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Waste Management Research 2008; 26; 163

DOI: 10.1177/0734242X07082138

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Release of nitrogen and trace metal species from field stacked biosolids

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Concerns over elevated nitrate (NO_3^-) levels found in groundwater near former biosolid stockpiling locations resulted in the Maine Department of Environmental Protection (MDEP) imposing stricter regulations governing the stockpiling of biosolids in October 2002. The goals of this study were to measure the amount and speciation of nitrogen (N) and trace metals leaving stockpiled biosolids and travelling through the soil column. The biosolids were placed on plastic-lined cells to collect all leachate. Ammonium (NH_4^+), ranging from 2000 to 4900 mg L^{-1} , was the dominant N species (90% of total N) in the leachate from the Class B lime-stabilized biosolids in the lined cell experiment. Nitrate (NO_3^-) and nitrite (NO_2^-) concentrations were negligible, remaining below 0.25 and 0.1 mg L^{-1} , respectively. Dissolved organic carbon (DOC) concentrations as high as 8900 mg L^{-1} and chemical oxygen demand (COD) as high as 37 000 mg L^{-1} were measured in the leachate leaving the lined cell. Fifteen zero-tension pan lysimeters (ZTP-lysimeter) were installed in a 90 m^2 plot at depth intervals of 30, 60, and 100 cm. Leachate passing through the soil column underlying the biosolids stockpile was collected in the ZTP-lysimeters. The average ZTP-lysimeter NH_4^+ concentrations ranged from 1400 mg L^{-1} at 60 cm depth to 145 mg L^{-1} at 90 cm depth. The average ZTP-lysimeter DOC concentrations ranged from 2000 mg L^{-1} at 60 cm to 525 mg L^{-1} at 90 cm. Trace metal determinations of the leachate collected from the lined cell and ZTP-lysimeters showed arsenic loading rates exceeded the state limits of 0.5 $\text{kg ha}^{-1} \text{ year}^{-1}$ by an order of magnitude. Arsenic concentrations were in excess of several thousand milligrams per litre in the lined-cell leachate and several hundred milligrams per litre in the ZTP-lysimeters as deep as 90 cm under the biosolid stockpile. Phosphorus, iron and manganese in excess of several thousand milligrams per litre were observed in both the lined-cell leachate and ZTP-lysimeters. Significant concentrations of other trace metals were found at depth in the zero-tension ZTP-lysimeter plot. Trace metals were largely mobilized by the DOC from the biosolids and due to the presence of anaerobic environment, especially in the underlying soil.

Keywords: Biosolids, trace metals, nutrients, groundwater quality, wmr 1069-2

Introduction

Biosolids are derived from sewage sludge and must be treated to meet land-application criteria specified by the US EPA part 503 rules developed in 1993 (US EPA 1993) to ensure safe agronomic use. Class A biosolids have been treated to reduce pathogens to natural background concentrations. Class B biosolids have been treated to reduce pathogens, but not to background concentrations. The uses of Class B biosolids are

restricted by well defined site suitability and access control regulations; land application is a primary outlet for this class.

Field application of biosolids, especially in agricultural areas, is practiced widely (Wang 1997; Yager *et al.* 2003). Field stacking of biosolids is an allowed practice in many countries and states to accommodate cropping schedules and seasonal field conditions (US EPA 2000). Hundreds to thousands of

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DOI: 10.1177/0734242X07082138

Received 26 July 2006; accepted in revised form 29 May 2007

cubic metres of material can be stored and then applied in one application. Field stacking is typically done in agricultural areas that are remote from public access and may occur on bare ground (USEPA 2000, Peckenham 2005).

Biosolids composition is well studied (e.g. Hall & Williams 1984, Stukenberg *et al.* 1993, Berti & Jacobs 1998, Kellog *et al.* 2000, Estes & Buob 2001; Shober *et al.* 2002, Chaney *et al.* 2004). The nutrient content of Maine's biosolids is: 0.1 to 8% nitrogen; 0.1 to 3.2% phosphorus; and 0.0 to 0.8% potassium (Peckenham 2005). Biosolids also contain trace metals such as chromium (Cr), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), nickel (Ni), and zinc (Zn) with individual concentrations > 100 ppm, along with a variety of other metals at lower concentrations (Cappo 1984, Bell *et al.* 1991, McBride *et al.* 2004, Basta *et al.* 2005). Field application of biosolids, both as amendments and for storage purposes, can result in the release of nutrient and metal species into the surrounding environment (Gerritse *et al.* 1982, Clapp *et al.* 1994, McBride *et al.* 1997, Shober *et al.* 2002).

Elevated concentrations of nitrate (NO_3^-) levels in groundwater surrounding agricultural and grazing lands have been reported (Smith *et al.* 1998a, b, Maticic 1999, Smith & Frost 2000, Maeda *et al.* 2003), but there is little information about the release of NO_3^- from biosolids stockpiles. Leaching of NO_3^- from biosolids stockpiles was suggested as the source of nitrogen plumes measured in a gravel pit reclamation site, and from repeated stacking of chicken manure at the same site (McDowell & Chestnut 2002). Even though some studies have reported the significant impacts of manure stockpiling and cattle slurry lagoons on groundwater quality (Goody *et al.* 2002), research that exclusively measured the N losses from field stacked biosolids is lacking.

