CHARACTERIZATION OF LEACHATE PRODUCED FROM SULPHUR-CONTAMINATED SOILS[1]

by

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Abstract. Reclaiming soils that are heavily contaminated with elemental sulphur is a problem specific to a small part of the oil and gas industry. Characterization of leachate produced from sulphur-contaminated soils will aid in the evaluation of potentially related environmental and health impacts. Leachate was collected at initiation and completion of a greenhouse growth trial from pots containing four sulphur levels (<0.1%, 4%, 9%, and 14% total sulphur) and four reclamation treatments(no treatment, limestone, limestone and manure, and manure). Leachate samples were analyzed for pH, electrical conductivity, nitrate-nitrogen, ammonium-nitrogen, sulphate-sulphur and phosphate-phosphorous. Selected subsamples were acidified and analyzed for soluble aluminum, iron and manganese. Leachate produced from unlimed, sulphur-contaminated soils were strongly acidic while the pH of leachate produced from limed, sulphur-contaminated soils was neutral. Electrical conductivity values were highest in leachate produced from unlimed, sulphur-contaminated soils. Leachate produced from unlimed sulphur-contaminated soils also contained elevated levels of nitrogen, phosphorous, and sulphur as well as elevated levels of soluble aluminum, iron and manganese. Leachate produced from limed, sulphur-contaminated soils contained elevated levels of sulphate-sulphur. Results obtained from this experiment indicate that a delay in neutralizing a soil that is heavily contaminated with elemental sulphur could cause a movement of soluble metals down the soil profile and may also cause a drain of potentially available nutrients that are required for plant growth.

Additional key words: Acid soils, metals, plant nutrients.

Introduction

Reclaiming soils that are heavily contaminated with elemental sulphur is specific to a small part of the oil and gas industry. Alberta produces 95% of Canada's elemental sulphur by converting the hydrogen sulphide present in sour oil and gas to elemental sulphur (Hyne 1977).

The majority of sour gas plants built prior to the mid 1970's stored elemental sulphur by pouring molten sulphur into a large block. The block was poured on top of a basepad, which was also usually formed from molten sulphur. Many of the basepads were poured directly onto soil, with minimal ground preparation.

There are approximately 105 block basepads at 34 locations in western Canada (Hyne and Schwalm 1983). These basepads range from a few hundred to fifty thousand square meters in area (Hyne 1986). Since 1980 few, if any, new basepads have been established in western Canada.

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The clean-up and reclamation of former sulphur block basepad sites can be a difficult process. As much as 30% sulphur may remain in the soil once the initial clean-up phase is completed. Little information is available about the biological, chemical and physical condition of soils underlying basepads. Data collected to date indicate that the soils can be highly acidified and that large amounts of sulphate-sulphur can accumulate and be leached into the underlying soils (Leitch and Nyborg 1985). The effect of sulphur contamination on the quality of leachate produced from soils was studied in a greenhouse experiment to determine if there were any related environmental and health risks potentially associated with these soils.

The leachate trials outlined in this paper were conducted as part of a larger experiment that investigated the effects of sulphur contamination in soil and evaluated the reclamation effectiveness of particular amendments on these soils (Leggett 1987, Leggett and Parkinson 1988).

Materials and Methods

Experimental Set-up

Soil for a greenhouse experiment was taken near a sour gas plant located in south-central Alberta. Sulphur-contaminated soil was removed from the 0-15 cm deep zone below a freshly exposed portion of the basepad. This soil had a total sulphur concentration of 14%. The control soil came from the same zone depth of a farmer's field located northwest of the gas plant. The control soil was from the same soil map unit as the contaminated soil, had a neutral pH and contained less than 0.1% total sulphur.

Different levels of sulphur were obtained by mixing proportions of contaminated and uncontaminated soils together. The final total sulphur concentrations of the soils used in the experiment were less than 0.1% (control), 4%, 9% and 14%. For each sulphur level the reclamation treatments were no treatment, treatment with limestone ($CaCO_3$) at a rate of three times the detected sulphur, treatment with both $CaCO_3$ and cattle manure (at a rate equivalent to 40 tonnes/ha) and treatment with cattle manure only. Agricultural limestone containing less than 5% $MgCO_3$ was used.

Sixteen pots comprised one replicate (4 sulphur levels x 4 treatments). There were 5 replicates in each trial for a total of 80 pots. Each pot contained 3500g of soil plus amendments, and was planted with reed canary grass.

