Comparison of Nutrient and Metal Loadings with the Application of Swine Manure Slurries and Their Liquid Separates to Soils

D. Kumaragamage,* O. O. Akinremi, and G. J. Racz

Abstract

The accumulation of phosphorus (P) and metals is a serious concern with the continuous application of manure to agricultural soils. Solid–liquid separation of swine slurry is a promising approach to reduce P and metal loadings through application of separated liquid (SL) as a nutrient source. However, little information is available on nutrient and metal loadings with the application of SL compared with unseparated raw manure (RM). We analyzed element concentrations and calculated nutrient and metal loadings for RM and their respective SL applications, considering an application rate of 100 kg total nitrogen (N) ha⁻¹. Samples of SL were obtained through three separation techniques: (i) centrifugation without a flocculant, (ii) centrifugation with a flocculant, and (iii) rotary press with a flocculant. Irrespective of separation technique, calculated P loadings with the application of SL were only 50 to 70% of that of RM at equivalent rates of total N yet exceeded crop removal rate. In contrast, calculated K and Na loadings with SL application were significantly greater than with RM, indicating a possible build-up of K and Na in soil. Calculated Ca and Mg loadings were significantly greater with RM than with SL. Loadings of Al, As, Ba, Cd, Cr, Fe, Mn, Ni, Pb, Sn, Se, Ti, and V were low, whereas Cu and Zn loadings were above crop removal rates for RM and SL. For solid–liquid separation to provide a lasting solution to the problem of P and metal accumulation, the SL must be supplemented with commercial N fertilizer to meet crop N demand.

Core Ideas

- P loading with SL was only 50 to 70% of that of swine slurry applied at equivalent rate of N.
- Loadings of K and Na with SL were higher, indicating a possible salt build-up in soil.
- Cu and Zn loading with application of swine slurry and SL were above crop removal rates.
- Loadings of other trace metals were low with both SL and raw swine slurry application.

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Abbreviations: RM, raw manure; SL, separated liquid.
P. Many techniques have been developed for solid–liquid separation, with varying efficiencies in separating dry matter, nutrients, and metals (Hjorth et al., 2010; Kumaragamage et al., 2013; Popovic et al., 2012). Compared with simple, mechanical separation techniques of swine slurry, the more advanced, chemical flocculant-assisted separation techniques ensure a greater separation efficiency of P and metals (Kumaragamage et al., 2013; Olatuyi et al., 2014; Popovic et al., 2012).

Very limited information is available on loadings of P, other nutrients, and metals to soils with the application of SL as compared with the application of unseparated raw manure (RM). Popovic et al. (2012) observed that Cu and Zn loadings to the soil were markedly lower when SL was applied to supply the crop’s N requirement than Cu and Zn loadings with the application of unseparated raw slurry. They also observed that Cu and Zn loadings were the smallest with application of SL produced by optimized separation treatments that included flocculation and coagulation. More information is needed on nutrient and metal loadings with the application of SL compared with RM to help identify the risks and benefits of using SL as a nutrient source after solid–liquid separation. Our hypothesis is that the application of SL as an N source to crops would result in the accumulation of fewer nutrients and metals in comparison to RM and their SL that were obtained from three separation techniques and calculated and compared nutrient and metal loadings with soil application of RM and their respective SL.

Materials and Methods

Solid–Liquid Separation of Swine Slurry

Four raw manure samples (RM1, RM2, RM3, RM4) obtained from a commercial phytase-fed finisher barn at different times were separated using commercial-scale separation techniques at the Prairie Agricultural Machinery Institute in Manitoba, Canada. The manure samples were taken from an in-ground mixing tank outside the barn within a period of 9 wk before being pumped into earthen manure storage. The separation techniques used were (i) centrifuging without flocculant for RM1, (ii) centrifuging with a cationic flocculant Core Shell 71300 for RM2, and (iii) rotary press with a cationic flocculant Zetag 7875 for RM3 and RM4. More details regarding the flocculants, separation techniques, and collection of representative separated samples (liquid and solid) for RM1, RM2, and RM3 are described elsewhere (Kumaragamage et al., 2013; Olatuyi et al., 2014). The flocculants and separation technique used for RM4 were identical to that of RM3, with the exception that the flocculant rate used for RM4 was slightly higher (0.48 g L⁻¹ of manure slurry) than for RM3 (0.34 g L⁻¹ of manure slurry).