Trace metal release from biosolids has been linked to microbiological activity (Qureshi *et al.* 2003), water solubility (McBride 1998), and breakdown of organic compounds that bind metals in the soil (Martinez *et al.* 2003). McBride *et al.* (1999) showed that shallow groundwater collected at an experimental plot that had a single high-dosage application of biosolids 15 years previously had elevated levels of several trace metals when compared with a nearby control plot that had received no biosolids. Richards *et al.* (1998) found excess trace metals such as boron (B), cadmium (Cd), Ni and Zn in soil porewater at a site treated with biosolids 20 years earlier. The evidence for increased trace metal concentrations in waters surrounding biosolid application sites, including a 20-year long watershed scale experiment in Minnesota (Clapp *et al.* 1994), is mixed due to natural heterogeneity of soils, uncertainties in biosolids compositions, and lack of control sites (Gerritse *et al.* 1982, Stukenberg *et al.* 1993, Berti & Jacobs 1998, Babarick *et al.* 1998, Sloan *et al.* 1998, National Research Council 2002).

Accounting for all the metals in land-applied biosolids by material balance has yielded varying results (Babarick *et al.* 1998, McBride *et al.* 1999; Sloan *et al.* 2000). In developing regulations, the US EPA has assumed that trace metal loadings are attenuated by the high organic content of biosolids

and that metal leaching from biosolids is not of concern (Qureshi *et al.* 2003). Baveye *et al.* (1999) reported trace metal losses of 36 to 60% in biosolids-amended soils tested to depths of 75 cm, and similar results have been reported by Chang *et al.* (1984), Bell *et al.* (1991), Dowdy *et al.* (1991), and Richards *et al.* (1998). The lack of mass balance for metals may be caused by differing analytical extraction methods (Dowdy *et al.* 1991) and lateral dispersion coupled with soil erosion (McGrath & Lane, 1989). In a long-term study conducted by Bergkvist *et al.* (2003), it was found that 99% of the Cd applied via bi-annual biosolids applications spanning 41 years was accounted for in the top 17 cm of the soil profile. By contrast, Yingming & Corey (1993) found only 15% of the applied Cd from biosolids remained in the topsoil after 11 years.

This work focuses on measuring the concentrations and loading rates of leachable constituents and evaluating the size fractionation of trace metals in biosolids leachate: arsenic (As), copper (Cu), selenium (Se), Cd, Ni and Zn. These metals were chosen because there are regulatory limits for annual and lifetime loading to soil. Aluminium (Al), Fe and Mn were included because they form (oxyhydr)oxides that can remove anions and trace metals from solution. Metal (oxyhydr)oxides dissolve via acid-catalysed and reductive dissolution mechanisms that result in the release of the absorbed species (Merrington *et al.* 2003, Speir *et al.* 2003, Richards *et al.* 2004, Lin *et al.* 2004, Basta *et al.* 2005).

Concerns over high NO_3^- levels in groundwater collected from wells near former biosolids stockpiling sites prompted the Maine Department of Environmental Protection (MDEP) to enact stricter siting regulations in October 2002 (CMR 06-096). As part of establishing these rules, the MDEP commissioned this field study to determine whether stricter regulation of field stockpiling of biosolids was justified. This study was expanded to examine trace metal release and transport from field-stacked biosolids. Metal contamination of groundwater due to stockpiling of biosolids presents a special case of risk for rural water quality. The existing state regulations focus on metal loading to soil and are nearly silent on metals in groundwater.

Materials and methods

Plot designs

Two experimental plots were constructed and stacked with municipal Class B biosolids at the University of Maine's research farm. The local soils are formed in a dense glacial till, Marlow silt loam (coarse loamy isotropic frigid oxyaquic Haplorthod) and Woodbridge silt loam (coarse loamy mixed mesic aquic Dystrudept) that meets Maine's stockpiling suitability criteria for slope (< 5%), drainage class (moderately well-drained), and texture (silt loam).

The plots consisted of: (1) a pair of lined cells to capture all leachate and runoff generated, and (2) a zero-tension pan lysimeter (ZTP-lysimeter) plot to capture flow through the soil beneath field-stacked biosolids. The design is described in detail in Peckenham *et al.* (2005) and ZTP-lysimeter samplers are known to have varying capture efficiencies (Jemison

& Fox, 1994; Zhu *et al.* 2002). Briefly, one 3.6 m × 22.5 m cell and one 1.8 m × 30.5 m cell were constructed by removing the vegetation and sloping the base inward. A 1.27 mm thick impermeable PVC sheet was placed over the base and a slotted plastic pipe was placed along the centreline to collect the leachate. The PVC barrier and the drain pipe were covered with 18 cm of fine sand to facilitate flow of the leachate. Lime-stabilized class-B biosolids were stacked to a depth of 0.6 to 0.8 m as unloaded from truck or tractor bucket.

The watershed from stockpile surfaces was collected using an open pipe system placed along the perimeter of the stockpile. The leachate and runoff flow rates were measured using 90° V-notch weirs and flow meters (ISCO® 4230). Auto-samplers triggered by the flow meters collected 100 mL samples for every 100 L of flow (Peckenham *et al.* 2005).

The ZTP-lysimeter plot was 6 m × 15 m, and consisted of 15 stainless steel pan lysimeters (45 cm × 45 cm × 5 cm) placed at 0.3, 0.6, and 1 m below grade. The ZTP-lysimeters were individually drained into buried polyethylene containers. Samples were collected using a manual vacuum pump. Lime-stabilized biosolids were stacked to a maximum thickness of approximately 1 m.

