ranges from 20 to 30 pCi g^{-1} in central Florida phosphogypsum and from 9 to 15 in northern Florida phosphogypsum (Roessler et al. 1979, Mays and Mortvedt 1986, May and Sweeney 1980). The concern with fluoride is the possible development of fluorosis (a bone and tooth disorder) in grazing animals from consuming plants with high concentrations of fluoride. Radium-226 may also be absorbed by plants and be ingested by animals. In addition, ^{226}Ra gives rise to Radon-222 (^{222}Rn) gas, which may diffuse out of the phosphogypsum into the atmosphere. There is also concern about the quality of surface runoff waters and leachate from phosphogypsum stacks.

This study is part of a research program with the primary objective of developing effective, but low cost, methods of establishing vegetation on phosphogypsum stacks when they are closed. One approach to coping with the chemical and physical properties of phosphogypsum is to cover it with soil or overburden. This approach has been accepted by the Florida Department of Environmental Protection (FDEP), but the costs of the earthmoving operations are high and there are environmental impacts on the soil borrow areas. However, FDEP regulations (Florida Administrative Code [FAC] 17-673) do allow for the possibility of using amended phosphogypsum as a final cover for phosphogypsum stack closure, if the amended phosphogypsum can can be shown to effectively sustain vegetation and meet performance standards. This paper examines vegetation establishment directly on phosphogypsum compared to the use of modest depths (7.5 to 15 cm) of overburden cover. It also compares the effects of establishing vegetation with or without an overburden cap on plant uptake of fluoride and ^{226}Ra, on the efflux of ^{222}Rn gas, and on the quality of runoff water.

Materials and Methods

A study was designed to compare the effects of 0, 7.5 and 15 cm of overburden cover on vegetation establishment on a 3.5:1 slope at the IMC-Agrico (IMCA) New Wales phosphogypsum stack in central Florida. The three overburden depths were replicated three times on plots measuring 30 m downslope and 7.6 m along the contour. A site was cleared on June 20, 1991, on the east side of the phosphogypsum stack. An existing bench, located immediately above the experimental site was widened to allow truck access and sloped to intercept surface runoff and route it away from the study plots. Dolomitic limestone was applied at the rate of 1.5 metric ton/ha on plots that did not receive any overburden cover (0 overburden depth) and the site was disked. Overburden application began June 24 and was completed on June 25, 1991. Overburden from a mined site south of the gypsum stack (site of the new stack expansion) was trucked and dumped on the bench and upper slope of the appropriate study plots. A D-3 dozer spread the overburden downslope over the phosphogypsum, guided by a series of survey stakes marked with the proper depths (0, 7.5 or 15 cm). On the afternoon of June 25, 1991, the site was lightly scarified with a disk harrow and the overburden depth study plots were broadcast seeded with 29 kg common bermudagrass (*Cynodon dactylon*) and 58 kg browntop millet (*Brachiaria ramosa*) per ha. Fertilizer was broadcast at the per ha rates of 78 kg N, 20 kg P_2O_5, 39 kg K_2O, 10 kg Mg, 2.4 kg Fe, 1.5 kg Mn, 0.5 kg Zn, 0.25 kg Cu, and 0.15 kg B. Bahiagrass hay mulch was blown on the overburden depth plots at 4 metric tons ha^{-1}, and the hay was crimped in with a cultipacker roller and the dozer tracks. The plots were fertilized again in June 1992 and June 1993 with 56 kg N, 28 kg K_2O and 14 kg Mg per ha. In the spring of 1994, 56 kg ha^{-1} of N as isobutyldienediurea (IBDU) was applied.

