

# On-Farm Assessment of Biosolids Effects on Soil and Crop Tissue Quality

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## ABSTRACT

**Agronomic use of biosolids as a fertilizer material remains controversial in part due to public concerns regarding the potential pollution of soils, crop tissue, and ground water by excess nutrients and trace elements in biosolids. This study was designed to assess the effects of long-term commercial-scale application of biosolids on soils and crop tissue sampled from 18 production farms throughout Pennsylvania. Biosolids application rates ranged from 5 to 159 Mg ha<sup>-1</sup> on a dry weight basis. Soil cores and crop tissue samples from corn (*Zea mays* L.), soybean (*Glycine* spp.), alfalfa (*Medicago sativa* L.), orchardgrass (*Dactylis* spp.) hay, and/or sorghum [*Sorghum bicolor* (L.) Moench] were collected for three years from georeferenced locations at each farm. Samples were tested for nutrients, trace elements, and other variables. Biosolids-treated fields had more post-growing season soil NO<sub>3</sub> and Ca and less soil K than control fields and there was some evidence that soil P concentrations were higher in treated fields. The soil concentrations of Cu, Cr, Hg, Mo, Mn, Pb, and Zn were higher in biosolids-treated fields than in control fields; however, differences were ≤0.06 of the USEPA Part 503 cumulative pollutant loading rates (CPLRs). There were no differences in the concentrations of measured nutrients or trace elements in the crop tissue grown on treated or control fields at any time during the study. Commercial-scale biosolids application resulted in soil trace element increases that were in line with expected increases based on estimated trace element loading. Excess NO<sub>3</sub> and apparent P buildup indicates a need to reassess biosolids nutrient management practices.**

**A**GRONOMIC USE OF BIOSOLIDS remains controversial, despite widespread adoption of the practice during the past 20 years. Concerns include the contamination of surface and ground waters due to losses of N and P, as well as soil accumulation and plant uptake of trace elements that could have adverse effects on humans, animals, or crops. The USEPA promulgated the Standards for the Use and Disposal of Sewage Sludge (USEPA, 1993) to protect human health and the environment. These regulations, commonly referred to as “Part 503,” are based on scientific risk assessment and establish sludge quality limits, pathogen reduction methods, and limits on the amounts of trace elements that can be applied to a site. The Part 503 regulations require monitoring of trace element additions to soil; however, they do not require continued testing of soils to which biosolids have been applied, soil testing at depths below the plow layer, or soil nutrient analysis. There is also no requirement for the sampling of crop tissue grown on treated soils for major plant nutrients or trace elements. Consequently, the long-term effects of agronomic bio-

solids use on soil and crop quality have rarely been monitored in nonresearch situations.

Most U.S. states, including Pennsylvania, stipulate that application of biosolids to agricultural land must be done at agronomic rates that are most commonly based on the N needs of the crop to be grown. Application rates are based on inorganic N content plus expected mineralization of organic N because most biosolids N is organic. When organic N is mineralized and nitrified it can, if not taken up by a crop, be lost as NO<sub>3</sub> through leaching or surface runoff. This could contribute to eutrophication of surface waters or to human health concerns (blue-baby syndrome). Smith (1996) maintained that the risk of excessive NO<sub>3</sub> leaching from soils receiving long-term biosolids application is small, as long as the N needs of the crop are not exceeded. Long-term biosolids applications may lead to increases in soil P above optimum values (Kelling et al., 1977; Peterson et al., 1994) since N-based application will supply P in excess of crop needs (Smith, 1996; Stehouwer et al., 2000; Pierzynski, 1994). High soil P values increase the possibility that runoff or erosion losses could contaminate surface waters with P and contribute to eutrophication (Chaussod et al., 1986; Sharpley et al., 1996; Sims et al., 1998, 2000).

The Part 503 regulations identify several trace elements present in biosolids that could pose health risks to humans (As, Cd, Hg, Pb, Se, and Zn), plants (Cu, Ni, and Zn), or animals (Mo) (Page and Chang, 1994). When compared with control soils, it has been determined that trace elements accumulate in the plow layer of biosolids-amended soils (McGrath and Cegerra, 1992). Many long-term studies have indicated that trace elements remain in the plow layer in forms relatively unavailable to crops (McGrath, 1987; McGrath and Lane, 1989; Chaney and Ryan, 1993). However, there is evidence of some accumulation of trace elements at depths below the zone of biosolids incorporation (Campbell and Beckett, 1988; Dowdy et al., 1994).