Analysis of Unseparated Raw Manure and Separated Liquid

A total of eight manure samples comprising four raw manures and four separated liquids were analyzed in duplicate. Each SL sample was a composite subsample taken during separation. All manure samples were analyzed for solid content and water content. Samples of RM and SL were thoroughly mixed in a 20-L pail, and a 500-mL subsample was taken for analysis. Subsamples of manure were analyzed within 48 h for pH, electrical conductivity, total N, and ammonium-N (NH₄–N). Another set of subsamples was stored at -13°C until laboratory analysis for P, K, and other elements.

Electrical conductivity and pH of manure samples were determined using a conductivity meter (Accumet AB30, Fisher Scientific) and a pH meter (Accumet AB15, Fisher Scientific) in triplicate at 25°C. Total Kjeldahl N was determined by digesting a 5.0-g manure sample with 12.5 mL concentrated H₂SO₄ and two digestion catalyst tablets (3.5 g potassium sulfate, 0.1 g cupric sulfate, and 0.1 g titanium dioxide per tablet) in a block digester (FOSS North America) at 400°C for 1 h. Samples were cooled and analyzed with a FOSS Kjeltec automated steam distiller where NH₄–N was collected and automatically titrated with 0.1 mol L⁻¹ HCl.

For analysis of total P, K, and other elements, RM and SL samples were digested with concentrated nitric acid in a microwave digestion vessel. The sealed vessel was heated in a MARS5 microwave digester (CEM Corp.) to 175°C over 5 min and held at this temperature for an additional 15 min. The sample digests were diluted to 100 mL with deionized water and analyzed by inductively coupled argon plasma–oceanic emission spectroscopy (Thermo iCAP 6500 Duo). The total concentrations of P, potassium (K), calcium (Ca), magnesium (Mg), sulfur (S), silver (Ag), aluminum (Al), arsenic (As), bariunm (Ba), beryllium (Be), boron (B), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), molybdenum (Mo), sodium (Na), nickel (Ni), lead (Pb), antimony (Sb), selenium (Se), titanium (Ti), thorium (Th), vanadium (V), and zinc (Zn) were determined.

Calculation of Nutrient and Non-Nutrient Metal Loadings with Application of Raw Manure and Separated Liquid

The loadings of nutrients and non-nutrient metals with RM and SL were calculated for application rates that would most likely be used in crop production. Swine slurry (unseparated RM) and SL are commonly used as a N source at rates of about 100 kg total N ha⁻¹ yr⁻¹ or slightly higher (Meade et al., 2011; Schlegel et al., 2015; Xu et al., 2013). Thus, a comparison of loadings of nutrients and metals using RM at 100 kg total N ha⁻¹ versus SL at an equivalent rate was made.

Statistical Analysis

Analysis of variance of nutrient and metal concentrations in RM and SL and calculated loadings were performed using the MIXED procedure in SAS software, version 9.2 (SAS Institute, 2008), considering different sources of RM (RM1, RM2, RM3, and RM4), and their respective SL separated using different techniques, as pseudo-replicates. Mean separation was determined using the Tukey–Kramer test at P < 0.05. Input data were tested for normality using the Shapiro–Wilk’s test. In cases where the residual (error terms) in the untransformed data did not follow a normal distribution (Shapiro–Wilk’s value <0.8), the data were log-transformed before analysis and back-transformed to obtain geometric means. For all statistical analyses, significance was determined at P < 0.05.
Results and Discussion