Overburden from the 15 cm depth treatment, phosphogypsum from the plots without overburden,

Table 1. Chemical analyses of overburden and phosphogypsum at the IMCA site [1]

	pH	EC	Melich I Extractable Nutrients (mg kg^{-1})				
			Mg	K	P	Mn	Zn
Overburden (Grassed)	5.0	2.01	45.9	32.8	2193	2.43	2.37
Phosphogypsum (Grassed) [2]	5.3	2.36	97.5	7.3	- [3]	0.33	0.64
Phosphogypsum (Bare)	4.5	2.35	4.0	4.8	- [3]	0.15	0.89

[1] Mean values of three replicates
[2] Dolomitic limestone applied at rate of 1.5 metric ton per ha
[3] Data not available

and phosphogypsum from a barren, untreated area were analyzed for pH, electrical conductivity, and Melich I extractable Mg, K, P, Mn, and Zn. The overburden was higher in pH, Mg, K, Mn, and Zn than the bare phosphogypsum, but the dolomite-amended phosphogypsum was higher in pH and Mg than the overburden (Table 1).

Percent plant cover was determined by the line-point method (three transects of 30 points each per plot) near the end of each growing season. Samples of bermudagrass tissue were collected from the overburden depth plots at the IMC site on October 24, 1991, and were analyzed for ^{226}Ra activity, and for concentrations of F, selected heavy metals and nutrients.

Radon-222 fluxes from the ground surface were measured in February, March and May 1992, with the aid of 25cm diameter activated charcoal canisters (Hartley and Freeman 1985) placed for 24 hours on the grass-covered overburden depth plots and a barren (no vegetation or overburden cover) phosphogypsum "control" plot. To assure a good canister to soil seal, the grass was clipped to a height of approximately 4 cm, a canister was placed on the ground, and bermudagrass stolons (runners) and "soil" were cut with a knife around the perimeter of the canister. The canister was then pressed downward and twisted to attain good ground contact. On the plots barren of vegetation, canisters were pressed onto the surface and phosphogypsum was packed around the outside perimeter of the canisters to help assure a good seal. The ground surface beneath the charcoal-laden portion of each canister was not disturbed. Following the 24-h period in the field, charcoal from each canister was removed, placed in a sealed container, and sent to the Florida Health and Rehabilitative Services laboratory in Orlando for radon flux determination.

Runoff was collected during the summer of 1994 from bare phosphogypsum, from phosphogypsum with a bermudagrass cover (planted June 1991), and from plots with 15 cm of overburden cover and bermudagrass. There were three replicate plots of each surface treatment. Water was collected in troughs made from 30 cm diameter PVC pipe cut in half, and the water then flowed through 5 cm diameter plastic pipes into 190 L plastic barrels. The troughs, pipes and barrels were rinsed with deionized water periodically, and runoff water samples were collected the morning after the next storm following the rinsing. The troughs were 2.9 m long, and the watershed above the troughs averaged 17.2 m in length (50 m^2 in area). A bermed terrace (roadway) above the test plots diverted runoff water from the rest of the gypstack away from our plots. Volume of runoff was determined after each storm event and samples were analyzed for electrical conductivity and fluoride (ion selective fluoride electrode).

Results

Plant Cover and Growth. Full details on the effects of the overburden cap on plant cover by species is contained in Richardson et al. (1995) and Richardson (1994). By the end of the 1992 growing season, grass yields were equal with or without the overburden cap (Table 2), but because of the greater

Table 2. Plant yields (kg dry weight per ha) in September 1992 as affected by depth of overburden cover at the IMCA phosphogypsum stack.(1)

	Overburden Depth (cm)		
	0	7.5	15
Grasses	5098 a	5081 a	4808 a
Broadleaved plants	199 b	1476 a	1265 a
Total	5297 b	6557 a	6073 a

(1) Values within rows followed by the same letter are not significantly different at the 0.05 level. Data are means of three replicates.

Table 3. Percent cover as affected by overburden depth at the IMCA phosphogypsum stack in 1993. [1]

	Overburden Depth (cm)		
	0	7.5	15
Bermudagrass	95.8 a	97.9 a	90.6 a
Broomsedge	14.9 a	0.3 b	0.0 b
Hairy Indigo	1.0 c	51.7 b	72.6 a
Dog Fennel	1.0 a	1.4 a	1.0 a
Saltbush	1.7 a	0.7 a	3.8 a
Litter	1.0 b	2.1 ab	9.4 a
Bare Ground	0.0 a	0.0 a	0.0 a