The stockpiling experiments for both the lined cell and the ZTP-lysimeters were performed twice. The first experiment (December 2002 to May 2003) tested 125 m³ of biosolids in the lined cells and 90 m³ in the ZTP-lysimeter plot. Freezing temperatures made flow quantification difficult to impossible. The second experiment (July 2003 to May 2004) tested 52.5 m³ of biosolids in the lined cell and 90 m³ in the ZTP-lysimeter plot. The road sand on the bottom of the lined cell was allowed to be rinsed by several rain storms between experiments, whereas the ZTP-lysimeter plot was scraped lightly and aerated for 30 days before restacking.

Sampling methods

The biosolids, aqueous samples, and soils samples were analyzed for nitrogen species, total organic carbon, metals, and pH. In addition, biosolids were analyzed for solids and liquids for chemical oxygen demand. A random grid was used to collect 10 biosolids or soil samples for compositing. Compositing biosolids were mixed in a plastic container and then re-sampled. Compositing biosolids samples were sampled immediately after stacking and just before stack dismantling. Soil samples were collected with a soil sampling tube (25.4 cm). All samples were stored on ice during transportation and kept at 4°C until analysed (within 7 days).

Data reported here are for the second stockpiling experiment. Aqueous samples from the lined cells were collected from the auto-samplers every 3 to 14 days. The individual 500 mL polyethylene bottles in the auto-samplers were transferred into Whirl-Pak® bags and placed on ice in coolers for transport. Daily rainfall data collected at the farm was used and verified with four rain gauges located at the field test site.

The ZTP-lysimeters were sampled approximately monthly from June 2003 to October 2003. Additional samples were collected in May and July 2004. Samples from each individual

ZTP-lysimeter were placed in an acid-washed 500 mL polyethylene bottle. The bottles were kept on ice for transport.

Analytical methods

Biosolids pH was determined by adding 70 mL of de-ionized (DI) water to approximately 35 mL of biosolids in a Nalgene® bottle. The bottle was capped, thoroughly shaken and the pH of the slurry measured after 15 min (Corning Model 430 meter with an Orion Model 8104-BN probe or Hach pH pen for field measurement). The dissolved oxygen (DO) concentration was measured at the collection point for the auto-sampler (where the leachate drained into a collection bucket) with a YSI portable field meter during several rain events.

Ammonium samples were filtered through 0.45 µm polypropylene filters and acidified to a pH of 2.0 with 1 : 1 H₂SO₄. The NH₄⁺ was analysed on an ALPKEM Flow Solution™ IV Automated Analyser using the Berthelot reaction according to EPA Method 353.2 (automated nitrate-nitrite determination). Nitrate was measured on the same analyser colorimetrically after its reduction by Cd to NO₂⁻ at pH 7.5, while NO₂⁻ was determined directly without the reduction step. Total Kjeldahl nitrogen (TKN) was determined on approximately 5 mL of leachate, or 5 g of dried biosolids (105°C). Samples were digested in a mixture of sulfuric acid, potassium sulfate and cupric sulfate at 400°C for 1 h to convert amino-nitrogen from most organic materials into NH₄⁺. The digested sample was concentrated by heat distillation into standardized boric acid in a receiving flask. Total N was quantified as NH₄⁺ by nesslerization. Dissolved organic nitrogen (DON) was determined by subtracting the concentrations of NH₄⁺ from TKN.

Dissolved organic carbon (DOC) in the leachate, runoff and ZTP-lysimeter samples was determined using an OI Analytical Model 1010 Total Carbon Analyser. Chemical oxygen demand (COD) was determined using the Hach Method 430/435, Hach COD reactor Model 45600 and a DR/2000 spectrophotometer. Percentage solids were determined gravimetrically after heating the samples for 24 h at 110°C, and loss on ignition (LOI) was determined by heating the samples at 550°C in a muffle furnace for 3 h.

Metal concentrations were measured on an ICP-AES (Perkin-Elmer Optima 3300XL), after sample extracts were filtered (0.45 µm membranes). Standard additions of a stock solution with known metal concentrations were made to a leachate sample to test the matrix effect on the ICP-AES measurements.

Ultrafiltration experiments were performed on the leachate from the lined cells to characterize the metal release mechanisms and speciation. Unacidified samples diluted 1 : 100 with DI water. A Millipore Amicon Model 8200 Stirred Ultrafiltration Cell utilizing cellulose membranes with pore sizes of 10 000 NMWL (10 kDa) and 1000 NMWL (1 kDa) was used. Each 0.45 µm-filtered sample utilized the following operational procedure: 200 mL of diluted filtrate was placed in the ultrafiltration cell under an N₂ atmosphere at 50 psi. The first 100 mL of filtrate passing the 10 kDa membrane was collected and 30 mL was retained for analysis. The remaining

70 mL was placed in the cleaned ultrafiltration cell with a 1 kDa membrane, and the procedure was repeated until 35 mL of filtrate passing the 1 kDa membrane was collected for analysis. All ultrafiltration membranes were flushed with 100 mL of DI water prior to sample addition to remove any residual impurities on the membranes. The ultrafiltration cell was triple rinsed with DI water after each run. Sample collection glassware was acid washed and rinsed with DI water after each sample run. Statistical analyses [analysis of variance (ANOVA)] of solution chemistries for the different levels of the ZPT lysimeter plot were performed using SYSTAT v. 11.

