Published January 8, 2015

Journal of Environmental Quality

TECHNICAL REPORTS

WASTE MANAGEMENT

Uptake Coefficients for Biosolids-Amended Dryland Winter Wheat

Ken A. Barbarick,* James A. Ippolito, and Jacob McDaniel

Abstract

The USEPA adapted a risk assessment approach in biosolids regulations that includes the use of an uptake coefficient (UC) (i.e., the ratio of plant concentration to quantity of element added) to determine limitations on selected elemental additions. The nature of the risk assessment requires UCs to be constants. Our hypothesis was that the UC for Cu, Fe, Mo, Ni, P, and Zn for biosolids-amended dryland winter wheat (*Triticum aestivum* L.) decreases with multiple biosolids applications at the same location. We applied up to 10 applications to two sites (designated North Bennett A and B) in eastern Colorado at rates from 2.24 to 11.2 Mg ha^{-1} per application from 1993 to 2013. Results indicated that grain concentrations for all six elements followed no discernible trend as the number of biosolids applications increased. The UC values for these elements compared with the number of biosolids applications followed exponential decay models (R^2 ranged from 0.329 to 0.879). Consequently, UC values will likely not provide constants for risk assessment where multiple biosolids applications are made on the same site. We found that the slope between cumulative elemental removal by grain (kg ha^{-1}) to the cumulative amount of element added with biosolids (kg ha^{-1}) provides a constant over the number of biosolids additions (R^2 ranged from 0.471 to 0.990). As compared with the USEPA approach, our strategy of looking at cumulative changes may provide better estimations of wheat-grain concentrations for risk assessment of biosolids-borne elements.

A foundation for the USEPA biosolids land-application regulations (40 CFR Part 503) is risk assessment of plant trace metal uptake (USEPA, 1993). The risk assessment depends on the use of plant uptake coefficients (UCs):

$$\mathrm{UC} = \frac{\left(\text{mg plant element kg}^{-1}\right)}{\left(\text{kg added element ha}^{-1}\right)} \quad [1]$$

where UC is the plant concentration divided by the quantity of added trace metal. The USEPA chose geometric means of UCs from studies before 1992, with the inherent assumption that UCs were constants. Recent studies, however, have provided more complete information on UCs for various crops.

Sukkariyah et al. (2005) determined Cd, Cu, Ni, and Zn UCs for romaine lettuce (*Lactuca sativa* L.) and radish (*Raphanus sativus* L.) 17 to 19 yr after biosolids applications (up to 210 Mg ha^{-1}). The UCs for these two crops varied from 6.25 times greater for Ni in radish globes to 20 times lower for Cu in lettuce compared with the USEPA UC values. Granato et al. (2004) land-applied ~540 Mg biosolids ha^{-1} from 1974 to 1984 and then monitored corn (*Zea mays* L.) for 13 yr after biosolids applications ceased (1985–1997). These authors concluded that USEPA's UC risk assessment models overpredicted corn grain uptake of Cd, Cu, Ni, and Zn. Chen et al. (2009) found that As, Cd, and Pb plant uptake factors determined in 70 California vegetable fields were probabilistic and fit log-normal distributions; they did not fit the USEPA model. Extensive evaluation of field and greenhouse studies led O'Connor et al. (2001) to estimate that the Mo UCs for nonleguminous plants were <0.5 (mg plant Mo kg^{-1} per kg Mo applied ha^{-1}). Mullen et al. (2005) found the UC for Mo in unwashed winter wheat (*Triticum aestivum* L.) forage was 0.36, compared with the USEPA value of 0.42.

The aforementioned studies focused on UC determinations for either single biosolids applications or at one sampling time after addition. An important issue not addressed was how the UC for a given crop may change where continuous biosolids application at agronomic rates is practiced. Barbarick et al. (1995) found evidence that P, Cu, Mo, Ni, and Zn follow an exponential rise to a maximum model for winter wheat grain

J. Environ. Qual. 44:286–292 (2015)
doi:10.2134/jeq2014.08.0350
Received 18 Aug. 2014.
Accepted 10 Nov. 2014.
*Corresponding author (ken.barbarick@colostate.edu).