Leachate Collection

At initiation and completion of the growth trial all pots were watered, by weight, to approximately field capacity plus 150 g. Leachate was collected by draining excess water into collection containers. Selected subsamples from the second leachate trial were acidified for metal analyses (aluminum, iron and manganese).

The pH and conductivity measurements were taken immediately following collection and filtration. The nitrate- and ammonium-nitrogen, phosphate-phosphorous and sulphate-sulphur concentrations of the filtrates were determined using a Technicon II Auto Analyzer. Samples were either analyzed within 48 hours of collection or frozen for analysis at a future date.

All data were tested for significance with a two-way analysis of variance and the Scheffe multiple range test. The accepted level of significance for all tests was $p < 0.05$. Values presented in each table followed by the same letter(s) are not significantly different.

Results

Similar trends in all measured parameters were noted from both leaching trials. Treatment of the soils with manure had no significant effect on any of the parameters measured (Tables 1-7). Leachate produced from unlimed, sulphur-contaminated soils were strongly acidic (Table 1). The pH of leachate generated from control soils were mildly acidic, while the pH of leachate produced from limed, sulphur-contaminated soils were neutral.

Conductivity values were highest in leachate produced from unlimed, sulphur-contaminated soil (Table 2). Within this group, conductivity values increased with increasing sulphur concentration. Leachate from limed, sulphur-contaminated soil were at least ten-fold lower than those from their unlimed counterparts.

Leachate produced from unlimed, sulphur-contaminated soils contained elevated levels of both nitrate- and ammonium-nitrogen (Tables 3 and 4). Nitrate values remained constant as soil sulphur levels increased, whereas ammonium levels increased with increasing soil sulphur concentration. Ammonium-and nitrate-nitrogen in leachate produced from control soils and from limed, sulphur-contaminated soils were not significantly different.

Elevated phosphate-phosphorous levels were found in leachate from unlimed, sulphur-contaminated soils but there was no clear trend with increasing soil sulphur concentration (Table 5). Phosphate concentrations in leachate from limed soils were not significantly different than those from control soils.

Sulphate-sulphur levels were highest in leachate produced from unlimed, sulphur-contaminated soils (Table 6). Limed soils contained the next highest levels, while leachate from control soils had the lowest sulphate concentrations.

Soluble aluminum, iron and manganese in leachate

Unlike other leachate data, metal determinations were made only on selected samples taken following the growth trial. Leachate from manure-treated soils were not analyzed for soluble metals. As only two replicate determinations were made, an analysis of variance was not conducted.

Leachate from unlimed soils contained higher levels of soluble aluminum, iron and manganese than did leachate from control soils (Table 7). Leachate from limed soils contained lower levels of measured soluble metals than did leachate from control soils. Highest levels of metals were recorded from pots containing unlimed 9% sulphur-contaminated soils. Further investigations would be required to determine if this trend is merely a function of low sample replication.

Discussion

The objective of the leaching trials was to chemically characterize the leachate produced from the various soil mixtures. No attempt

TABLE 1. The pH of leachate produced from pot soil used for growth trials. Data are means (n=5) ± standard deviations.

Soil Sulphur (%)	Reclamation Treatments: No Treatment	Limestone	Limestone and Manure	Manure
a) Prior to growth trial				
<0.1	5.3 ± 1.0^{bc}	4.6 ± 1.2^{b}	5.1 ± 0.9^{bc}	5.8 ± 1.0^{bc}
4	1.7 ± 0.2^{a}	7.2 ± 0.3^{c}	6.8 ± 0.7^{c}	1.8 ± 0.1^{a}
9	0.9 ± 0.2^{a}	7.0 ± 0.2^{c}	6.9 ± 0.3^{c}	0.9 ± 0.1^{a}
14	0.5 ± 0.1^{a}	6.9 ± 0.2^{c}	7.1 ± 0.1^{c}	0.5 ± 0.1^{a}
b) Following growth trial				
<0.1	4.9 ± 0.7^{b}	5.0 ± 0.2^{b}	5.5 ± 0.5^{b}	5.3 ± 0.3^{b}
4	1.3 ± 0.4^{a}	7.1 ± 0.4^{c}	$7.3 + 0.1^{c}$	1.4 ± 0.2^{a}
9	1.0 ± 0.1^{a}	$7.3 + 0.2^{c}$	7.4 ± 0.1^{c}	1.0 ± 0.1^{a}
14	0.7 ± 0.1^{a}	7.5[1]	7.5 ± 0.1^{c}	0.7 ± 0.2^{a}

TABLE 2. Electrical conductivity (mS cm^{-1}) of leachate produced from pot soil used for growth trials. Data are means (n=5) + standard deviations.

