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Cadmium Concentrations in New Zealand Pastures: Relationships to Soil and Climate Variables

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Cadmium (Cd) is a nonessential element that occurs at above-background concentrations in many New Zealand (NZ) soils. Most of this Cd is due to the historical application of single superphosphate that was made from Nauru phosphate rock containing between 400 and 600 mg Cd kg^{-1} P. Pasture Cd uptake exacerbates the entry of Cd into animal products. We sought to determine the critical environmental factors affecting Cd uptake in NZ pastures and to calculate the likely Cd intake of sheep and cattle. We tested 69 pastures throughout NZ for a range of variables, including Cd. Soil Cd and pasture Cd were positively correlated with soil P and soil concentrations of other elements found in phosphate fertilizers. We found that no single environmental variable adequately predicted pasture Cd uptake. Nevertheless, pseudo-total soil Cd and Cd extracted using a 0.05 mol L^{-1} $Ca(NO_3)_2$ solution were positively correlated with pasture Cd. Although soil pH, soil Fe, and soil Cd provided an excellent predictor of the $Ca(NO_3)_2$–extractable soil Cd fraction, regression models explained just 38% of the variation of the Cd concentration in pasture grasses. Incorporating the effect of pasture species composition is a crucial next step in improving these models. A calculation of the likely exposure to Cd of sheep and cattle revealed that no pastures tested resulted in sheep and cattle ingesting Cd at a rate that would result in breaching muscle-tissue food standards. For offal products, which the NZ meat industry does not sell for human consumption, food safety standards exceedence was calculated in a few cases.

Cadmium (Cd) is a nonessential element (Kabata-Pendias and Mukherjee, 2007) that is readily taken up by plants and transferred through fodder and food products into animals and humans (Cadmium Working Group, 2011a). Enrichment of Cd in soils has been reported worldwide through the land application of biosolids, industrial effluents, and phosphate fertilizers with high Cd concentrations (Hooda, 2010; Kabata-Pendias and Pendias, 2001; Kabata-Pendias and Mukherjee, 2007). Atmospheric deposition of Cd is often as significant as input via soil amendments in some highly populated and industrialized countries, and fertilizer Cd dominates in other countries with lower populations and less industrialization, such as New Zealand (NZ) (Hooda, 2010; Taylor, 2007). Several authors have reported enrichment of Cd in NZ's pasture soils (Gray et al., 1999; Kim, 2005; Roberts et al., 1994; Schipper et al., 2011; Taylor, 1997; Taylor, 2007). Most Cd in these soils originates from mineral phosphate fertilizers (primarily single superphosphate), which may contain up to 280 mg Cd kg^{-1} P (Loganathan et al., 2003) (Cadmium Working Group, 2011b). In a NZ soil survey, Taylor et al. (2007) reported an average Cd concentration in pasture soils of 0.43 mg kg^{-1} ($n = 825$). This is more than double the background concentration of 0.16 mg kg^{-1} ($n = 372$) (Taylor, 2007).

Pastureland is the main type of farming in NZ, covering 13.8 million hectares (Loganathan et al., 2003). Grazing animals take up Cd mainly via consumption of herbage; intake through soil ingestion is generally lower (Lee et al., 1996; Loganathan et al., 2003). Food safety standards have been set to limit the concentration of Cd in foodstuffs. Exceeding such standards might result in trade restrictions and serious economic damage for producers (Cadmium Working Group, 2011a). The most important factor affecting food-chain transfer is soil–plant uptake (Hooda, 2010). The bioavailability of Cd in soil depends on its (total) concentration, pH, organic matter content, clay content, soil moisture conditions, and availability of macro- and micronutrients (Grant et al., 1999; Welch and Norvell, 1999). The use of a single soil-to-plant transfer coefficient for a given

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Abbreviations: ADI_{Cd}, acceptable daily intake for cadmium; BAF, bioaccumulation factor; DI_{Cd}, daily intake of cadmium; dw, dry weight; NZ, New Zealand.

plant species is not appropriate to describe plant Cd uptake in a wide range of soils (Rodrigues et al., 2012; Römkens et al., 2009).

Hooda (2010) suggested that Zn and Cd should be considered together to understand either of them in detail. The two elements are often cocontaminants (the typical Zn:Cd ratio is 200:1), have many similar properties in soil and plants, and are readily absorbed and translocated to plant shoots (Hooda, 2010). Synergistic and antagonistic effects of Zn on Cd uptake by plants are known, with antagonistic effects usually observed under non–Zn deficiency conditions (Grant et al., 1999). Whereas Zn is often deficient in agricultural systems, Cd is often a food-chain toxicity risk (Hooda, 2010).