[1] Bermudagrass seeded 1991. Data are mean values of three replicates. Values within rows followed by the same letter are not significant at the 0.05 level. The sums of columns may total greater than 100% because of overlapping vegetation.

invasion of the annual legume, hairy indigo (*Indigofera hirsuta*), on the overburden cap than on the phosphogypsum, total plant yield was greater with the overburden cap . By the end of the 1993 growing season, plant cover was excellent (there was no bare ground) (Table 3). Bermudagrass provided most of the effective ground cover, but there was considerable overlapping cover from the more erect and taller hairy indigo. Dog fennel (*Eupatorium capillifolium*) and saltbush (*Baccharis halimifolia*) commonly invaded the plots but were controlled with an herbicide mixture of 2,4-D plus dicamba in late June to early July. In the spring of 1994, dog fennel, saltbush and hairy indigo were pulled manually from the plots to minimize the potential competitive effects of these taller species on the bermudagrass cover during the runoff study.

Plant Mineral Content. The mineral analyses of bermudagrass tissue, as affected by overburden cover on the IMCA phosphogypsum stack, are shown in Table 4. Overburden cover had no significant effects on the concentrations of Ca, Mg, K, Fe, Cu, B or Al in bermudagrass tissue. Tissue phosphorous was

Table 4. Concentrations of various elements in bermudagrass tissue as affected by depth of overburden cover at the IMCA phosphogypsum stack [1]

	Overburden Depth (cm)		
	0 [2]	7.5	15
		-----------g kg^{-1}---------	
Ca	4.58 a	4.65 a	4.74 a
Mg	2.12 a	2.02 a	2.17 a
K	8.59 a	8.86 a	10.06 a
P	3.52 a	2.50 b	2.92 b
		------------mg kg^{-1}---------	
Mn	130.2 b	215.6 a	228.5 a
Fe	60.0 a	76.2 a	68.0 a
Zn	13.8 b	36.5 a	36.2 a
Cu	3.5 a	3.8 a	7.2 a
B	3.2 a	3.0 a	2.8 a
Al	35.0 a	58.3 a	45.0 a
Cd	0.0 b	2.7 a	3.2 a
Ni	0.0 a	0.0 a	0.5 a
F	895.0 a	835.0 a	670.0 b

(1) Mean values of three replicates. Values within rows followed by the same letter are not significantly different at the 0.05 level.

(2) Phosphogypsum without overburden cover was treated with 1.5 metric tons dolomite per ha.

higher without overburden cover, while manganese, zinc and cadmium were higher with overburden cover. There were no significant differences in the effects of the 7.5 cm or the 15 cm overburden depths on tissue mineral content, except in the case of fluoride, which was lower when plants were grown with 15 cm of overburden cover. The fluoride values were nevertheless very high for all treatments and could lead to fluorosis if the bermudagrass were consumed in large quantities by livestock (Alcordo and Rechcigl 1995).

Radioactivity. The activity of ^{226}Ra in bermudagrass tissue actually increased with increasing depth of overburden cover over phosphogypsum (Table 5), and linear regression analysis produced an r^2 value of 0.92.

The presence of bermudagrass drastically reduced the ^{222}Rn flux from phosphogypsum, based on the charcoal canister technique employed in this study (Table 5). Radon flux from phosphogypsum was reduced 50 percent by the bermudagrass cover, while 15 cm of overburden plus bermudagrass reduced the radon flux to 25 percent of that from bare phosphogypsum. The mean value of radon flux measurements from the grassed plots with the various depths of overburden cover differed significantly only in May. Radon flux measurements in February and March indicated a decrease in radon flux with increasing overburden depth, however the differences were not significant. The correlation coefficients (r) for radon flux versus overburden depth were -0.74 in February (r^2 = 0.54), -0.66 in March (r^2 = 0.44) and -0.82 in May (r^2 = 0.66). The correlations were significant at the 0.05 level in February and May, and nearly significant (0.052 level) in March.

Runoff Water Quality. The electrical conductivity and fluoride data indicate a marked improvement in runoff water quality with a vegetation cover (Table 6). Runoff volume was reduced when there was a cover of vegetation. There were no statistically significant differences between the vegetated plots with or without the 15 cm of overburden cover.