Agronomic biosolids management practices and regulations are based on an extensive body of scientific research concerning transformations, availability, fate, and transport of biosolids nutrients and trace elements. This literature has been reviewed by Smith (1996) and we will not attempt to review it here. Almost all of this research has been conducted using experiments where parameters such as soil type, biosolids source and quality, and biosolids application methods, rates, and timing are carefully controlled. Such controls are absent when biosolids are actually used in production agriculture as a substitute for traditional nutrient or lime inputs. Furthermore, when biosolids are used for crop production, they become one component of a complex farm manage-

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**Abbreviations:** CPLR, cumulative pollutant loading rate.

**Table 1. County locations and soil series and taxonomic names for soils sampled at the farms included in this study.**

County	Soil series†	Taxonomic name
Beaver	Wharton (NB)	fine-loamy, mixed, active, mesic Aquic Hapludults
Berks	Gilpin (B)	fine-loamy, mixed, active, mesic Typic Hapludults
Blair	Athol	fine-loamy, mixed, active, mesic Ultic Haplualfs
Centre	Hublersburg	clayey, illitic, mesic, Typic Hapludults
Columbia	Hagerstown	fine, mixed, semiactive, mesic Typic Hapludalfs
Cumberland	Allenwood	fine-loamy, mixed, semi-active, mesic Typic Hapludults
Dauphin	Duffield	fine-loamy, mixed, active, mesic, Ultic Hapludalfs
Franklin	Hagerstown	fine, mixed, semiactive, mesic Typic Hapludalfs
Fulton	Weikert (NB)	loamy-skeletal, mixed, active, mesic Lithic Dystrudepts
Juniata	Bedington (B)	fine-loamy, mixed, active, mesic Typic Hapludults
Lancaster 1	Hagerstown	fine, mixed, semiactive, mesic Typic Hapludalfs
Lancaster 2	Edom	fine, illitic, mesic Typic Hapludalfs
Lebanon	Bedington	fine-loamy, mixed, active, mesic Typic Hapludults
Northampton	Manor	coarse-loamy, micaceous, mesic Typic Dystrudepts
Schuylkill	Berks	loamy-skeletal, mixed, active, mesic Typic Dystrudepts
	Washington	fine-loamy, mixed, mesic, Ultic Hapludalfs
	Berks (B)	loamy-skeletal, mixed, active, mesic Typic Dystrudepts
Susquehanna	Watson (B/NB)	fine-loamy, mixed, active, mesic Typic Fragiudults
Union	Shelmadine (NB)	fine-loamy, mixed, mesic Typic Fragiaquults
	Volusia	fine-loamy, mixed, active, mesic, Aeric Fragiaquults
York	Hagerstown	fine, mixed, semiactive, mesic Typic Hapludalfs
	Elliber	loamy-skeletal, mixed, semiactive, mesic Typic Hapludults
	Glenelg	fine-loamy, mixed, semiactive, mesic Typic Hapludults

† The term B indicates treated (biosolids) field, while NB indicates control (no biosolids) field.

ment system. The goal of our research was to assess the effects of repeated biosolids use on soil and crop quality within the context of production agriculture systems. Our objective was to determine if repeated substitution of biosolids for traditional inputs has resulted in measurable differences in soil and crop concentrations of nutrients and trace elements, and in soil pH, organic C, and acidity. We intentionally included a broad range of soils, crops, biosolids, and farming systems in the study to provide a comprehensive assessment of agronomic biosolids use.

## MATERIALS AND METHODS

Eighteen production farms with a history of repeated commercial agricultural biosolids application were selected for inclusion in the study. At each farm, a field that received biosolids (treated) was paired with a field that never received biosolids (control). The paired fields were selected to be similar with respect to soil type, crops grown, and management history. At most sites the paired fields were adjacent with similar soil series and similar management practices; however, at the Franklin and Schuylkill farms soils were not from the same series (Table 1). The pairing of fields was necessary because time zero background samples (or data) were not available for most of the treated fields. Our assumption was that before the use of biosolids, the soil and crop quality parameters we measured were similar in both members of a pair because field management was similar for each pair. For example, at farms that use manure, the field management practices for both fields were similar except that in some years biosolids were substituted for manure. In years when biosolids was not applied, it is likely that if the control field received manure, the biosolids-treated field also received manure. We were not attempting to measure absolute change in the variables, but rather to measure any differences between paired fields resulting from the use of biosolids as an agronomic input. Pairing was also the basis for all statistical testing as is described in detail below.