Mechanistic models capable of predicting soil–plant transfer have not been developed (Swartjes, 2011). Various empirical models have been proposed to describe Cd accumulation by plants as a function of soil properties (e.g., pH, total Cd, Cd that is extractable using a dilute salt solution and soil organic matter content) (Smolders, 2001; Tudoreanu and Phillips, 2004). Empirical models are more limited in scope than mechanistic models; however, because they are based on observations, they provide a degree of accuracy not necessarily present in mechanistic models (Tudoreanu and Phillips, 2004). Empirical models are only useful in specific environments within the range of their calibration. They must be validated or recalibrated with local soil and environmental conditions (Swartjes, 2011). Little information about the proposed model variables is known other than under experiment-specific conditions. There is a lack of studies investigating the large number of environmental and plant factors suspected to influence Cd uptake under field conditions.

We aimed to elucidate the most important environmental and edaphic factors affecting Cd concentrations in NZ pastures with a view to identifying factors that could be measured along with soil Cd to predict the risk of plant Cd uptake. Furthermore, we sought to use the model of Rodrigues et al. (2012) to assess the daily intake of Cd by sheep and cattle on various pastures.

Materials and Methods

Soil and Pasture Sampling

Soil and pasture samples from 69 sites were collected during the NZ summer (Dec. 2009 to Feb. 2010). The sampling locations, given in Supplemental Table S1, were determined using a GPS receiver (accuracy, 8–10 m). Most of the pastures were intensive cattle or dairy farms, which typically receive high rates of P fertilization. Pastures were in the vegetative growth phase, and a visual inspection indicated that they contained minimal senescent material. The median total annual rainfall at the collection sites was 1838 mm (range, 424–5406 mm). The median average annual air temperature ranged from 8.6 to 16.4°C. The range of rainfall and temperature characteristics reflects the geographical spread and difference in elevation. Pasture samples were cut 2 to 3 cm above ground on an area of approximate 0.2 m^2. Nongraminaceous weeds were removed from samples to minimize the effect of species composition in the study. The composition of graminaceous species was typical for dairy farm pastures in NZ, dominated by perennial ryegrass [*Lolium perenne* (L.)], with some pastures containing small amounts of brown top [*Agrostis capillaris* (L.)]. Roberts and Longhurst (2002) reported that a typical NZ pasture contains approximately 70% ryegrass, 20% white clover, and 10% weeds. A composite sample of the underlying topsoil was obtained by means of a soil corer (10–15 cores per sampling location taken from a depth of 0–10 cm).

Chemical Analyses

Plant samples were washed in deionized water and dried at 60°C until a constant weight was obtained. Samples were milled using a Cyclotech type 1093 cyclone grinder with an aluminum rotor. Plant material (0.5 g) was digested in 5 mL HNO_3. The digests were diluted with Milli Q (EASYpure RF, 18.3 MΩ-cm, Barnstead) to a volume of 25 mL and filtered with Whatman 52 filter paper (pore size, 7 μm).

Soil samples were dried for 7 d at room temperature. The aggregates were separated and passed through a 2-mm nylon sieve. Soil pH was measured in Milli Q at a solid:water ratio of 1:2.5 using a Metler Toledo pH meter. The grain size distribution of the soils was determined using the pipette method according to the reference methods of the Swiss federal agricultural research stations (FAL et al., 1996).

Pseudo-total element concentrations in soil (henceforth referred to as "total") were measured in acid digests. Half a gram of soil was digested in 5 mL aqua regia (BDH Aristar nitric acid 69%, BDH Aristar hydrochloric acid 37%). Extractable elemental concentrations in soil were measured by weighing 5 g of soil into 40-mL centrifuge tubes and adding 30 mL 0.05 mol L^{-1} $Ca(NO_3)_2$ (BDH AnalaR [$Ca(NO_3)_2 \times 4H_2O$]). Tubes were agitated for 2 h on an end-over-end shaker, centrifuged for 10 min at 15,000 rpm, and filtered using Whatman 52 filter paper. Oxalate-extractable concentrations were measured according to the method described elsewhere (Blakemore et al., 1987). All extracts were stored at 4°C before the analysis.