K.A. Barbarick and J. McDaniel, Dep. of Soil and Crop Sciences, Colorado State Univ., 200 W. Lake St., Fort Collins, CO 80523-1170; J.A. Ippolito, USDA-ARS-NWISRL, 3793 North 3600 East, Kimberly, ID 83341-5076. Assigned to Associate Editor Cesar Plaza.

Abbreviations: L/E, Littleton/Englewood Wastewater Treatment Plant; UC, uptake coefficient.

concentrations versus the accumulative amount of element added. Li et al. (2012) also observed an exponential rise to a maximum uptake response for wheat grain Zn and Cu after biosolids land application. These results suggest that UC values would subsequently decrease as the elements are continually added through biosolids application. Consequentially, the assumption that UCs are constants would be invalid.

Our hypothesis was that winter-wheat grain UC values for P, Cu, Fe, Mo, Ni, and Zn will decrease significantly as the number of biosolids applications increase. The findings of Barbarick et al. (1995) that Cu, Mo, Ni, and Zn wheat-grain concentrations versus cumulative element addition follow an exponential rise to a maximum regression model prompted this hypothesis. We determined the UC values for pollutants of interest (Cu, Mo, Ni, and Zn) and for two nutrients (P, Fe) that are over 1% concentration in the biosolids used in the current study. We used linear decline (Eq. [2]) and exponential decay (Eq. [3]) models to test this hypothesis.

$$UC = a - b(\text{number of biosolids applications}) \quad [2]$$

where a is the y axis intercept and b is the slope, and

$$UC = c\left(e^{-d(\text{number of biosolids applications})}\right) \quad [3]$$

where c is the amplitude of the curve and d is the rate constant.

Model choice is based on the equation that has the highest R^2 and lowest probability level and standard error of the estimate.

Materials and Methods

Plot Design

The biosolids research locations for this study were approximately 20 km north of Bennett, CO (referred to as North Bennett; 39.9563 N, 104.462 W) and were initiated in the summer of 1993. We have followed a wheat-fallow rotation on two fields (site A was initiated in 1993, and site B was initiated in 1994) consisting of Weld loam (fine, smectitic, mesic Aridic Argiustoll) (NRCS, 2014). The initial soil pH of the top 20 cm was 6.9 at both sites. The pH across treatments and both sites in 2012 and 2013 ranged from 6.9 to 7.8. The anaerobically digested biosolids from the Littleton/Englewood, CO Wastewater Treatment Plant (L/E; Table 1) were sand-bed dried for approximately 60 d before application. We manually applied biosolids at rates of 0, 2.24, 4.48, 6.72, 8.96, and 11.2 dry Mg biosolids ha^{-1} to 1.8 by 17.1 m plots from 1993 through 2013. The 4.48 dry Mg biosolids ha^{-1} application rate is the agronomic rate for both sites (Barbarick et al., 2000). Each year in late July or early August, we distributed the material evenly over the plots. We manually applied urea fertilizer (46–0–0) at rates of 0, 22.4, 44.8, 67.2, 89.6, and 112 kg N ha^{-1} to separate plots at the same time as biosolids application. Four replications of each treatment at each site were randomized in a complete block design (24 biosolids and 24 N fertilizer plots). Wheat grain was harvested and yields were determined in July each year, except for 2000–2001 at site B where the crop was lost to hail damage. Ground grain samples were digested in concentrated HNO_3 and analyzed for P, Cu, Fe, Mo, Ni, and Zn concentrations by inductively coupled plasma–atomic emission spectrophotometry (Soltanpour et al., 1996). Barbarick and Ippolito (2007, 2008) provide further research-site details.