Sample collection began in 1998 from 13 farms in 13 Pennsylvania counties. Five more sites were added the following year for a total of 18 farms in 17 counties sampled in 1999

and 2000. The average amount of biosolids applied in this study was 53.71 Mg ha<sup>-1</sup> (dry matter) for the history (2–18 yr) of the sites. Application amounts ranged from 5.06 Mg ha<sup>-1</sup> at the Juniata County site, to 159.04 Mg ha<sup>-1</sup> at the Fulton County site (Table 2). Soil samples were collected in the fall from five georeferenced locations within each field. Georeferencing allowed us to collect samples from the same locations ( $\pm 1$  m) in each year of the study. Soil samples were collected at three depth increments (0 to 10, 10 to 20, and 20 to 40 cm) with a 5-cm bucket auger. Approximately 2.5 cm of soil was removed from the top of the bucket auger when soils were collected from the 10- to 20- and 20- to 40-cm depths to eliminate sample contamination with soil that may have fallen from higher in the profile. The soil samples were air-dried and ground to pass a 2-mm sieve. Before chemical analysis, the five samples taken within each field were composited within depth increments to create one sample per field per depth.

Available NH<sub>4</sub> and NO<sub>3</sub> were extracted from 1 g of soil with 10 mL of 2 M KCl as described in Griffin (1995). The extracts were analyzed using a Technicon Auto-Analyzer II (Technicon Industrial Systems, Tarrytown, NY). The Mehlich 3–extractable P, K, and Ca were determined using the method described in Wolf and Beegle (1995). Total and inorganic carbon were analyzed using a TOC-5000A total organic carbon analyzer with the SSM-5000A solid sample module (Shimadzu, Kyoto, Japan). Organic carbon was determined by subtracting the amount of inorganic carbon from the amount of total carbon. Soil pH was determined in a 1:1 (v/v, soil to water) suspension using a pH electrode and acidity was determined using the SMP buffer method (Eckert and Sims, 1995). Soil samples were analyzed for Hg using USEPA Method 7471 and digests were analyzed using an AS-90 flow injection mercury system (PerkinElmer, Wellesley, MA). Soil samples were also digested for total extractable elements by USEPA Method 3050B (USEPA, 1986) and digests were analyzed for Cd, Cr, Cu, Pb, Mn, Mo, Ni, and Zn using a Thermo Jarrell Ash 61E inductively coupled plasma atomic emission spectrometer (Thermo Elemental, Franklin, MA) and for As and Se using a Zeeman 5100 graphite furnace (PerkinElmer).

Crop tissue samples were collected at each of the five georeferenced soil sampling locations in each treated and control field. The tissue collected was representative of the portion

**Table 2. Crop rotations, farm type, biosolids type, cumulative biosolids application, and years in which biosolids were applied.**

Site	Crops†	Manure used?	Biosolids stabilization‡	Amount of biosolids applied Mg ha <sup>-1</sup>	Years that biosolids were applied
Beaver	A	yes	anaerobic	101.49	1984–2001
Berks	C	yes	anaerobic	11.92	1994–1998
Blair	C	yes	anaerobic	20.81	1991–1996, 1997–2000
Centre	C, A	yes	anaerobic	15.61	1991, 1993, 1999
Columbia	C, S	no	lime	37.90	1995–1998
Cumberland	C	yes	lime	40.36	1986, 1989, 1997, 2000
Dauphin	M	yes	anaerobic	47.04	1980, 1981, 1984, 1990, 1991, 1996–1998
Franklin	C, S	no	anaerobic	42.22	1988, 1991–1995, 1997
Fulton	C, A	yes	anaerobic	159.04	1986–1999
Juniata	C	no	anaerobic	5.06	1997, 1998
Lancaster 1	C, S	yes	anaerobic	108.26	1986–1995, 2000
Lancaster 2	A	yes	anaerobic	118.23	1990–1994, 1996, 1999, 2000
Lebanon	C, S	no	anaerobic	12.10	1996, 1998
Northampton	C	no	aerobic	10.24	1997–1999
Schuylkill	C, S	no	anaerobic	50.18	1992, 1994–1997, 2000
Susquehanna	H	no	anaerobic	99.79	1982–1995, 1995, 1997–2000
Union	C, S	yes	anaerobic	16.13	1994–1997, 1999
York	C, S	no	lime	64.33	1988–1993, 1995–1997

† A, alfalfa; C, corn; H, hay; M, corn, sorghum, and sorghum–sudangrass mix; S, soybean.