Results and discussion

The biosolids stockpile generated negligible volumes of leachate for the first 10 to 14 days and then leachate flux increased, peaked at approximately 60 days, and thereafter declined gradually (Figure 1). All surface liquid was incorporated into the biosolids or evaporated. The predicted leachate volume was less than the precipitation input. After 3 weeks, leachate volume exceeded precipitation input for the remainder of the experiment. Release of stored liquid from the stockpile was approximately 40 000 L (762 L m⁻³ biosolids) over 100 days, a net increase in total solids from 25.8% to 31.0%.

Leachate chemistry: nutrients and pH

The dominant N species was NH₄⁺ with maximum concentration exceeding 4800 mg L⁻¹. NO₃⁻ and NO₂⁻ concentrations remained below 0.21 and 0.07 mg L⁻¹, respectively, constituting a very small fraction of total N (Figure 1). On average, DON represented approximately 10% of the total N released. All nitrogen species' concentrations increased over time. COD concentrations peaked after day 29 at (37 000 mg L⁻¹) and DOC peaked at day 26 (8900 mg L⁻¹); concentrations declined slowly and steadily for the remainder of the experi-

ment. The dissolved oxygen (DO) concentration measured at the collection point for the auto-sampler was consistently < 0.1 mg L⁻¹.

The measured leachate release rates and concentrations of N species from the experimental stockpile (0.081 ha) were used to calculate the 90 day loading rates (kg ha⁻¹) in comparison with the maximum stockpile size allowed by the MDEP (0.20 ha) (Table 1). Application rates of land-applied biosolids are regulated to account for N demand of crops. When calculating these application rates, the percentage inorganic (mineral) N and availability of organic N in the biosolids were considered. Approximately 90% of the total N in the leachate was mineral. The calculated loading from initial leachate exceeded the agronomic demand at the stockpile by a factor of 66 within the footprint of the stockpile. After 90 days the N loading rose to 145 times the agronomic demand. Based on LOI measures, biosolids organic carbon over the 90 days fell from 27.6% to 24.9% by weight. Carbon loss based on leachate analyses showed a loss of 290 kg, while biosolids LOI changes showed a loss of 350 kg. Nitrogen analyses of the biosolids showed an insignificant decrease in NH₄⁺ (2500 to 2300 mg kg⁻¹), a net decrease in NO₃⁻ + NO₂⁻ (6.7 to 2.5 mg kg⁻¹) and a net decrease in TKN (3.2 to 0.9 wt.%). The biosolids analyses show an N loss of 240 kg whereas leachate analyses show a 145 kg N loss. The discrepancies in C and N mass balances may be attributed to gaseous CO₂ and N₂ formation. Release of CO₂ and N₂ from biosolids-amended soils have been estimated to be between 10 and 25% of the total released from the biosolids (Smith *et al.* 1998a, b, Balint *et al.* 2002; Roelle *et al.* 2002).

Lime was added to the biosolids at the wastewater treatment plant to increase pH to approximately 12. Calcium loss and pH decline in the leachate tracked together with calcium concentrations rising to 1000 mg L⁻¹ near day 20, then rapidly declining to < 100 mg L⁻¹. At the same time leachate pH fell from > 11 to approximately 7.5.

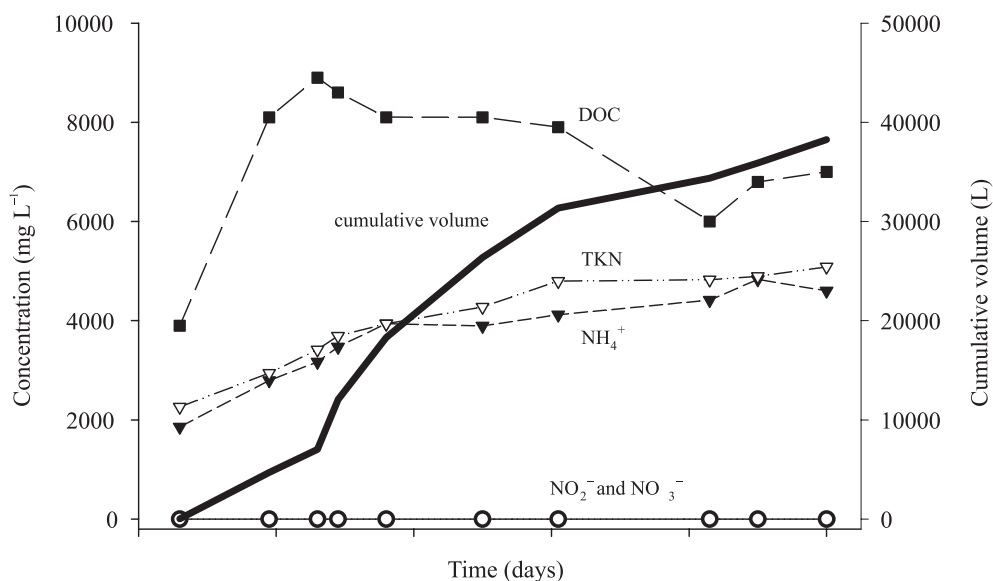


Fig. 1: Lined cell leachate cumulative volume and concentrations of DOC and nitrogen species.

Table 1: Comparison of 90 day nutrient and metal loadings and projected annual loading limits from the lined cell stockpile.