Properties of Raw Manure and Separated Liquids

Results comparing the efficiency of the three separation techniques in partitioning solids, P, and metal elements to the solid separate have been previously described (Kumaranagama et al., 2013; Olatuyi et al., 2014). In general, the rotary press with a cationic flocculant was superior to centrifuge techniques (with or without flocculant) in partitioning solids, P, and metals to the separated solids. The use of a flocculant in the centrifuge technique significantly enhanced the partitioning efficiency. Results contained herein compare the concentrations of P and metals in RM and SL and compare P and metal loadings to soils with applications of RM and SL, regardless of the separation techniques used.

The four unseparated RMs had solid contents from 23.5 and 55.3 g kg\(^{-1}\), whereas their SL had solid contents between 6.7 and 14.1 g kg\(^{-1}\) (Table 1). The pH values were near neutral and similar for the RM and their respective SL. Electrical conductivity values in RM varied widely from 4.9 to 17.0 mS cm\(^{-1}\), with similar electrical conductivity values in their respective SL. On a fresh (wet) weight basis, RM had total N concentrations ranging from 2.56 to 3.42 g kg\(^{-1}\). Total N concentrations in the SL were considerably lower than in the RM (range, 1.53–2.56 g kg\(^{-1}\)). Based on the total N concentration, the calculated rates of manure application to provide 100 kg N ha\(^{-1}\) were 34, 30, 39, and 29 t (fresh weight) ha\(^{-1}\) for RM1, RM2, RM3, and RM4, respectively. The calculated application rates for the respective SL1, SL2, SL3, and SL4 were greater, at 39, 47, 66, and 56 t (fresh weight) ha\(^{-1}\), respectively.

Element Concentrations in Raw Manure and Separated Liquid

Both RM and SL had high concentrations of NH\(_4\)-N, with mean NH\(_4\)-N concentrations of 1798 and 1575 mg kg\(^{-1}\), respectively, which was about 59% of total N in RM and about 80% of total N in SL (Table 2). Total N and NH\(_4\)-N concentrations were typical for raw swine slurries (Chelme-Ayala et al., 2011; Le et al., 2008; Sørensen and Amato, 2002) and SL of swine slurries (Vanotti et al., 2009; Walker et al., 2010). Mean total K concentrations in RM and SL were high (approximately 1200 mg kg\(^{-1}\)). Potassium exists in a highly soluble form in manure, and therefore high concentrations in RM and SL are to be expected (Saey et al., 2005). Both RM and SL had high concentrations of Na (about 600 mg kg\(^{-1}\)) that were comparable to Na concentrations previously reported for swine slurry (Brenchin and McDonald, 1994). Sodium is a nonessential nutrient and therefore is biologically a less significant element, but accumulation in soil may cause sodicity, resulting in soil quality deterioration (Brenchin and McDonald, 1994; Lal et al.,