Discussion

An overburden cap of up to 15 cm in depth was not necessary for the establishment of an excellent cover of bermudagrass on phosphogypsum in this study. Good plant growth on phosphogypsum in the greenhouse and at three field sites was observed by Richardson et al. (1995) when the pH was 4.8 to 5.3; when adequate nitrogen, potassium and magnesium were supplied; and when adapted plant species such as bermudagrass or switchgrass (*Panicum virgatum*) were planted. However, the overburden cap did modify the edaphic environment (e.g. the overburden appeared to be a source of nutrients such as potassium, manganese and zinc [see Table 1]), and it did increase the number of plant species that became established on the phosphogypsum stacks. For example, hairy indigo

Table 5. Radium levels in bermudagrass tissue and radon flux from the ground surface at the IMC phosphogypsum stack as affected by overburden cover and by plant cover.[1]

	Control (no plants)	Overburden Depth (cm) (with plant cover) 0	7.5	15
^{226}Ra in plant tissue (pCi g^{-1})	--	0.6 c	1.1 b	1.8 a
^{222}Rn flux (pCi m^{-2} s^{-1})				
Feb. 1992	18.3 a	5.6 b	3.5 b	1.1 b
Mar. 1992	32.4 a	15.1 b	12.7 b	8.3 b
May 1992	22.8 a	18.5 b	8.1 bc	6.9 c
Average	24.5	11.4	8.1	5.4

[1] Mean values of three replicates. Values within rows followed by the same letter are not significantly different at the 0.05 level.

Table 6. Effects of vegetation cover and depth of overburden cap on volume, electrical conductivity and fluoride concentration of stormwater runoff from a phosphogypsum stack.[1]

	Bare	Overburden Depth (cm) (with plant cover) 0	15
Runoff Volume (L)	51 b	13 a	13 a
Electrical Conductivity (umho cm^{-1})	1621 b	53 a	46 a
Fluoride (ppm)	5.7 b	1.1 a	1.2 a

[1]Values are means of three replicates per sampling date and twelve sampling dates. Values within rows followed by the same letter are not significantly different at the 0.05 level.

did better on the overburden cap at the IMCA site, and at other phosphogypsum sites, overburden (or other amendments) enhanced weeping lovegrass (*Eragrostis curvula*) and bahiagrass (*Paspalum notatum*) establishment (Richardson et al. 1995). Thus an overburden cap or other amendments may improve species diversity, but they are not necessary for certain adapted plant species. Some species, such as broomsedge (*Andropogon virginicus*) may even do better on phosphogypsum than overburden, probably because of less plant competition on the phosphogypsum (see Table 3).

Fluoride uptake was high in bermudagrass growing on phosphogypsum, even with the overburden cap, although the 15 cm overburden cap did somewhat reduce the fluoride concentration of bermudagrass. However, the fluoride concentrations were many times the levels considered safe for grazing animals (Rechcigl et al. 1992, Church 1979). One solution is to exclude grazing animals from phosphogypsum stacks or make sure that phosphogypsum grown plants are only a small part of the diet. Application of a thicker cap of low fluoride soil material, combined with plant species that are less prone to accumulate fluoride, such as bushy beardgrass (*Andropogon glomeratus*) and bahigrass (*Paspalum notatum*) (Richardson et al. 1995), might decrease forage F concentrations to more acceptable levels. It may also be possible to reduce the solubility of fluoride by lime application.

Radium-226 in phosphogypsum or overburden was not measured at the IMCA study site, but typically in central Florida, phosphate overburden has much lower ^{226}Ra levels (average about 5 pCi g^{-1}) than phosphogypsum (average about 26 pCi g^{-1}) (Roessler et al. 1979 and 1980). Therefore it was surprising that the levels of ^{226}Ra in bermudagrass increased, rather than decreased, with increasing thickness of the overburden cap. It is possible that the ^{226}Ra in the overburden may have been in a more soluble form. Soil pH, soil calcium, and the chemical form of the radium compounds in the soil may all affect plant uptake of radium (Simon and Ibrahim 1988, Mortvedt 1992). The ^{226}Ra levels in bermudagrass grown on phosphogypsum in this study are similar to the ^{226}Ra levels in stargrass (*Cynodon nlemfuensis*), a relative of bermudagrass, grown in central Florida on waste phosphatic clay containing 22 pCi g^{-1} of ^{226}Ra (Mislevy et al. 1989, 1990). Bahiagrass grown on phosphatic clay contained less than one-third the radium level of the stargrass. This suggests that bahiagrass might take up less ^{226}Ra than bermudagrass when planted on overburden-capped phosphogypsum (but bahiagrass establishment was poor on uncapped phosphogypsum at another site [Richardson et al. 1995]).