‡ Method of biosolids stabilization: anaerobic or aerobic digestion and lime stabilization.

of the crop that would be harvested by the farmer. Crop samples were dried to a constant mass at 70°C and ground to pass a 2-mm sieve. Total crop N was determined by the combustion method (Campbell, 1991; Pella, 1990) using an NA1500 elemental analyzer (Fisons Instruments, Beverly, MA). The dry ash method (Dahlquist and Knoll, 1978) was used to destroy organic matter in the crop tissue samples. Dilute nitric acid was then added to dissolve the ash and the resulting solutions were analyzed for P, K, Ca, Cd, Cu, Pb, Mn, Mo, Ni, and Zn using inductively coupled plasma–atomic emission spectroscopy (ICP–AES).

### Data Analysis

A strip-plot design with time as a repeated measure (replicate) was used to analyze treatment effects on variables over the 3-yr course of the study. Data were analyzed using SAS software (SAS Institute, 1999). Data were evaluated to determine overall treatment effects on each variable. Data were unbalanced due to sampling of fewer farms in 1998 requiring the use of sequential sums of squares for hypothesis testing. The farm × treatment interaction was used as the error term to test for treatment effect. If there was a significant treatment × year interaction, the variable could not be analyzed in this fashion. In this case a replicated measurement *t* test design was used to determine significance of the variable for each individual year of the study (Steel and Torrie, 1980). The difference between the treated and control fields was determined for each variable at each site. The mean of these differences was then taken to determine the average difference between treated and control fields for all sites. The mean of the differences was compared with the variance of the paired differences using a significance level of  $\alpha = 0.10$ .

Soil trace element increases were converted from concentration units (mg kg<sup>-1</sup>) to mass per area units (kg ha<sup>-1</sup>) to allow comparison with the USEPA Part 503 CPLRs. Such conversion has previously been done by assuming a biosolids mixing depth of 15 cm and a soil mass of  $2 \times 10^6$  kg ha<sup>-1</sup> 15 cm<sup>-1</sup>, which is equivalent to a bulk density of 1.33 g cm<sup>-3</sup> (USEPA, 1995). Since we had no bulk density measurements, we used a bulk density of 1.33 g cm<sup>-3</sup> to convert concentration increases into mass increases for each of the three soil depth increments. The trace element mass increase in each depth increment was summed to give the total increase and then expressed as a fraction of the CPLR.

## RESULTS AND DISCUSSION

### Soil Quality Parameters

Soil chemical variables that are considered to be basic indicators of agricultural soil quality include soil organic carbon, pH, acidity, and plant-available macronutrients (NH<sub>4</sub>, NO<sub>3</sub>, P, K, Ca, and Mg) (Doran and Parkin, 1994). When soil amendments are used that could alter soil concentrations and bioavailability of micronutrients and other trace elements, these should also be considered as indicators of soil quality (Sims and Pierzynski, 2000). Our on-farm assessment showed that biosolids use affected several of these chemical parameters.

### Carbon

Combined analysis of all years of data showed that differences between control and treated fields for organic and total carbon were not significant (Table 3). Data from individual years and depths showed that measured differences were small and inconsistent. Because most biosolids generated in Pennsylvania typically contain 60 to 65% organic matter (Stehouwer, 1999b), an increase in soil carbon was expected due to repeated biosolids application. Two factors probably account for the absence of carbon increases in this study. First, the addition of biosolids stimulates soil microbial activity and much of the added organic matter may be rapidly mineralized. Second, the application of manure to some of the fields during years that biosolids were not applied added organic matter, confounding any effect of biosolids on soil organic C levels.

### Nitrate and Ammonium

Combined analysis of all three years of data showed that differences in soil NH<sub>4</sub>-N were not significant (Table 3). Accumulation of soil NH<sub>4</sub> in treated fields was not expected since NH<sub>4</sub> concentration in biosolids is relatively low. Also, NH<sub>4</sub> incorporated into the soil is readily converted into NO<sub>3</sub> by nitrifying bacteria or, when biosolids are not incorporated, lost due to ammo-

**Table 3. Mean background concentrations (control) and magnitude of change ( $\Delta$ ) for soil pH, C, and nutrients.**