Element concentrations were measured using ICP–OES (Varian 720 ES). Wageningen (ISE 921, IPE 100) and NIST (1573a) reference materials were analyzed for quality assurance. The results of these analyses are given in Table 1. The C and N content of soil and plant material was determined using an Elementar Vario MAX CN element analyzer.

Correction for Cadmium Arising from Surface Contamination

Because sample washing cannot remove all soil particles from field-collected plant material (Robinson et al., 2009), the plant concentrations were corrected for soil contamination using a method described by Robinson et al. (2008). Iron was used as the indicator element with the minimal measured value in plant of 44.4 mg kg^{-1} dry weight (dw) as baseline value. The masses of soil calculated using Fe as an indicator were in reasonable agreement with the corresponding masses when Al or Cr was used as indicator element ($R = 0.68$; $R = 0.76$ [Pearson]).

Geographic Information System Analysis

Climate data (raster dataset with 500 m spatial resolution) representing medians of the period 1971 to 2000 were obtained from the National Institute of Water and Atmospheric Research

Table 1. Results of the chemical analysis of the reference samples.

Reference material	Element	Certified value		Measured value	
		Mean	Confidence interval	Mean	SD
		mg kg^{-1}			
		Plant material			
NIST 1573	Cd	1.52	0.04	1.32	0.02
	Zn	30.9	0.7	28.9	0.31
IPE 100	Cd	0.085	0.003	0.076	0.011
	Zn	33	0.099	34.6	1.3
		Soil material			
ISE 921	Cd	2.32	0.1	2.58	0.3
	Zn	502	14	518	66

(Tait and Zheng, 2007). The value of the raster cell overlying the sampling point was ascribed to that sample using the ESRI ArcGIS 9.3 software. The average annual solar radiation is henceforth referred to as radiation, the total annual rainfall as rainfall, and the average annual air temperature as temperature.

Statistical Analysis

Correlations between variables were described by Spearman rank correlation coefficients. To identify the best linear regression models, an exhaustive search (i.e., an all-subset regression) across all predictor variables was conducted. The best model for each number of explanatory variables was selected given that all estimated coefficients were significant ($\alpha = 0.05$, F-test using type III sums of square). Variables showing positively skewed frequency distributions [organic C, total Mn, total S, oxalate-extractable Zn (ox-Zn), oxalate-extractable Cd (ox-Cd), $Ca(NO_3)_2$–Zn, $Ca(NO_3)_2$–Cd, rainfall, pasture N, pasture Zn, and pasture Cd] were log-transformed before regression analysis. The relative importance metric LMG (Lindemann, Merenda, and Gold) was calculated to describe the importance of explanatory variables (Lindeman et al., 1980). Statistical analyses were undertaken with the software R (R-Core-Team, 2012) and Excel 2007. For correlation analysis, the R package *Hmisc* (Harrell, 2012) was used. For model selection and assessment of the importance of model variables, the R packages *leaps* (Lumley, 2009) and *relaimpo* (Grömping, 2006) were used.

Results and Discussion

Total Cadmium and Zinc Concentrations in Soil

Table 2 shows that the Cd concentration in topsoil ranged from 0.25 to 1.58 mg kg^{-1} (mean, 0.89 mg kg^{-1}). Earlier NZ surveys reported mean Cd concentrations of 0.4 mg kg^{-1} for pasture soils (Roberts et al., 1994; Taylor, 2007). The discrepancy in our results may be because most of our samples were taken from farmland that was under intensive dairy farming, which receives higher fertilizer inputs than the extensive grazing systems included in the aforementioned studies. Comparing them with the mean background concentration of 0.16 mg kg^{-1} (Taylor, 2007), our values are consistent with the frequently reported accumulation of Cd in NZ soils due to anthropogenic inputs.

The clay contents ranged from 3 to 40% (mean, 23%). These values are typical values for relatively young soils (Scheffer et al., 2010), as are the soils found in the NZ landscape. Total soil Cd was positively correlated with clay content, total Fe, Mn, Al, P, Ca, and Zn (Table 3). Total Cd was negatively correlated with sand content and site temperature. The positive correlations with clay content and total concentrations of Fe, Mn, and Al are consistent with the binding of Cd by metal oxides and clay minerals (Scheffer et al., 2010).

That Cd was positively correlated with P and Ca is consistent with the numerous reports that these elements coaccumulate due to fertilizer use (Loganathan et al., 2003). Despite the presence of 11% S in single superphosphate, the total Cd concentration did not correlate with the total concentration of S in soil (Table 3). This is not surprising because S as sulfate leaches from NZ soils, only being retained in significant quantities as organic S (Edmeades et al., 2005; Ghani et al., 1991).