Soil Sulphur (%)	Reclamation Treatments: No Treatment	Limestone	Limestone and Manure	Manure
a) Prior to growth trial				
<0.1	1.4 ± 0.7^{a}	1.2 ± 0.3^{a}	0.9 ± 0.4^{a}	1.2 ± 0.3^{a}
4	33 ± 5.1^{c}	3.2 ± 0.1^{b}	3.4 ± 0.1^{b}	32 ± 5.0^{c}
9	74 ± 18^{d}	3.5 ± 0.2^{b}	3.8 ± 0.2^{b}	71 ± 13^{d}
14	141 ± 21^{e}	3.1 ± 0.4^{b}	4.4 ± 0.2^{b}	143 ± 14^{e}
b) Following growth trial				
<0.1	0.6 ± 0.3^{a}	0.7 ± 0.3^{a}	0.5 ± 0.3^{a}	0.7 ± 0.3^{a}
4	47 ± 8^{c}	3.2 ± 0.4^{b}	$3.3 + 0.2^{b}$	42 ± 4^{c}
9	61 ± 10^{c}	3.5 ± 0.2^{b}	3.7 ± 0.2^{b}	58 ± 5^{c}
14	108 ± 22^{d}	2.58[1]	3.9 ± 0.4^{b}	124 ± 22^{d}

1 n = 1

TABLE 3. NO_3-N (ug ml^{-1}) in leachate produced from pot soil used for growth trials. Data are means (n=5) ± standard deviations.

Soil Sulphur (%)	Reclamation Treatments: No Treatment	Limestone	Limestone and Manure	Manure
a) Prior to growth trial				
<0.1	30±14[bc]	42±22[c]	21±10[bc]	15±7[bc]
4	291±39[d]	17± 7[bc]	19± 6[bc]	418±85[d]
9	408±73[d]	10± 4[b]	11± 4[bc]	398±95[d]
14	660±74[d]	2± 2[a]	8± 2[b]	618±87[d]
b) Following growth trial				
<0.1	0.4±0.3[1,a]	11±4[2,b]	ND	0.2±0.2[2,a]
4	473±84[c]	0.5±0.1[1,a]	0.7±0.5[a]	578±119[c]
9	522±119[c]	0.8±0.4[a]	0.4±0.2[a]	515±107[c]
14	674±104[c]	ND	1 ± 2[a]	706±151[c]

TABLE 4. NH_4-N (ug ml^{-1}) in leachate produced from pot soil used for growth trials. Data are means (n=5) ± standard deviations.

Soil Sulphur (%)	Reclamation Treatments: No Treatment	Limestone	Limestone and Manure	Manure
a) Prior to growth trial				
<0.1	0.5±0.2[ab]	0.5±0.1[ab]	0.4±0.2[a]	0.4±0.2[a]
4	574±68[c]	0.8±0.3[ab]	1.3±0.4[ab]	861±162[cd]
9	1177±384[d]	1.4±0.3[ab]	1.9±0.3[b]	1264±317[d]
14	1841±238[d]	1.1±0.6[ab]	1.9±1.0[b]	1668±241[d]
b) Following growth trial				
<0.1	0.5±0.3[a]	0.5±0.2[a]	0.6±0.2[1,a]	0.4±0.2[a]
4	925±110[b]	0.3±0.1[a]	0.4±0.3[a]	1034±164[bc]
9	1275±213[bc]	0.5±0.1[a]	0.4±0.2[a]	1548±173[bc]
14	1729±284[c]	0.2[3]	0.7±0.5[a]	2065±373[c]

1 n=3
2 n=2
3 n=1
ND Not determined

TABLE 5. PO_4-P (ug ml^{-1}) in leachate produced from pot soil used for growth trials. Data are means (n=5) ± standard deviations.