Depth	1998		1999		2000		Three years	
	Control	$\Delta$	Control	$\Delta$	Control	$\Delta$	Control	$\Delta$
<b>cm</b>								
<b>pH (unitless)</b>								
0–10	6.52	0.12	6.50	0.11	6.54	0.24†	6.52	0.16
10–20	6.61	0.12	6.52	0.14	6.54	0.19	6.55	NA‡
20–40	6.68	0.21	6.58	0.14	6.71	0.07	6.65	0.13
<b>mg kg<sup>-1</sup></b>								
<b>Ca</b>								
0–10	1639	867†	1648	704†	1880	602†	1731	710†
10–20	1602	586†	1560	658†	1872	277	1686	499†
20–40	1375	390	1306	460†	1633	132	1444	321†
<b>NO<sub>3</sub>-N</b>								
0–10	8.78	12.54†	14.87	11.34†	10.74	2.47	11.74	8.40†
10–20	6.63	11.03†	15.38	9.62†	9.02	1.92	10.72	7.16†
20–40	3.40	9.87†	8.37	9.24†	5.30	5.62†	5.92	8.08†
<b>NH<sub>4</sub>-N</b>								
0–10	11.24	-1.77	4.71	1.13	5.23	-0.78†	6.63	-0.34
10–20	7.87	0.46	3.61	0.89†	4.46	0.06	5.05	0.47
20–40	5.11	0.00	2.86	-0.09	3.40	0.08	3.65	0.00
<b>P</b>								
0–10	243.9	13.8	140.0	53.7†	108.8	68.3†	156.1	NA
10–20	194.6	9.8	112.1	18.8	92.5	28.6	126.8	20.0
20–40	90.9	-11.4	53.6	-5.8	40.3	2.7	58.6	-4.2
<b>K</b>								
0–10	176.3	-14.7	184.2	-25.4†	162.4	-32.6†	174.1	-25.2
10–20	114.3	-2.4	109.3	-15.6†	118.4	-21.6†	114.0	-14.3
20–40	92.9	-8.4	88.6	-7.8	89.8	-10.2	90.2	-8.8
<b>Organic C (<math>\times 10^4</math>)</b>								
0–10	1.94	-0.03	2.17	-0.14	2.30	0.07	2.16	-0.03
10–20	1.43	0.11	1.54	0.12	1.79	0.04	1.60	0.09
20–40	0.74	-0.02	0.75	0.21†	0.98	0.05	0.83	0.09
<b>Total C (<math>\times 10^4</math>)</b>								
0–10	1.94	-0.02	2.18	-0.14	2.31	0.07	2.16	-0.03
10–20	1.44	0.11	1.55	0.11	1.79	0.05	1.61	0.09
20–40	0.75	-0.01	0.77	0.20†	0.99	0.05	0.85	0.09

† Significant at the  $\alpha = 0.10$  level.‡ Three-year change is not available due to significant treatment  $\times$  time interaction. All three years were analyzed separately.

nia volatilization (Smith, 1996; McFarland, 2001; Robinson and Polglase, 2000).

In contrast, combined analysis of all three years of data indicated that the concentration of soil NO<sub>3</sub>-N was higher in biosolids-treated fields at all depth increments (Table 3). Analysis of individual years showed that in only one of nine year-depths, the increase in NO<sub>3</sub> not significant. While the magnitude of NO<sub>3</sub> increase varied from year to year, overall, the amount of post-growing season NO<sub>3</sub> in biosolids-treated fields was on the order of twice that in nonbiosolids fields. Excess soil NO<sub>3</sub> increases the likelihood of loss to surface and ground water and ultimately increases the risk of environmental problems such as eutrophication. Excess post-growing season NO<sub>3</sub> could result from any of several factors that this study was not designed to elucidate. One possibility is that mineralization constants used to predict plant-available N are too low, resulting in overapplication of biosolids (Pierzynski, 1994; Smith, 1996). The excess NO<sub>3</sub> could also result from mineralization after crop uptake of N ceases. Whenever there is sufficient soil moisture and soil temperatures are above freezing, N mineralization can occur, leading to accumulation of nitrate in the soil and significant nitrate leaching during the nongrowing season (McFarland, 2001). Both of these factors indicate a need to reassess methods used

to determine N-based agronomic loading of biosolids and the need to implement N conservation practices such as planting cover crops that will take up excess NO<sub>3</sub>-N and reduce NO<sub>3</sub> leaching.