Statistical Analyses

SAS Proc Mixed (SAS, 2014) was used to calculate the ANOVA for each grain concentration and UC. We used SAS Proc Reg (SAS, 2014) to find the slope of plant concentration versus

Table 1. The solids, Cu, Fe, Mo, Ni, P, and Zn content of the Littleton/Englewood, Colorado biosolids applied to dryland winter wheat at the two North Bennett sites in eastern Colorado, from 1993–2013.

Year applied	Site receiving	Solids (wet basis)	Cu	Fe	Mo	Ni	P	Zn
		g kg^{-1}	mg kg^{-1}					
1993	A	880	558	15,200	27.1	85.0	27,100	942
1994	B	880	493	11,100	19.6	65.3	16,600	816
1995	A	600	458	12,000	13.4	42.3	17,400	816
1996	B	730	657	17,000	20.2	51.7	23,000	652
1997	A	530	459	15,100	8.0	34.7	35.900	422
1998	B	680	236	13,700	5.5	15.2	11,800	301
1999	A	640	256	19,200	8.0	15.2	10,200	198
2000	B	800	352	18,000	6.4	1.1	36,000	370
2001	A	740	398	26,300	13.1	8.0	19,900	428
2002	B	930	326	19,700	15.7	7.6	19,000	351
2003	A	550	594	21,000	15.2	10.9	24,000	418
2004	B	920	652	36,700	18.2	13.9	30,000	767
2005	A	930	234	9,500	3.6	4.7	14,000	206
2006	B	800	458	12,300	23.4	7.1	24,300	165
2007	A	930	656	13,600	5.9	10.6	17,000	270
2008	B	—†	—	—	—	—	—	—
2009	A	716	880	13,900	31.8	15.0	24,500	872
2010	B	842	546	17,600	5.5	8.2	18,900	356
2011	A	925	800	19,000	5.5	14.2	22,000	693
2012	B	925	800	17,600	9.0	12.0	22,000	693

† No biosolids were applied. Farmer grew sunflowers (*Helanthus annus* L.) to help control a jointed goat grass (*Aegilops cylindrical* Host) infestation.

quantity of element added. The linear model was determined without an intercept (Eq. [1]); therefore, the calculated slope represents the UC. Model equations, R^2 values, probability levels, and standard errors were found using SigmaPlot (2009) version 11.2.

Results and Discussion

Barbarick et al. (1995) found that six applications of up to 144 Mg L/E biosolids ha^{-1} in a wheat-fallow rotation resulted in winter wheat grain P, Cu, Mo, Ni, and Zn concentrations versus the cumulative amount of element added fitting an exponential rise to a maximum regression model. In the current study at a different set of locations, the maximum amount of L/E biosolids applied was 112 Mg ha^{-1} in 10 applications. We found no discernable pattern of grain concentration versus elemental additions (nonsignificant changes over time); all regression models produced probability levels >0.05 (Fig. 1). One major difference between the two studies was the amount of biosolids added. The current study was designed to study