### Table 1. Solid content, pH, electrical conductivity, and total N concentration of unseparated raw manure (RM) and their liquid separates.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Unseparated/ separated liquid</th>
<th>Solid content(\dagger) g kg(^{-1})</th>
<th>pH(\dagger)</th>
<th>Electrical conductivity(\dagger) mS cm(^{-1})</th>
<th>Total N concentration(\dagger) g kg(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>RM1</td>
<td>unseparated</td>
<td>26.1 (0.48)</td>
<td>6.9 (0.04)</td>
<td>5.4 (0.2)</td>
<td>2.93</td>
</tr>
<tr>
<td>RM1</td>
<td>separated liquid</td>
<td>14.1 (0.02)</td>
<td>7.1 (0.06)</td>
<td>5.2 (0.4)</td>
<td>2.56</td>
</tr>
<tr>
<td>RM2</td>
<td>unseparated</td>
<td>41.9 (0.64)</td>
<td>6.7 (0.03)</td>
<td>17.0 (1.1)</td>
<td>3.38</td>
</tr>
<tr>
<td>RM2</td>
<td>separated liquid</td>
<td>12.1 (0.27)</td>
<td>7.1 (0.04)</td>
<td>16.0 (0.9)</td>
<td>2.24</td>
</tr>
<tr>
<td>RM3</td>
<td>unseparated</td>
<td>23.5 (0.28)</td>
<td>7.0 (0.02)</td>
<td>5.0 (0.2)</td>
<td>2.56</td>
</tr>
<tr>
<td>RM3</td>
<td>separated liquid</td>
<td>6.7 (0.23)</td>
<td>7.0 (0.03)</td>
<td>4.7 (0.3)</td>
<td>1.53</td>
</tr>
<tr>
<td>RM4</td>
<td>unseparated</td>
<td>55.3 (0.52)</td>
<td>6.9 (0.04)</td>
<td>4.9 (0.2)</td>
<td>3.42</td>
</tr>
<tr>
<td>RM4</td>
<td>separated liquid</td>
<td>10.3 (0.19)</td>
<td>7.2 (0.03)</td>
<td>5.2 (0.2)</td>
<td>1.78</td>
</tr>
</tbody>
</table>

\(\dagger\) Means of three replicates. Values in parentheses are SD.

### Table 2. Mean comparison for concentrations (fresh weight basis) of ammonium-N and total elements in unseparated raw manure and their liquid separates.

<table>
<thead>
<tr>
<th>Nutrient or element</th>
<th>Unseparated raw manure(\dagger) mg kg(^{-1})</th>
<th>Separated liquid(\dagger) mg kg(^{-1})</th>
<th>P value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total N</td>
<td>3073 (407)(\dagger)</td>
<td>2028 (463)(\dagger)</td>
<td>0.001</td>
</tr>
<tr>
<td>NH(_4)-N</td>
<td>1798 (168)(\dagger)</td>
<td>1575 (125)(\dagger)</td>
<td>0.077</td>
</tr>
<tr>
<td>P</td>
<td>808 (138)(\dagger)</td>
<td>320 (90)(\dagger)</td>
<td>0.001</td>
</tr>
<tr>
<td>K</td>
<td>1211 (86)(\dagger)</td>
<td>1127 (234)(\dagger)</td>
<td>0.526</td>
</tr>
<tr>
<td>Na</td>
<td>577 (15)(\dagger)</td>
<td>604 (22)(\dagger)</td>
<td>0.089</td>
</tr>
<tr>
<td>Ca</td>
<td>550 (85)(\dagger)</td>
<td>244 (85)(\dagger)</td>
<td>0.002</td>
</tr>
<tr>
<td>Mg</td>
<td>495 (86)(\dagger)</td>
<td>183 (54)(\dagger)</td>
<td>0.001</td>
</tr>
<tr>
<td>S</td>
<td>350 (77)(\dagger)</td>
<td>165 (36)(\dagger)</td>
<td>0.005</td>
</tr>
<tr>
<td>Zn</td>
<td>69.2 (8.6)(\dagger)</td>
<td>19.8 (14.4)(\dagger)</td>
<td>0.001</td>
</tr>
<tr>
<td>Fe</td>
<td>46.4 (8.8)(\dagger)</td>
<td>13.4 (9.5)(\dagger)</td>
<td>0.002</td>
</tr>
<tr>
<td>Cu</td>
<td>24.7 (6.4)(\dagger)</td>
<td>8.2 (6.3)(\dagger)</td>
<td>0.010</td>
</tr>
<tr>
<td>Al</td>
<td>18.3 (5.2)(\dagger)</td>
<td>4.4 (2.0)(\dagger)</td>
<td>0.003</td>
</tr>
<tr>
<td>Mn</td>
<td>12.0 (2.3)(\dagger)</td>
<td>3.4 (1.5)(\dagger)</td>
<td>0.001</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Nutrient or element</th>
<th>Unseparated raw manure(\dagger) mg kg(^{-1})</th>
<th>Separated liquid(\dagger) mg kg(^{-1})</th>
<th>P value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti</td>
<td>0.90 (0.23)(\dagger)</td>
<td>0.27 (0.13)(\dagger)</td>
<td>0.003</td>
</tr>
<tr>
<td>Ba</td>
<td>0.84 (0.20)(\dagger)</td>
<td>0.27 (0.11)(\dagger)</td>
<td>0.002</td>
</tr>
<tr>
<td>V</td>
<td>0.73 (0.15)(\dagger)</td>
<td>0.30 (0.05)(\dagger)</td>
<td>0.002</td>
</tr>
<tr>
<td>Mo</td>
<td>0.39 (0.12)(\dagger)</td>
<td>0.20 (0.04)(\dagger)</td>
<td>0.031</td>
</tr>
<tr>
<td>Ni</td>
<td>0.30 (0.06)(\dagger)</td>
<td>0.15 (0.05)(\dagger)</td>
<td>0.008</td>
</tr>
<tr>
<td>Pb</td>
<td>0.27 (0.08)(\dagger)</td>
<td>0.10 (0.01)(\dagger)</td>
<td>0.006</td>
</tr>
<tr>
<td>Sb</td>
<td>0.21 (0.03)(\dagger)</td>
<td>0.13 (0.04)(\dagger)</td>
<td>0.020</td>
</tr>
<tr>
<td>Cd</td>
<td>0.06 (0.01)(\dagger)</td>
<td>0.03 (0.01)(\dagger)</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Cr</td>
<td>0.04 (0.02)(\dagger)</td>
<td>0.02 (0.01)(\dagger)</td>
<td>0.048</td>
</tr>
</tbody>
</table>