Soil Sulphur (%)	Reclamation Treatments: No Treatment	Limestone	Limestone and Manure	Manure
a) Prior to growth trial				
<0.1	0.2±0.1[a]	0.1±0.1[a]	0.6±0.2[a]	0.4±0.1[a]
4	53±16[b]	0.4±0.1[a]	0.6±0.1[a]	72±56[b]
9	169±40[c]	0.5±0.1[a]	0.7±0.1[a]	225±36[c]
14	116±25[bc]	0.5±0.1[a]	0.7±0.1[a]	183±47[c]
b) Following growth trial				
<0.1	0.2±0.2[a]	0.1±0.1[a]	0.4±0.2[1,a]	0.3±0.2[a]
4	96.6[2]	0.1±0.1[a]	0.3±0.1[a]	123±22[c]
9	161±24[cd]	0.2±0.1[a]	0.3±0.1[a]	232±37[d]
14	78±17[b]	0.3[2]	0.4±0.1[a]	147±34[c]

TABLE 6. SO_4-S (mg ml^{-1}) in leachate produced from pot soil used for growth trials. Data are means (n=5) ± standard deviations.

Soil Sulphur (%)	Reclamation Treatments: No Treatment	Limestone	Limestone and Manure	Manure
a) Prior to growth trial				
<0.1	0.7±0.4[ab]	.47±.18[b]	.43±.31[a]	.59±.26[1,ab]
4	38.1±10.8[c]	1.6±0.1[b]	1.6±0.1[b]	37.4±8.1[c]
9	82.0±26.5[c]	1.5±0.1[b]	1.6±0.1[b]	81.7±12.6[c]
14	90.7±11.7[c]	1.4±0.3[b]	1.6±0.1[b]	55.8±23.8[c]
b) Following growth trial				
<0.1	0.3±0.2[1,a]	0.3±0.1[a]	0.5±0.3[2,a]	0.3±0.2[a]
4	50.9±26.2[3,c]	2.3±0.2[b]	2.4±0.1[b]	77.6±11.6[2,c]
9	85.5±5.8[c]	2.4±0.3[b]	2.6±0.2[b]	82.5±22.9[c]
14	67.2±7.8[c]	ND	3.1±0.6[b]	63.9±16.2[c]

1 n=4
2 n=3
3 n=2
ND Not determined

TABLE 7. Soluble aluminum, iron, and manganese (ug ml^{-1}) in leachate produced from selected pot soil used for growth trials. Data are means (n=2) ± standard deviations.

Soil Sulphur (%)	Metal Measured	Reclamation Treatments: No Treatment	Limestone	Limestone and Manure	Manure
<0.1	Al	0.12±0.11	ND	ND	ND
	Fe	0.73±0.44			
	Mn	0.03±0.01			
4	Al	778±3	0.01±0.01	ND	ND
	Fe	4817±1439	0.12±0.04		
	Mn	514±34	0.01±0.01		
9	Al	1082±293	0.01±0.00	ND	ND
	Fe	8220±3224	0.12±0.04		
	Mn	419±169	0.01±0.01		
14	Al	526±163	0.01[1]	ND	ND
	Fe	2554±1055	0.02[1]		
	Mn	13.8±5.7	0.18[1]		

1 n=1
ND Not determined

was made to relate plant nutrient concentrations in leachate with those measured in the soils.

Acidic leachate were produced from all unlimed, sulphur-contaminated pots, while leachate from limed pots were neutral. The two highest sulphur levels resulted in pH values of less than 1.0.

Sulphate-sulphurconcentrations occurred in acidic leachate in levels as high as 91 mg/ml while concentrations in leachate from limed soils ranged up to 26 mg/ml. Conductivity values were greater in leachate from unlimed soils than in those from control or limed soils. The increase in soluble salts present in acidic leachate was likely due to the presence of high levels of sulphate salts. High levels of other nutrients, such as nitrogen and phosphorous, were also present in acidic leachate. In contrast, amounts of nitrogen and phosphorous leached from limed soils were not significantly higher than those from control soils. The use of manure as a reclamation amendment appeared to be ineffective in mitigating the effects of soil acidification such as the leaching of metals and nutrients.

Soluble aluminum, iron and manganese concentrations in leachate produced from limed soils were well below 1 ug/ml, while concentrations in the acidic leachate were recorded in the hundreds of ug/ml. These results indicate the potential for metal contamination from sulphur-contaminated soils that are not limed immediately following basepad removal. Leaving a soil that is heavily contaminated with elemental sulphur exposed and unlimed is not only a potential health hazard as a result of the metal concentrations that could potentially reach the groundwater, but also a drain of potentially available nutrients that are required for plant growth.

The results obtained in this study suggest that leachate from limed, sulphur-contaminated soils would not present a hazard to local groundwater supplies but acidic leachate from unlimed, sulphur-contaminated soils may contaminate groundwater. This study indicates that soils acidified as a result of elemental sulphur contamination require rapid neutralization in order to prevent formation of acidic leachate.

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