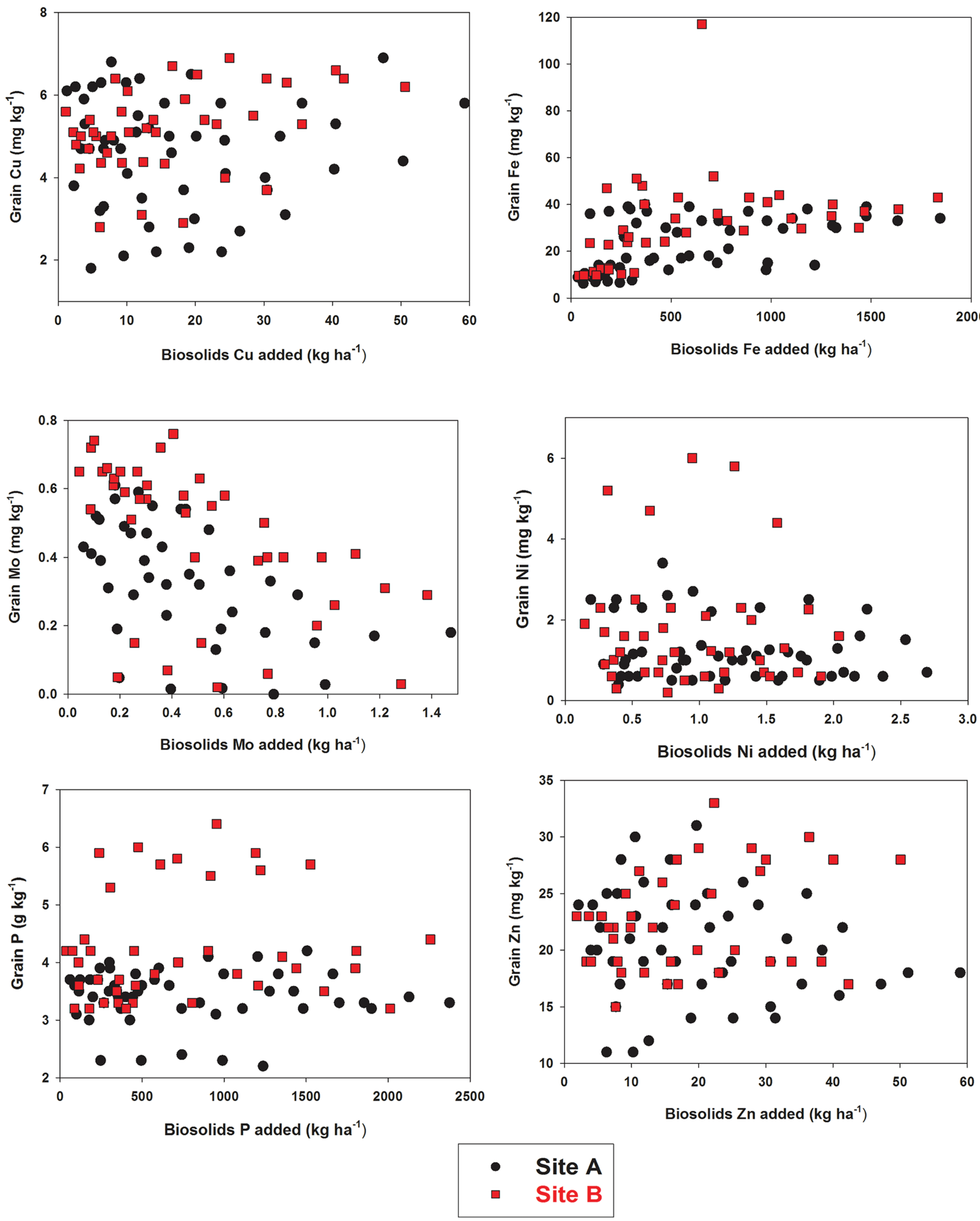

Fig. 1. Grain concentrations compared with biosolids additions of Cu, Fe, Mo, Ni, P, and Zn at the North Bennett research sites, 1993 to 2013.

plant and soil N and trace element changes within a range of agronomic rates. The Barbarick et al. (1995) study included rates to test the impact of up to 10 times the agronomic rate. The lower applications in our current study apparently were not conducive to developing an exponential rise to a maximum regression model.

The ANOVAs showed that the factors that affected the UCs for Cu, Fe, Mo, Ni, P, and Zn were the research site and the number of biosolids applications. Biosolids rate did not affect the UC values primarily because of the low rates of application (data not shown). Consequently, we developed regression models for Eq. [2] and [3] based on the number of biosolids applications for both research sites. For each element tested, the exponential decay model (Eq. [3]) generally had superior R^2 values, probability levels, and standard errors than the linear decline model (Eq. [2]) (Table 2). Figure 2 indicates that the probability level for all models was <0.01 except for Fe at both sites and Ni at site B. The R^2 values for the Cu, Mo, P, and Zn exponential decay models exceeded 0.77, whereas the R^2 values for Fe and Ni were <0.40. These results refute the inherent assumption in the USEPA risk analyses for the 503 biosolids regulations that the UC is constant. In fact, using the USEPA (1993) grain and cereals UC values for Ni (0.03) and Zn (0.027) and the regression models for each site separately and combined (Fig. 2), we calculated that about 19 and 18 biosolids applications would be required to reach the USEPA UC values for Ni and Zn, respectively. The USEPA risk assessment approach does not use UC for the other elements we studied.