\(\dagger\) Means of four unseparated manures or separated liquids. Values in parentheses are SD.

\(\dagger\) Means within a row followed by the same lowercase letter are not significantly different at P ≤ 0.05.
Concentrations of major and secondary nutrients (N, P, K, Ca, Mg, and S) were greater than 300 mg kg\(^{-1}\) in RM and followed the order \(\text{NH}_4^-\text{N} > \text{K} > \text{P} > \text{Ca} > \text{Mg} > \text{S}\). In the SL, concentrations of major and secondary nutrients were >150 mg kg\(^{-1}\) and followed an order similar to that of RM. Concentrations of K, P, and Ca when converted to a dry weight basis were somewhat lower than the median concentrations (dry weight basis) reported for pig manures, but Mg concentrations were similar (Sager, 2007). Cationic micronutrient concentrations ranged from 12 to 70 mg kg\(^{-1}\) in RM and from 3 to 20 mg kg\(^{-1}\) in SL and followed the same order in RM and SL, with concentrations of Zn > Fe > Cu > Mn. Of micronutrients that exist in an anionic form in soil, the concentration of Mo was very low but detectable (0.39 and 0.2 mg kg\(^{-1}\) in RM and SL, respectively), whereas B concentration was below the detection limit in both RM and SL digests (Table 2). Concentrations of micronutrients in RM of the present study are comparable to those reported by Sager (2007) for pig manure on a dry weight basis.

Of the metals other than nutrients, Al was in high concentration in RM (18 mg kg\(^{-1}\)) and SL (4 mg kg\(^{-1}\)). All other metals were found in very low concentrations, with detectable levels of Ti, B, V, Ni, Pb, Sc, Cd, and Cr ranging from 0.04 to 0.9 mg kg\(^{-1}\) in RM and from 0.02 to 0.3 mg kg\(^{-1}\) in SL (Table 2). Concentrations of Ag, As, Co, Be, Se, and Tl were below detection limit for some or all RM and SL digests, and hence the data are not shown. Sager (2007) also reported that trace elements (e.g., Ni, Pb, As, and Hg) were found in manure at rather low concentrations.