	Calculated 90-day loading (kg ha ⁻¹)	Maximum annual loading (kg ha ⁻¹)	Percentage of allowable annual loading
COD	139246	no standard	–
TOC	33011	no standard	–
TKN	16205	112	14500
NH ₄ ⁺	15605	varies	–
DON	1140	varies	–
NO ₃ ⁻	0.45	varies	–
NO ₂ ⁻	0.13	no standard	–
Arsenic	5.35	0.5	1070
Cadmium	0.031	1.9	1.6
Copper	0.97	75	1.3
Nickel	1.27	20	6.4
Selenium	0.52	5	10.4
Zinc	0.37	140	0.3

Leachate chemistry: trace metals

Time-series trends in concentrations of metals (Al, As, Cd, Cu, Fe, Mn, Ni, Se, and Zn) and P in the leachate of lined cells are shown in Figure 2. The leachate Fe and Mn concentrations peaked at 130 and 17 mg L⁻¹, respectively. It is likely that Fe and Mn were released during microbially-catalysed

reductive dissolution of their respective (oxyhydr)oxides under the anoxic conditions in the interior of the biosolids pile (Lovley & Phillips 1988). The sharp drop in concentrations after 20 days may signal the depletion of available Fe- and Mn-(oxyhydr)oxides (von Gunten & Zobrist 1993, Amirbahman *et al.* 2003), and the Fe concentration rebound after 40

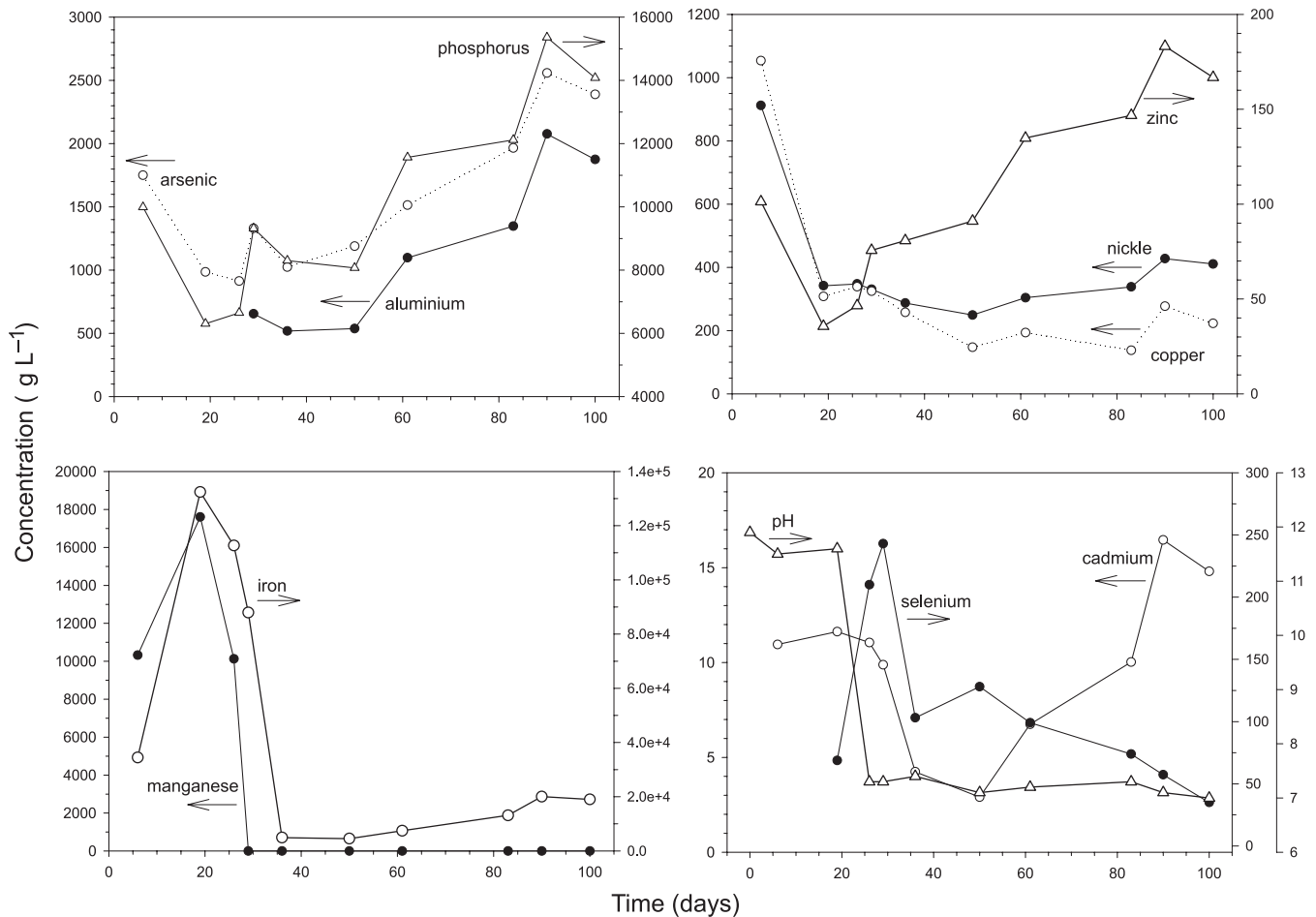


Fig. 2: Lined cell leachate trace metal concentration: 9 July 2003 to 17 October 2003.

Table 2: Leachate ultrafiltration results for lined cell and zero-tension pan (ZTP) lysimeter (June 2003).