If a constant that represents the effect of elemental additions on plant response is a consideration in risk assessment, then an alternative approach is to use the slope of the cumulative quantity of elemental plant removal to the cumulative amount of element added to estimate elemental grain concentration. For all elements tested as shown in Fig. 3, the removal versus amount added followed linear models for both sites and for the combined data except Mo, which followed an exponential rise to a maximum at sites A and B. All R^2 values were >0.76. The slopes for the linear models also indicate that 0.40 to 0.42, 0.05 to 0.06, 2.1 to 2.6, 6.6 to 13, and 1.6 to 1.7% of the biosolids-applied Cu, Fe, Ni, P, and Zn, respectively, were removed by the grain for any given number of biosolids applications at agronomic rates.

These results support the hypothesis that the UCs for dryland winter wheat grain Cu, Fe, Mo, Ni, P, and Zn decrease as the number of biosolids applications increases. The UC versus biosolids application followed exponential decay models for all elements at site A and the combined data and all but Fe and Ni at site B. Consequently, the variable nature of UC argues against its use in risk assessment of biosolids impact on winter wheat that receives multiple biosolids applications. We suggest that the ratio between the cumulative removal of an element to the cumulative amount of an element added in biosolids application should be used in determining wheat-grain concentrations for risk assessment of Cu, Fe, Ni, P, and Zn. This ratio was found to be constant over the number of biosolids application. The amount of Cu, Fe, Ni, and Zn removed after any given number of biosolids application ranged from about 0.05 to 2.6%, whereas P removal ranged from 6 to 13%.

Table 2. The R^2 values, standard errors, and probability levels for linear and exponential decay models for the winter wheat uptake coefficients for Cu, Fe, Mo, Ni, P, and Zn compared with the number of Littleton/Englewood, Colorado biosolids applications at the two North Bennett sites in eastern Colorado, from 1993 to 2013.

Element	Site	Model	R^2	SE	*P* value
Cu	A	linear	0.551	0.26	0.01
	A	exponential decay	0.900	0.12	<0.01
	B	linear	0.593	0.23	0.02
	B	exponential decay	0.842	0.14	<0.01
Fe	A	linear	0.249	0.024	0.08
	A	exponential decay	0.300	0.023	0.06
	B	linear	0.487	0.018	0.03
	B	exponential decay	0.461	0.019	0.04
Mo	A	linear	0.727	0.41	<0.01
	A	exponential decay	0.962	0.15	<0.01
	B	linear	0.780	0.58	<0.01
	B	exponential decay	0.953	0.27	<0.01
Ni	A	linear	0.407	0.79	0.03
	A	exponential decay	0.664	0.59	<0.01
	B	linear	0.131	1.4	0.20
	B	exponential decay	0.112	1.4	0.22
P	A	linear	0.589	3.0	<0.01
	A	exponential decay	0.894	1.5	<0.01
	B	linear	0.567	1.9	0.02
	B	exponential decay	0.847	3.7	<0.01
Zn	A	linear	0.643	0.57	<0.01
	A	exponential decay	0.887	0.32	<0.01
	B	linear	0.675	0.54	0.01
	B	exponential decay	0.822	0.40	<0.01