Concentrations of all elements were normally distributed (Shapiro–Wilk’s W > 0.8). On a fresh weight basis, the concentrations of \(\text{NH}_4^-\text{N}\) and total elements, with the exception of Na, were greater in unseparated RM than in SL (Table 2). The decrease in concentrations in SL over RM was statistically significant \((P < 0.05)\) for all elements (nutrients and metals) other than \(\text{NH}_4^-\text{N}, \text{K}, \text{and} \text{Na}\). The decrease in element concentrations in SL compared with RM could be due to the preferential partitioning of some elements to the solid separate during solid–liquid separation, thus reducing the concentrations in SL. In addition, SL had a smaller proportion of solids and a greater proportion of water compared with RM; thus, element concentrations are more diluted in SL than in RM on a fresh weight basis. In contrast to other elements, \(\text{NH}_4^-\text{N}, \text{K}, \text{and} \text{Na}\) concentrations, on a fresh weight basis in SL, were not significantly different from those in the RM (Table 2). Because \(\text{NH}_4^-\text{N}, \text{K}, \text{and} \text{Na}\) in manure predominantly exist in dissolved forms, these elements would be preferentially partitioned into the SL and, as a result, showed similar concentrations in RM and SL in spite of the greater proportion of water in SL diluting the element concentrations.

### Nutrient and Metal Loadings to Soils after Application of Raw Manure and Separated Liquid

The loadings of nutrient and metal elements with SL applied to provide 100 kg total N ha\(^{-1}\) were compared with loadings with RM applied to provide an equivalent rate of total N. The amount of \(\text{NH}_4^-\text{N}\) received at this rate of total N was 59 ± 6 and 80 ± 14 kg ha\(^{-1}\) for RM and SL, respectively (Fig. 1). Thus, SL provided significantly \((P = 0.03)\) greater amounts of \(\text{NH}_4^-\text{N}\) than RM. Loading of P was significantly \((P = 0.0002)\) less with SL than RM at an equivalent rate of N. This was expected because a larger proportion of P is partitioned to solid separate with solid–liquid separation, whereas a smaller proportion goes into SL (Kumaragamage et al., 2013; Peters et al., 2011; Popovic et al., 2012), resulting in a lower concentration of P in SL (Table 2). Loading of P with RM and SL was 26 ± 1 and 16 ± 2 kg ha\(^{-1}\), respectively (Fig. 1); thus, SL supplied only about 60% of P as RM at equivalent rates of total N. The total P loading with both RM and SL were comparable to estimated total P removal by modern crops, which is often in the range of 5 to 35 kg P ha\(^{-1}\) (Mikkelsen et al., 2014; Withers et al., 2001), depending on the crop. Thus, total P loading with the application of RM and SL at a rate of 100 kg N ha\(^{-1}\) can lead to a build-up of soil P for crops with lower P uptake, which could be avoided or reduced through application of SL instead of RM. For soils with already high soil test P levels, the application of SL instead of RM would reduce further build-up of P.

Loadings of K with SL were significantly greater \((P = 0.003)\) than with RM (56 ± 6 and 40 ± 4 kg ha\(^{-1}\), respectively) (Fig. 1). Thus, compared with RM, SL is a more concentrated source of K. However, at the rates applied, RM and SL provided only a fraction of the total crop requirement of K (Bauer et al., 2015; Masters et al., 2016), but the amount supplied was comparable to or greater than the K removal by most crops grown in western Canada (MAFRI, 2007). The continuous application of SL, with
a greater K content, may increase the exchangeable K in soils, leading to cation exchange sites dominated by K rather than Ca.