Analyte	Lined cell leachate		ZTP lysimeter pan 2-C	
	% passing 10 kDa	% passing 1 kDa	% passing 10 kDa	% passing 1 kDa
Aluminum	87	62	97	66
Arsenic	81	66	93	78
Cadmium	93	82	98	84
Copper	–	–	89	71
Iron	33	3	94	75
Manganese	0	0	95	76
Nickel	86	20	–	–
Selenium	80	67	99	99
Zinc	96	48	98	86
DOC	94	92	82	74

days may be due to the slow dissolution of the more crystalline Fe-containing minerals. The pH decrease to a circumneutral value coincided especially with the drop in Mn concentration. Mn minerals are quite insoluble at both alkaline and circumneutral pH values regardless of the redox potential (Krauskopf & Bird 2003), and as such, it is unlikely that changes in biosolid pH as reported here would result in significant mobilization or removal of Mn. Furthermore, the precipitation of rhodochrosite ($\text{MnCO}_{3(s)}$) is favoured at high pH, whereas in this study, Mn was mobile at a high pH (Figure 2).

Ultrafiltration showed that most of the Fe was colloidal (> 10 kDa) (Table 2). The Fe mobilization mechanism probably involved the reductive dissolution of Fe(III)-(oxy)hydroxides by Fe-reducing microbes under an anaerobic environment. Following its release, Fe(II) was oxidized under an oxic environment, and in the presence of a high DOC concentration, formed stable colloids coated with organic matter (von Gunten & Schneider 1991, Amirbahman & Ohlsen 1993).

Leachate Al, As, P, Zn and Cd concentrations were stable for 50 days, and then increased so that by 100 days, As, Al, P, Zn and Cd concentrations were nearly 2.5, 2, 8, 0.2 and 0.015 mg L⁻¹, respectively (Figure 2). These concentrations do not directly correlate with Fe and Mn, and therefore, it is unlikely that their mobilization was directly related to colloidal Fe and Mn mobilization. This is also in agreement with the ultrafiltration results (Table 2) as discussed below. Considering the relatively strong affinity of these species especially to the Fe(III) (oxy)hydroxide surface (Dzombak & Morel 1990), this lack of correlation with Fe may be attributed to the strong interaction of DOC with Fe colloids that covers their reactive surface sites, and complexation of the species with DOC. The lack of correlation between the abrupt pH drop between 20 and 30 days (Figure 2) and the release of these species also suggests that the pH drop was not directly responsible for the metal release from the biosolids. One possible mechanism for the Al, As, P and Zn release may be organic matter mineralization.

Aluminium was associated with the non-colloidal size range, < 10 kDa (Table 2). However, considering the solubil-

ity of gibbsite ($\text{Al}(\text{OH})_{3(s)}$), free Al concentration at a circumneutral pH should be < 1 µg L⁻¹ (Stumm & Morgan 1996), and therefore, Al was probably complexed by the fulvic acid or the low molecular weight organic acid fractions of DOC. The presence of low molecular weight organic acids was especially important in this study, since 92% of DOC was < 1 kDa (Table 2). Zn, Cd and As were also largely in the non-colloidal fraction < 10 kDa (Table 2); association of metal cations such as Zn and Cd with DOC is well-documented (Tipping 2002). Redman *et al.* (2002) and Lin *et al.* (2004) observed As complexation with DOC, especially in the presence of bridging cations such as Fe. Therefore, the As in the leachate may have been truly dissolved or associated with DOC. P was also not expected to be associated with the Fe and Mn colloids as their respective concentrations did not correlate (Figure 2). Soluble P, however, may possibly have a direct microbial association and be part of large biomolecules or associated with the DOC via bridging cations.

Concentrations of Cu and Ni were approximately 1 mg L⁻¹ on day 6 and then 0.4 mg L⁻¹ for the duration of the experiment. Se concentration remained stable at approximately 0.17 mg L⁻¹ until day 25 after which it declined to approximately 0.03 mg L⁻¹ at 100 days. Jensen & Christensen (1999) studied the physical size and colloidal nature of metals found in landfill leachate and observed that nearly 80–95% of Cd, Cu, K, Mg, Na, Ni, Pb and Zn were < 1 kDa; nearly half of Ca and Mn were < 1 kDa; and Fe was 85% > 1 kDa, which is consistent with data reported here.

All of the regulated metal concentrations in leachate were below Maine's annual loading limits with the exception of As at 90 days. Extrapolating linearly from day 90 to day 240 (8 months, maximum allowable field stacking duration), all of the regulated metals listed in Table 2, with the exception of As, would still be below the annual loading limits.

ZTP-lysimeter: dissolved carbon and nitrogen species

The average concentrations of N species and DOC at various ZTP-lysimeter depths varied markedly for sample locations at 30, 60 and 90 cm depths (Table 3). The June 2003 samples

Table 3: Zero-tension pan lysimeter filtrate test results (June 2003).