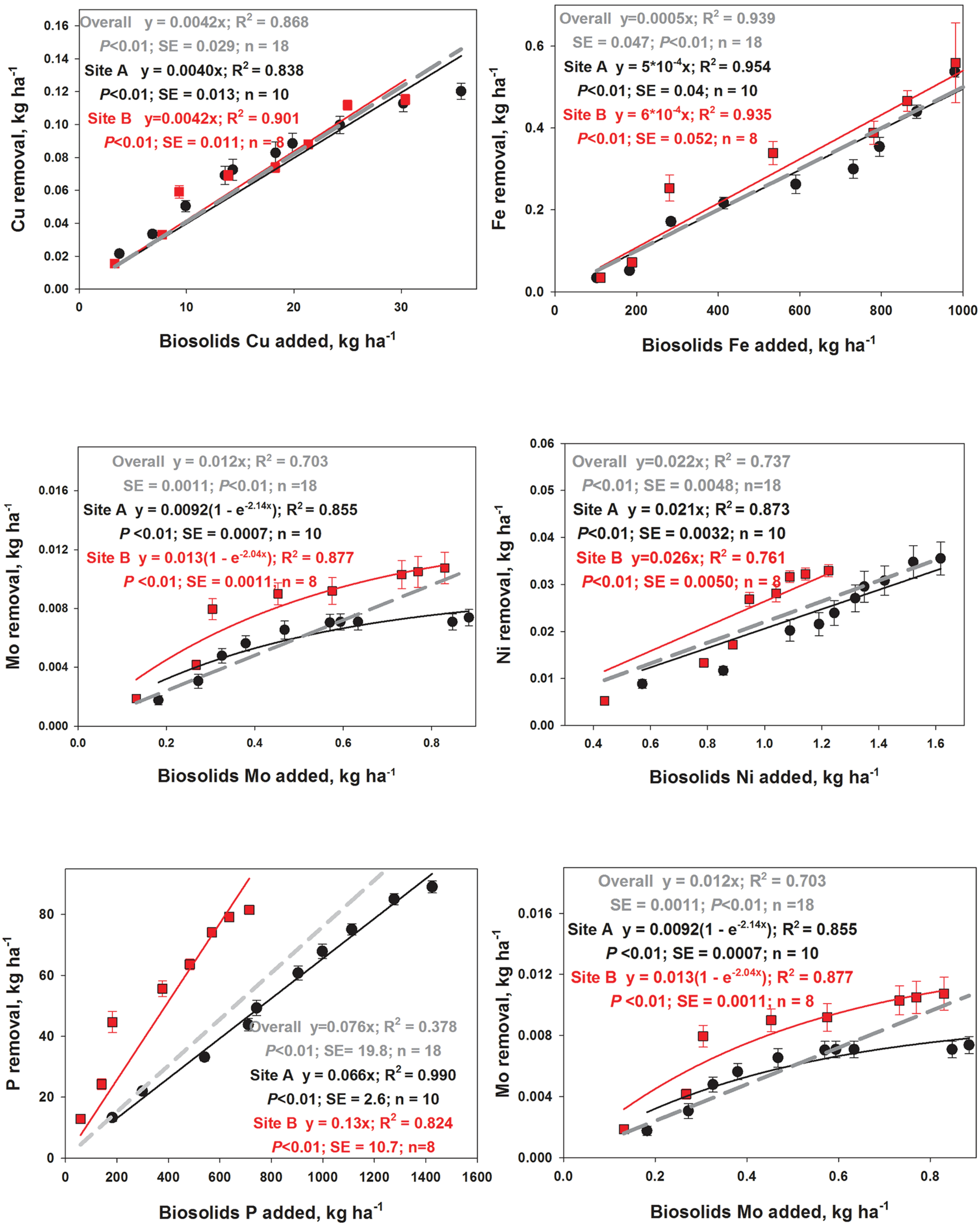

Fig. 2. Uptake coefficients for Cu, Fe, Mo, Ni, P, and Zn relative to number of biosolids applications at the North Bennett research sites, 1993 to 2013.