Loading of Na followed trends noted for K (Fig. 1), with SL supplying \(30 \pm 6\) kg ha\(^{-1}\), which was significantly \((P = 0.01)\) greater than the loading of Na with RM \((19 \pm 6\) kg ha\(^{-1}\)). The Na loading observed by Brechin and McDonald (1994) with a similar rate of swine slurry application \((32\) kL ha\(^{-1}\)) as used in this study \((\approx 34\) kL ha\(^{-1}\)) was much smaller \((8.3\) kg Na ha\(^{-1}\)) yet resulted in a significant increase in the tissue Na concentration of barley. Although the Na tissue concentrations they observed were not high enough to adversely affect plant growth, the authors suggested that continued use of high rates of swine slurry, particularly in drier environments, might result in the build-up of Na in the soil. With the application of SL in the present study, the loading of Na was nearly 60\% greater than that of RM and was almost 4-fold of Na loading observed with unseparated swine slurry by Brechin and McDonald (1994). This shows that continuous application of SL may enhance Na build-up in soils relative to RM, which may adversely affect soil properties.

Loadings of Ca and Mg, in contrast, were significantly greater \((P < 0.001)\) with RM than with SL (Fig. 1). With SL and RM, loadings of Ca and Mg were small \((<18\) and \(<12\) kg ha\(^{-1}\) for RM and SL, respectively) compared with amounts typically found in soils and hence will have little or no effect on Ca and Mg concentrations in soils. Loadings of S were low with both RM and SL \((11\) and \(8\) kg ha\(^{-1}\), respectively), but the difference in S loadings with RM and SL was significant \((P = 0.009)\). Crop removal of S is typically in the range of \(10\) to \(30\) kg S ha\(^{-1}\) (Masters et al., 2016; Norton et al., 2013) and depends on the crop and crop yields. Thus, the amount of S added to soils with RM or SL was about equal to or less than the amounts removed annually by most crops.

Mineral salts are routinely added to animal feed globally (Li-Xian et al., 2007), resulting in increased concentrations of Na, K, Ca, and Mg in the manure (Choudhary et al., 1996). Thus, successive application of manures to soil may lead to a build-up of salt concentration in soils. When converted to salt equivalents added per hectare, the sum totals of Na, K, Ca, and Mg added with RM and SL \((\text{at a rate of application to provide} \ 100\ \text{kg total N ha}^{-1})\) were similar at about \(4000\) equivalents ha\(^{-1}\) (data not shown). However, our calculated loading of elements in the present study indicates that applying SL instead of RM provides more Na and K and less of Ca and Mg to soils, which may change the proportion of exchangeable cations in soils and may adversely affect soil properties.

Calculated loadings of Fe with applications of RM and SL were \(1.5 \pm 0.1\) and \(0.6 \pm 0.3\) kg ha\(^{-1}\), respectively, and the corresponding loadings of Mn were \(0.39 \pm 0.03\) and \(0.16 \pm 0.05\) kg ha\(^{-1}\), respectively (Fig. 2). Thus, loadings of Fe and Mn with RM and SL applications were insignificant in comparison to the typical amounts of total Fe and Mn concentrations found in soils (Madrid et al., 2007; Moharami and Jalali, 2013) and were comparable to amounts removed through crop uptake (Masters et al., 2016). Hence, repeated applications of RM or SL will have little or no effect on total concentrations of Fe and Mn in soils. In contrast, loadings of Cu and Zn with application of RM and SL were much greater than the crop removal of these elements. The calculated loadings of Zn in the present study with RM and SL applications were \(2.3 \pm 0.2\) and \(0.9 \pm 0.5\) kg ha\(^{-1}\), respectively (Fig. 2), whereas the reported value for crop removal for Zn \((\text{converted to kg ha}^{-1})\) is \(<0.5\) kg ha\(^{-1}\) (Bauer et al., 2015; Masters et al., 2016; Xu et al., 2013). Similarly, the calculated loadings of Cu with RM \((0.8 \pm 0.5\) kg ha\(^{-1}\)) and SL \((0.4 \pm 0.2\) kg ha\(^{-1}\)) were much greater than the previously reported crop removal rates when converted to kg ha\(^{-1}\) \(<0.1\) kg ha\(^{-1}\) (Saha et al., 2013; Xu et al., 2013). Qian et al. (2003) estimated total Cu and Zn loadings of \(0.1\) and \(0.05\) to \(0.7\) kg ha\(^{-1}\), respectively, which are comparable to loadings in the present study, with swine manure application rates to provide \(100\) kg total N ha\(^{-1}\). Copper and Zn are added to animal feeds as nutritional supplements and/or to suppress diseases; hence, repeated manure applications have resulted in Cu and Zn accumulation in soils (Formentini et al., 2015; Xu et al., 2013). In spite of increased concentrations of total and available Zn in soils, some researchers observed that the total concentrations remained below the maximum regulatory levels suggested for the region and thus may not pose an environmental risk (Penha et al., 2015; Qian et al., 2003). Contrary to the above, Mantovi et al. (2003) reported soil Cu concentration in manured soils of northern Italy to be high according to the European legislation, but there was no risk of contamination of the food chain. Because the loadings of Cu and Zn in the present study were much above crop removal rates, Cu and Zn accumulations should be monitored with long-term manure applications.