### Available Phosphorus and Potassium

Many of the fields sampled in this study, both treated and control, had soil test P (Mehlich 3) levels above the optimum range for crop growth. Significant increases were noted in the surface soils in 1999 and 2000 biosolids-treated fields, which had approximately 50 mg kg<sup>-1</sup> more Mehlich 3 P than control fields (Table 3). Biosolids did not increase P at depths below 0 to 10 cm. This agrees with Maguire et al. (2000), who found higher concentrations of P where biosolids were applied, with the highest concentrations in the top 5 cm. Some of the P buildup in our study is undoubtedly due to repeated applications of dairy manure on both fields at many of the farms in the study. When applied at an N-based rate, both manure and biosolids will add P in excess of crop removal. For example, Pierzynski (1994) calculated that applying biosolids that contained 1.5% plant available nitrogen (PAN) and 1.0% total P to meet corn N requirements (151 kg PAN ha<sup>-1</sup>) adds 112 kg P ha<sup>-1</sup>, an annual surplus of 84 kg P ha<sup>-1</sup>. The environmental



**Table 4. Mean background concentrations (control) and magnitude of change in biosolids treated fields ( $\Delta$ ) for trace elements.**

Depth	1998		1999		2000		Three years	
	Control	$\Delta$	Control	$\Delta$	Control	$\Delta$	Control	$\Delta$
cm	mg kg <sup>-1</sup>							
0-10	7.00	-0.55	5.84	0.54	4.59	-0.06	5.69	0.03
10-20	6.62	-0.40	6.07	0.63	4.55	0.13	5.66	0.17
20-40	6.01	-0.87	6.61	0.75	4.41	0.43	5.64	0.20
	<b>Cr</b>							
0-10	25.87	4.86†	23.94	2.26†	26.15	2.48†	25.26	3.03†
10-20	30.54	-0.17	23.77	2.28†	25.34	5.62†	26.14	2.86
20-40	28.72	3.47†	29.90	1.38†	27.76	1.60†	28.80	2.02†
	<b>Cu</b>							
0-10	25.12	9.15†	23.58	10.53†	23.38	9.76†	23.91	9.88†
10-20	26.18	6.34†	23.64	6.55†	23.56	4.44†	24.28	5.72†
20-40	25.89	4.12†	23.79	4.24†	23.00	4.40†	24.05	4.27†
	<b>Hg</b>							
0-10	0.05	0.04†	0.06	0.04†	0.06	0.04†	0.06	0.04†
10-20	0.05	0.02†	0.05	0.01†	0.06	0.02†	0.05	0.02†
20-40	0.05	0.00	0.05	0.00	0.04	0.01†	0.05	NA‡
	<b>Pb</b>							
0-10	23.87	3.47†	23.42	3.35†	26.16	2.39	24.54	3.03†
10-20	24.56	0.68	21.82	3.96†	25.92	2.21	24.05	2.45
20-40	19.15	3.91†	17.19	3.93†	20.01	3.76†	18.74	3.86†
	<b>Mn</b>							
0-10	1018	335†	1050	268†	999	250†	1023	279†
10-20	1080	229†	1045	228†	1035	185†	1051	212†
20-40	802	236†	876	77	853	196†	848	163
	<b>Mo</b>							
0-10	3.48	0.14	3.96	0.57†	2.77	0.40	3.40	0.39†
10-20	3.61	0.21†	3.85	0.57†	2.83	0.25†	3.41	0.36†
20-40	3.30	0.22†	4.10	0.40†	2.92	0.27†	3.45	0.30†
	<b>Ni</b>							
0-10	20.68	2.91†	19.36	2.43†	21.00	0.18	20.31	NA
10-20	25.38	0.25	10.79	0.86	19.94	2.03	18.02	1.13
20-40	25.52	2.96	24.60	0.65	21.77	1.46	23.80	1.56
	<b>Zn</b>							
0-10	90.54	20.17†	90.58	15.77†	89.14	15.29†	90.03	16.76†
10-20	97.11	3.84	82.55	9.69†	87.10	4.38	88.08	6.19
20-40	84.56	5.48	83.92	12.72	77.93	19.56†	81.88	13.31

† Significant at the  $\alpha = 0.10$  level.‡ Three-year change is not available due to significant treatment  $\times$  time interaction. All three years were analyzed separately.

significance of these increases in observed P in this study cannot be assessed from the data we collected.

Combined analysis of all years indicated that, on average, biosolids-treated fields had lower concentrations of soil K than control fields. Soon et al. (1978) found that sludge application had a negligible effect on extractable soil K. When applied at N-based rates, the amount of K supplied by biosolids is often much lower than the amount needed by crops. This is due to the fact that K is water-soluble and is therefore removed from the sludge during dewatering (Pierzynski, 1994).