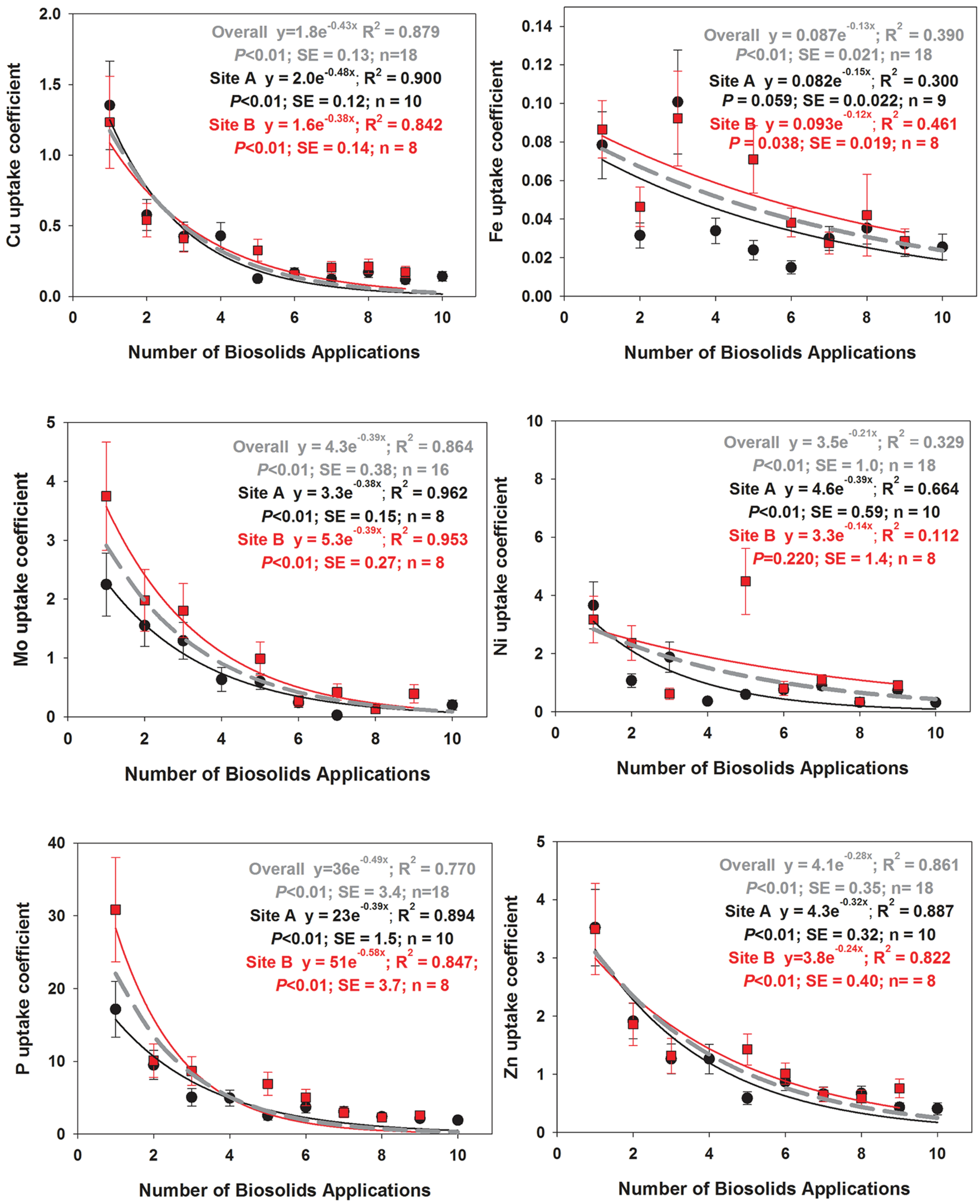

Fig. 3. Elemental grain removal relative to the quantity of biosolids-borne Cu, Fe, Mo, Ni, P, and Zn added at the North Bennett research sites, 1993 to 2013.