Loadings of Al were low \(<0.6\) kg ha\(^{-1}\) with application of RM and SL to provide \(100\) kg total N ha\(^{-1}\) (Fig. 2) and were negligible compared with total Al concentration in soil; hence, applications of RM or SL have little or no effect on total Al concentrations in soils. Loadings of Fe, Mn, Cu, Zn, and Al with application of SL were significantly \((P < 0.05)\) smaller (over \(50\%\) less; Table 2) than application of RM to provide equivalent rates of total N (Fig. 2). Sommer et al. (2015) obtained similar results, where loadings of Cu with the application of SL to a wheat field were lower than the crop uptake, whereas the loadings with separated solid was greater than crop uptake. Popovic et al. (2012) also observed that Cu and Zn loadings to the soil were markedly lower with the application of SL to supply the crop’s N requirement than with the application of unseparated raw slurry.

Loadings of B, Ti, Ba, V, Mo, Ni, Pb, Sb, Cd, and Cr with application of RM and SL at rates of \(100\) kg of N ha\(^{-1}\) were very low \(<40\) g ha\(^{-1}\) in comparison to loading of metals such as Cu, Zn, Fe, and Mn and hence are not likely to limit long-term manure applications to land. Loadings of the above metals with application of SL were less than with RM at equivalent rates of total N, and the differences in loadings between SL and RM were
significant ($P < 0.05$) for Ti, Ba, V, Ni, Pb, and Cd. Therefore, the risk of metals accumulation is less with the application of SL than RM for most metals.

**Conclusions**

There was high variation in elemental concentrations of RM and SL and hence high variations in the loadings of various elements. Application of SL at a rate to supply 100 kg total N ha$^{-1}$ supplied less P and metals compared with the application of RM. Thus, at equivalent rates of total N, using SL instead of RM as a N source in crop production would minimize the build-up of soil P and most metals. Loadings of Na, however, were greater with the application of SL than with RM. Monitoring for build-up of salts and Na in soils will have to be more frequent for soils treated with SL than with RM to maintain soil Na concentrations within acceptable levels and to prevent the development of salinity. Analysis of manures for elements added as supplements to feeds needs to be conducted for long-term manure management. For solid–liquid separation to provide a lasting solution to the problem of P and metal accumulation in soil, the SL must be applied at rates below that which may cause P build-up and metal toxicities to plants and/or soil microorganisms. The Zn, Cu, and other metals in animal feeds may have to be reduced to attain sustainability. In addition, synthetic fertilizer may need to be used to supplement manures to meet the crop N demand and thus maintain low loadings of P and metals.

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