Plants growing in ^{226}Ra-containing materials may have an effect on the release of radon to the atmosphere. Lewis and MacDonell (1990), for example, provided evidence that ^{222}Rn dissolved in soil water may be absorbed by plants and released to the atmosphere via transpiration. This mechanism of ^{222}Rn release would be most significant when a ^{226}Ra-containing material was covered by a low radioactivity soil cap and plant roots came into contact with the buried material. The data presented here indicate that the presence of plants growing directly on phosphogypsum will reduce the efflux of radon, and a thin (e.g. 15 cm thick) soil or overburden cap will further reduce the radon efflux.

Regulations of the Florida Department of Environmental Protection (FDEP) (Florida Administrative Code [FAC] 17-673.610[6][c]) allow the use of amended phosphogypsum as a final cover for phosphogypsum stack closure, if it will sustain vegetation and meet performance standards. In this study, phosphogypsum amended with dolomitic limestone at 1.5 metric ton ha^{-1}, which raised the pH to 5.3, was able to support vegetation cover comparable to that on overburden with a pH of 5. Other research has shown that the dolomitic limestone is not necessary if the pH of the phosphogypsum is near 5; if adequate nitrogen, potassium and magnesium fertilizers are supplied; and if adapted plant species such as bermudagrass are planted (Richardson et al. 1995). Runoff water quality (e.g. electrical conductivity and fluoride concentration) was equally good in this study from test plots with plants growing directly in phosphogypsum or in a 15 cm layer of overburden on top of the gypsum. This indicates that performance standards, with regard to vegetation cover, erosion control, and minimizing or eliminating contaminated runoff, should be accomplished equally well with either overburden or amended phosphogypsum as the surface dressing for sustaining vegetation cover.

A topic included in the FDEP regulations but not addressed in our research was the use of a low permiability barrier layer beneath the vegetated cover to retard or prevent infiltration of rainfall into the gypsum stack (with the goal of ultimately reducing the amount of acidic water seeping out of a closed phosphogypsum stack). The only phosphogypsum stack closed to date in Florida with a low permiability barrier layer on top is the Cargill (formerly Gardinier) stack adjacent to Tampa Bay (Morris and Garlanger 1994). A high density polyethylene (HDPE) geomembrane was applied only to the nearly-flat top of the stack, while the side slopes remained unlined. The entire surface of the gypsum stack was covered with mineral soil prior to planting vegetation. Our research indicates the soil cover on the side slopes is unnecessary and that amended phosphogypsum could also serve as the cover to support vegetation on the HDPE top liner.

The FDEP rules also allow for the use of compacted soil as a barrier layer on top of a phosphogypsum stack if permeability can be kept at or below 10^{-7} cm s^{-1} for stacks without bottom liners or 10^{-5} cm s^{-1} for stacks with bottom liners. Hird (1993) reported a permeability of approximately 10^{-6} cm s^{-1} for a naturally settled mixture of phosphatic clay and phosphogypsum in North Carolina. This suggests that a mixture of phosphatic clay and phosphogypsum could function as a top barrier layer, particularly if the mixture were compacted with mechanical pressure. This has not yet been demonstrated in Florida. Other Florida research (Richardson et al. 1995, Patel et al. 1995), plus studies in North Carolina (Hird 1993), clearly show that gypsum-clay mixtures can support excellent growth of vegetation. Future research should address the possibility of using alternative barrier layers that may be less costly yet may adequately control the infiltration of water and the production of leachate.

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