References

Barbarick, K.A., and J.A. Ippolito. 2000. Nitrogen fertilizer equivalency of sewage biosolids applied to dryland winter wheat. J. Environ. Qual. 29:1345–1351. doi:10.2134/jeq2000.00472425002900040043x

Barbarick, K.A., and J.A. Ippolito. 2007. Nutrient assessment of a dryland wheat agroecosystem after 12 yr of biosolids applications. Agron. J. 99:715–722. doi:10.2134/agronj2006.0221

Barbarick, K.A., J.A. Ippolito, and D.G. Westfall. 1995. Biosolids effect on P, Cu, Zn, Ni, Mo concentrations in dryland wheat. J. Environ. Qual. 24:608–611. doi:10.2134/jeq1995.00472425002400040009x

Barbarick, K.A., and J.A. Ippolito. 2008. Predicting soil-extractable zinc, phosphorus, iron, and copper in a biosolids-amended dryland-wheat agroecosystem. Soil Sci. 173:175–185. doi:10.1097/SS.0b013e3181652201

Chen, W., L. Li, A.C. Chang, L. Wu, R.L. Chaney, R. Smith, and H. Ajwa. 2009. Characterizing the soil-solution partitioning coefficient and plant uptake factor of As, Cd, and Pb in California croplands. Agric. Ecosyst. Environ. 129:212–220. doi:10.1016/j.agee.2008.09.001

Granato, T.C., R.I. Pietz, G.L. Knafl, C.R. Carlson, Jr., P. Tata, and C. Lue-Hing. 2004. Trace element concentrations in soil, corn leaves, and grain after cessation of biosolids application. J. Environ. Qual. 33:2078–2089. doi:10.2134/jeq2004.2078

Li, Q., X.Y. Guo, X.H. Xu, Y.B. Zuo, D.P. Wei, and Y.B. Ma. 2012. Phytoavailability of copper, zinc and cadmium in sewage sludge-amended calcareous soils. Pedosphere 22:254–262. doi:10.1016/S1002-0160(12)60012-5

Mullen, R.W., W.R. Raun, N.T. Basta, J.L. Schroder, and K.W. Freeman. 2005. Effect of long-term application of biosolids on molybdenum content and quality of winter wheat forage. J. Plant Nutr. 28:405–420. doi:10.1081/PLN-200049151

NRCS. 2014. Web soil survey. http://websoilsurvey.nrcs.usda.gov/app/WebSoilSurvey.aspx (accessed 1 Aug. 2014).

O'Connor, G.A., R.B. Brobst, R.L. Chaney, R.L. Kincaid, L.R. McDowell, G.M. Pierzynski, A. Rubin, and G.G. Van Riper. 2001. A modified risk assessment to establish molybdenum standards for land application of biosolids. J. Environ. Qual. 30:1490–1507. doi:10.2134/jeq2001.3051490x

SAS. 2014. SAS/STAT user's guide. Version 9.2. SAS Inst., Cary, NC.

SigmaPlot. 2009. SigmaPlot 11.2 user's manual. Systat Software, San Jose, CA.

Soltanpour, P.N., G.W. Johnson, S.M. Workman, J.B. Jones, Jr., and R.O. Miller. 1996. Inductively coupled plasma emission spectrometry and inductively coupled plasma-mass spectrometry. In: D.L. Sparks, editor, Methods of soil analysis. Part 3. Chemical methods. SSSA, Madison, WI. p. 91–139.

Sukkariyah, B.F., G. Evanylo, L. Zelanzy, and R.L. Chaney. 2005. Cadmium, copper, nickel, and zinc availability in a biosolids-amended piedmont soil years after application. J. Environ. Qual. 34:2255–2262. doi:10.2134/jeq2004.0369

USEPA. 1993. Standards for the use or disposal of sewage sludge. Fed. Regist. 58:9248–9415.