### pH, Acidity, and Calcium

Soil pH was evaluated for each year individually at the 0- to 10-cm depth due to the presence of significant treatment  $\times$  year effects and we found that biosolids-treated fields displayed higher pH than control fields only in 2000 (Table 3). There was no effect on pH below the 0- to 10-cm depth. By contrast, biosolids increased Ca at all soil depths. Increases in Ca while pH remained the same can be attributed to the use of lime-stabilized biosolids in some fields and to application of dolomitic limestone in many of the fields. Regulations in Pennsyl-

vania require that the pH be maintained above 6.0 when biosolids are applied to decrease the bioavailability of trace elements.

### Trace Elements

Combined analysis over all three years showed that the soil concentrations of Cr, Cu, Hg, Mn, Mo, and Zn were significantly higher in treated fields than in control fields at one or more depth increments (Table 4). Cadmium and Se concentrations were below instrument detection limits for all samples. There were no significant differences at any depth for As. Nickel and Mn showed evidence of treatment  $\times$  year interactions at the 0- to 10-cm depth and Hg data indicated the presence of a treatment  $\times$  year interaction at the 10- to 20-cm depth. These parameters were evaluated for each year individually at these depths using the paired *t* test (Table 4). Statistical analysis of biosolids affects on Cr and Ni did not include data from the Lancaster 2 site. These data were removed because the control field at the Lancaster 2 site had extremely high Cr and Ni concentrations in the soil (approximately 700 and 140 mg kg<sup>-1</sup>, respectively), compared with the treated field (ap-

**Table 5. Typical background levels for trace elements in noncontaminated soils and median concentration in Pennsylvania biosolids. Median values are based on more than 1000 analyses of biosolids produced in Pennsylvania in 1996–1997. Adapted from Stehouwer (1999a).**

Trace element	Typical background concentration range for surface soil	Median trace element concentrations in median Pennsylvania biosolids
	mg kg <sup>-1</sup> soil	mg kg <sup>-1</sup> biosolids
Arsenic	6–10	3.4
Cadmium	0.2–0.5	2.2
Chromium	15–36	35.1
Copper	17–65	505.0
Lead	8–22	62.0
Mercury	0.06–0.15	1.5
Nickel	7–18	22.0
Selenium	0.4–0.4	4.4
Zinc	19–82	694.0

proximately 70 and 30 mg kg<sup>-1</sup>, respectively). This can be explained by the presence of serpentine parent material that naturally contains trace elements at concentrations far higher than normal background levels. Chromium and Ni concentrations increased with soil depth in the profile providing further evidence that the high Cr and Ni concentrations resulted from parent material rather than from an anthropogenic source.

Since these regulated trace elements are normally present in biosolids at concentrations greater than normal soil background concentrations, such increases can be expected if the trace elements are conserved in the zone of biosolids incorporation. Table 5 compares typical surface soil background concentrations with median trace element concentrations in Pennsylvania biosolids. There was a very large difference between background concentrations of Cu, Zn, and Hg when compared with concentrations of these elements in biosolids. Therefore, increases in soil concentrations of these elements were expected following the addition of biosolids. Biosolids Cr is at the upper range typical for surface soils, and greater than the mean Cr concentration of control soils in this study (Table 4). Many studies have reported that trace elements accumulate in soils to which biosolids have been applied (Sposito et al., 1982; Chang et al., 1984; Canet et al., 1998). Conservation of biosolids trace elements is attributed to strong sorption on or coprecipitation with oxides of Mn and Fe in the biosolids, and strong sorption to soil minerals and organic matter that fixes the trace elements in relatively unavailable forms for long periods of time (Sommers, 1977; Emmerich et al., 1982; McGrath, 1987; Dowdy et al., 1994; McGrath and Cegera, 1992; Chaney and Ryan, 1993). Other researchers, however, have argued that trace elements may be released when added organic matter is mineralized, potentially allowing for movement through the soil profile and increased bioavailability (McBride, 1995). Bioavailability of trace elements was assessed by measuring crop tissue concentrations and by measuring trace element concentrations in soil below the usual depth of biosolids mixing.