Lysimeter ^a	DOC (mg L ⁻¹)	COD (mg L ⁻¹)	NO ₂ ⁻ (mg L ⁻¹)	NO ₃ ⁻ (mg L ⁻¹)	NH ₄ ⁺ (mg L ⁻¹)	TKN (mg L ⁻¹)
30-centre-1	67	1810	0.074	N.D.	167	223
30-centre-2	725	6520	0.102	N.D.	600	715
30-centre-3	1270	12660	0.218	N.D.	2400	2590
30-edge-1	79	120	N.D.	N.D.	23.5	38.8
30-edge-2	2030	20225	0.552	0.036	1960	2420
60-centre-1	4860	26700	0.443	0.031	2384	2840
60-centre-2	301	2690	0.062	N.D.	257	358
60-centre-3	55	384	0.012	N.D.	87.8	112
60-edge-1	6060	16075	0.036	N.D.	1340	3030
60-edge-2	76	1680	0.031	N.D.	149	198
90-centre-1	24	17	0.802	23.7	0.34	27.2
90-centre-2	270	3480	N.D.	N.D.	342	468
90-centre-3	54	246	12.5	8.8	0.88	24.6
90-edge-1	39	54	1.0	43.1	1.19	52.5
90-centre-2	37	422	N.D.	0.012	22.8	37.3

^aLysimeters are identified by depth (cm)-location under the stockpile-replicate number.

were collected after the removal of the first stockpile that was in place for 6 months, and before the emplacement of the second stockpile, which lasted from July 2003 to May 2004. The second and third samples were collected in September 2003 and May 2004, when the second stockpile was in place, and the fourth sample was collected in July 2004, 2 months after the removal of the second stockpile.

Average DOC concentrations at 30 and 60 cm depths ranged from 1000 to 4000 mg L⁻¹. DOC concentrations increased markedly at all three depths between June and September 2003, which was probably due to the fresh biosolids stockpile emplaced in July 2003. The DOC, on average, increased steadily with time at a depth of 90 cm. The DOC concentrations at 60 cm were consistently higher than at 30 cm, which may be due to differences in organic solubility or biological activity at shallower locations. The decline in DOC at 30 and 60 cm was perhaps attributable to the passing of a DOC front and the loss of degradable DOC.

Average NH₄⁺ concentrations ranged from 589 to 1030 mg kg⁻¹ at 30 cm with no clear temporal trend. The ZTP-lysimeter data at 60 and 90 cm, however, showed a general increase in average NH₄⁺ concentration over time. NO₃⁻ concentrations were close to the detection limit of 0.01 mg L⁻¹ at 30 and 60 cm depths prior to the emplacement of the biosolids stockpile. Only one sample collected at the beginning of the experiment had detectable NO₃⁻.

The variances observed by depth and ZTP-lysimeter location for the individual filtrates (Table 3) were not homogeneous and a standard ANOVA could not be applied. A non-parametric one-way ANOVA (Kruskal-Wallis method) was performed instead and NH₄⁺ was found to vary significantly by depth ($\alpha < 0.05$). There were no differences between pans located below the centre or edges of the pile. The significant

differences in concentrations seen at the same depth may be attributed to the existence of preferential flow paths. The installation of pan ZTP-lysimeters disturbs the existing soil structure, possibly creating preferential flow paths. Subsurface flow may also be restricted locally by biological growth plugging the open pores and pathways in the soil, as has been reported in landfill leachate column studies (Jensen & Christensen, 1999). The existence of preferential flow paths was also suggested by the greatly varying yield of liquid from the ZTP-lysimeters at the three different depths. ZTP-lysimeters at the 60 and 90 cm depths in particular failed to yield as much liquid as ZTP-lysimeters at the 30 cm depth. Preferential flow paths are commonly found in nature and have been documented to accelerate biosolids leaching into soil (Cambreco *et al.* 1996). The variability observed in June 2003 (Table 3) was representative of all sample dates; N species and DOC concentrations declined with depth but increased with time.

Soils tested before and after stockpiling on the ZTP-lysimeter plot showed that pH increased from 5.7 to 7.3, LOI decreased from 6.4 to 5.3% (w/w), NH₄⁺ increased from 3.9 to 824 mg kg⁻¹, and NO₃⁻ decreased from 6.9 to 0.6 mg kg⁻¹. It is clear that stockpiling of the biosolids elevated the pH and NH₄⁺ content in the soil. NO₃⁻ concentration declines were probably due to denitrification. The increase in soil pH was attributed to the lime added to the Class B biosolids utilized in this experiment

ZTP-lysimeter: metals

ZTP-lysimeter results for metals are presented for one sampling round for each of the 15 individual ZTP-lysimeter pans sampled in June 2003 (Table 4). As in the nutrients, spatial variability obscured vertical trends. Maximum concentrations

Table 4: Zero-tension pan lysimeter trace metal concentrations June 2003.

Depth	Analyte Al ($\mu\text{g L}^{-1}$)	As ($\mu\text{g L}^{-1}$)	Cd ($\mu\text{g L}^{-1}$)	Cu ($\mu\text{g L}^{-1}$)	Fe ($\mu\text{g L}^{-1}$)
30 cm	1280 \pm 1230	99.9 \pm 123	16.6 \pm 26	157 \pm 113	20080 \pm 23000
60 cm	1270 \pm 1280	101 \pm 103	5.17 \pm 5.5	222 \pm 172	27500 \pm 38600
90 cm	243 \pm 218	60.7 \pm 38	13.5 \pm 15	1080 \pm 2270	151 \pm 226
Depth	Analyte Mn ($\mu\text{g L}^{-1}$)	Ni ($\mu\text{g L}^{-1}$)	P ($\mu\text{g L}^{-1}$)	Se ($\mu\text{g L}^{-1}$)	Zn ($\mu\text{g L}^{-1}$)
30 cm	3640 \pm 3890	190 \pm 200	9090 \pm 11700	171 \pm 155	68.4 \pm 65
60 cm	7704 \pm 14000	227 \pm 258	5430 \pm 11700	149 \pm 110	89.7 \pm 96
90 cm	89.9 \pm 200	26.3 \pm 62	671 \pm 865	86.0 \pm 60	310 \pm 476

Results presented as mean \pm 1 standard deviation.

of Fe, Mn, P and Al were in the milligrams per litre range whereas all other metals were present at less than 1 mg L^{-1} (Table 4). Concentrations were generally lowest in the deepest ZTP-lysimeter. P concentrations increased slightly with time. The P concentrations were greatest at the 30 cm depth, suggesting that biosolids were the source and that underlying soil or microorganisms act as a sink for P. Arsenic concentrations increased over time and depth, with the highest single concentrations at 90 cm. Cu and Zn concentrations increased in September 2003, possibly due to the emplacement of a fresh biosolid stockpile in July 2003. Like As, Cu concentrations were highest at 90 cm suggesting that increases of these metals with depth is due to mechanisms such as dissolution of native mineral oxides. Dissolution of Fe(III) (oxy)hydroxides and subsequent release of As from a landfill-contaminated aquifer has been previously reported (Welch & Coleman 2003). Soil extractions before the emplacement of the biosolid stockpile showed total As and Cu concentrations of 6.1 and $14.3 \mu\text{g g}^{-1}$, respectively. Considering a solid density of 2.6 g cm^{-3} and porosity ranging between 0.3 and 0.5, the soil contained two to three orders of magnitude more As and Cu than observed in the ZTP-lysimeters so that some of the ZTP-lysimeter results may include metal released from the underlying soil. Concentrations of Cd and Ni ranged between 5–20 and 20–250 $\mu\text{g L}^{-1}$, respectively, and remained stable over time (data not shown here). The one-way non-parametric analysis of variance indicated significant differences ($\alpha < 0.05$) between concentrations of Fe, Mn, and P by ZPT-lysimeter level.

Ultrafiltration of samples from ZTP-lysimeter 2C in June 2003 showed that the metals were mostly $< 10 \text{ kDa}$, and were in truly dissolved form or complexed with DOC. DOC complexation was particularly suggested in the 1 and 10 kDa fractions. This size range held 31% of the Al, while 66% of the Al was $< 1 \text{ kDa}$. Since 74% of the DOC was $< 1 \text{ kDa}$, Al must be complexed with the DOC (Table 3). The loss of the smaller DOC size fractions in the ZTP-lysimeter was similar to the DOC loss observed in the lined-cell leachate (92% $< 1 \text{ kDa}$; Table 2). This may be due to the preferential consumption of the low molecular weight organic acids by soil micro-organisms or to DOC polymerization over time. In contrast to

mainly colloidal Fe and Mn in the lined-cell leachate (where there was an abrupt transition from anoxic to oxic conditions that can promote DOC-coated metal colloids). Fe and Mn in the ZTP-lysimeters were mostly in the size fraction $< 10 \text{ kDa}$.

Conclusions

Leachate from field-stacked biosolids contained significant concentrations of DOC and N, well in excess of typical agronomic demands. NH_4^+ was the dominant species of N, and organic N, as a minor species, represented approximately 10% of the total dissolved N. Both species contribute to the formation of NO_3^- in soil and groundwater. The loss of mineral N from biosolids stockpiles exceeded agronomic demand and will likely lead to N saturation in the soil near the stockpile with subsequent losses of NH_4^+ , NO_3^- and NO_2^- into groundwater (Jacobsen 1998, Smith *et al.* 1998a, b, Roelle *et al.* 2002). In addition, P is mobilized from the biosolids in the low mg L^{-1} concentration range.

The results of this study also indicate that Al, Fe, Mn, and As are released from a lime-stabilized Class B biosolids stockpile within 90 days of placement. Metal analyses showed that As concentrations exceeded state annual soil loading limits. Other regulated metals (Cd, Cu, Ni, Se and Zn) were also released within 90 days but at rates below the allowable limits. ZTP-lysimeter data showed that As, Cu, P and Se from the biosolids were mobile through the soil beneath the stockpile. Further, native soil-bound As and Cu were evidently mobilized downward in the soil. Other analytes, such as P and Se showed a general decrease with depth, suggesting that they directly originated from the biosolids. Ultrafiltration of the lined-cell leachate indicated that Fe and Mn were largely in colloidal form ($> 10 \text{ kDa}$), whereas other analytes were predominantly complexed by DOC or truly dissolved. Ultrafiltration of the ZTP-lysimeter leachate indicated that all metals, including Fe and Mn were predominantly complexed by the low molecular weight DOC, or truly dissolved ($< 1 \text{ kDa}$). The results of this study show that the current practice of allowing stockpiling of Class B biosolids on bare ground for up to 8 months should be re-examined to better assess the potential impact that N and trace metal leaching may have on long-term groundwater quality.

Acknowledgements

This project was funded by the Maine Department of Environmental Protection and the Maine Water Resources Research Institute with support and collaboration of the Department of the Interior, US Geological Survey and the University of Maine, under Grant No. 01HQGR0083. The views and con-

clusions contained in this document are those of the authors and should not be interpreted as necessarily representing the official policies, either expressed or implied, of the US Government, or the Maine Department of Environmental Protection. We thank Justin Jamieson, Ken Johnson and Liz Dyzek for their help in the field and laboratory.

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