#### Differences at the Twenty- to Forty-Centimeter Depth

The zone of incorporation for biosolids is generally within the top 10 to 20 cm of soil. Differences in trace element concentrations would not be expected at depths

greater than 20 cm because trace elements in biosolids tend to accumulate where they are placed. Our data indicate that the concentrations of Cu, Mo, and Pb at the 20- to 40-cm depth were higher for biosolids-treated fields than for control fields (Table 4). These results agree with studies by Sposito et al. (1982), Chang et al. (1984), and Campbell and Beckett (1988), who reported increases in trace elements below the zone of biosolids incorporation. Differences at depths below the zone of incorporation could be due to the deep incorporation of biosolids or to association of trace elements with soluble inorganic or organic ligands, or with mobile colloidal particles moving through the soil profile (McBride, 1995). It was beyond the scope of our study to determine the mechanisms that may have accounted for the observed increases in Cu, Mo, and Pb in the 20- to 40-cm depth.

#### Comparison with USEPA Part 503 Cumulative Pollutant Loading Rates

While a statistically significant trace element increase indicates that it is reasonable to conclude the observed difference was due to biosolids application, it provides no indication of any corresponding change in risk to plant, animal, or human health. Risk assessments used to develop the Part 503 regulations established the cumulative pollutant loading rates (CPLRs) as the maximum amount of an element that could be added to the soil without causing adverse effects on human, plant, or animal health (USEPA, 1993). One means of approaching the question of the potential risk to human and environmental health posed by the increases in trace element concentrations we found is to compare those differences with permissible increases under current regulations (Table 6). The largest difference occurred with Pb at 0.06 of the CPLR, while differences for other elements were <0.024. The small magnitude of these differences relative to USEPA-established safe levels indicates that the increased concentrations of some elements in these biosolids-treated fields have not increased the risk to human or environmental health.

#### Crop Tissue Composition

Biosolids application did not affect crop yield or tissue concentrations of N, P, K, Ca, Cu, Mo, Mn, Ni, and Zn

**Table 6. Estimated average trace element increases per hectare 40-cm depth expressed on a mass basis and as a fraction of cumulative pollutant loading rates (CPLRs).**

Trace element†	CPLR	Soil profile increase	
		Total	Fraction of CPLR
	kg ha <sup>-1</sup>		
Cu	1500	32.1	0.021
Pb	300	15.7	0.059
Hg	17	0.11	0.006
Ni	420	1.80	0.004
Zn	2800	65.9	0.024

† Estimated mass increases were calculated only for those trace elements whose soil concentrations were significantly larger in biosolids treated fields in the combined analysis of all three sampling years.

(data not shown). Other trace elements were below detection limits in the crop tissue samples (Cd and Pb, <0.04 and 0.25 mg kg<sup>-1</sup>, respectively) and therefore biosolids effects could not be assessed. These results indicate that while biosolids increased the soil concentration of some trace elements, their phytoavailability remained low since there was no effect on the concentration of the elements in crop tissue. Our results agree with those of Dowdy et al. (1994), who found no increase of trace elements in corn after 19 yr of sludge application, and McGrath (1987), who concluded that trace elements added to soil in biosolids are not efficiently removed by crops.

## CONCLUSIONS

Commercial application of biosolids to agronomic soils has led to the increase of NO<sub>3</sub>, P, and several trace elements in soil. Increased post-growing season NO<sub>3</sub> in biosolids-amended soils increases the risk of losses to surface and ground water. Further study is warranted to determine the cause of higher nitrate N concentrations in biosolids-treated fields so that nutrient management practices that decrease the risk of NO<sub>3</sub>-N leaching losses can be implemented. Increased surface soil P levels could be an environmental concern, particularly in watersheds with excessive P in surface waters. Again, further study is needed to determine the environmental availability of P in biosolids-amended soils before any conclusions can be reached regarding the environmental risk from P in biosolids. Decreases in soil K levels with ongoing biosolids use indicate that soil K levels should be monitored when biosolids are used and other sources of K applied as needed to maintain adequate K fertility.

Accumulations of trace elements in biosolids-amended soil were very small relative to CPLRs, indicating that these increases have not increased risk to human, animal, or plant health. Furthermore, the data provided no evidence of unexpectedly large concentrations of trace elements in biosolids-treated soils indicating that biosolids analysis and CPLR calculations are adequately controlling trace element additions to soil when biosolids are applied. However, increases in soil trace elements below 20 cm indicate that not all biosolids trace elements are being conserved within the depth of biosolids incorporation. The observed increases in soil trace elements and nutrients did not affect corresponding concentra-

tions in the harvested portion of the crop. Thus, there was no indication that current agronomic biosolids land application practices have affected either crop quality or increased the amounts of trace elements entering the food chain through crop uptake.

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