There was a positive correlation between Zn and P and Ca in addition to that between Cd and Zn, indicating that fertilizer was an important source of Zn. Like Cd, Zn occurs as impurity in phosphate rocks used for the production of superphosphate fertilizers (Syers et al., 1986). McGrath and Tunney (2010) observed a significant increase in soil Zn (on average, 8.19 mg kg^{-1} in the 0- to 20-cm soil layer) in a grazed pasture after 31 yr of superphosphate addition (930 kg kg^{-1} over 31 yr) compared with a control pasture receiving no superphosphate (McGrath and Tunney, 2010). For many years, phosphate rock from Nauru Islands, known to contain high concentrations of Cd (100 mg kg^{-1}) and Zn (1100 mg kg^{-1}), was used for the production of superphosphate applied to New Zealand and Australian soils (Cadmium Working Group, 2008). In a survey of Iranian arable soils used for wheat production, Karami et al. (2009) found a correlation between total Zn and available P (Olsen P). They concluded that superphosphate fertilizer with high Zn (100–1100 mg kg^{-1}), which is commonly applied in Iran, is an important source of Zn in these soils. Zinc might also accumulate in NZ pasturelands because it is used as a prophylactic against facial eczema in sheep and cattle (Anderson et al., 2012). Therefore, another explanation for the observed correlations may be that pastures that received higher rates of phosphate fertilizer also received higher rates of zinc as a result of the prophylactic facial eczema treatment.

$Ca(NO_3)_2$–Extractable Cadmium and Zinc Concentrations

The $Ca(NO_3)_2$–extractable fractions of soil Cd and Zn have been reported to be good predictors of plant Cd and Zn uptake in pasture soils (Black et al., 2011; Black et al., 2012). Table 2 shows the $Ca(NO_3)_2$–extractable fractions of Cd and Zn in the soils tested. The extractable concentrations were approximately three orders of magnitude lower than the total concentrations, consistent with observations that these elements are immobile in weakly acidic to alkaline soils (Loganathan and Hedley, 1997). Mean K_D values (total/extractable concentration coefficients) for Cd and Zn were approximately 3000 and 6000, respectively.

All sampled soils had pH values below 7.0 (mean, 5.5) (Table 2). Calcium nitrate–cadmium was negatively correlated with pH (Table 3). Variable charged binding sites are increasingly protonated at low pH and unavailable for binding Cd. In addition, there is increased competition with acidic metal cations (e.g., Al^{3+}) for binding sites under acidic conditions (Loganathan et al., 2012; Scheffer et al., 2010). Most of the sampled soils probably had a history of liming because acidity limits fertility, particularly N availability, on many NZ farms (Carran, 1992). There was a negative correlation between $Ca(NO_3)_2$–Cd and total Ca. This is likely due to an indirect effect related to the pH increase as a result of liming. The positive correlation of total Ca with pH (Supplemental Table S2) is consistent with this rationale. The positive correlation between $Ca(NO_3)_2$–Cd and rainfall is also suspected to be related to soil pH because higher rainfall was negatively correlated with pH (Supplemental Table S2). Basic cations tend to leach with the percolating rainwater, and the moist soil conditions allow increased biological activity, which produces acidity (Sparks, 2003). The negative correlation between $Ca(NO_3)_2$–Cd and the clay fraction and total Al is consistent with the aforementioned likely importance of Al-bearing minerals of the clay fraction in binding of Cd. The correlation with $Ca(NO_3)_2$–Zn reflects the geochemically similar behavior of Cd and Zn (Hooda, 2010). The $Ca(NO_3)_2$–Cd concentration was governed by factors influencing the solubility rather than by the total soil Cd, as there was no significant correlation with total Cd.

Tables 4 and 5 show regression models that use edaphic and climate variables to calculate $Ca(NO_3)_2$–extractable concentrations of soil Cd and Zn. In each model, between one and six environmental variables are used, with successive variables increasing the explanatory power (R^2_{adj}) but decreasing the ease of parameterization of the model. For Cd and Zn, pH was the most important soil parameter in explaining the $Ca(NO_3)_2$–extractable concentrations. Incorporating organic C, total Fe, total Cd, ox-Cd, and rainfall, linear regression explained 86% of the variation in $Ca(NO_3)_2$–Cd. Total Fe is incorporated with a negative sign, consistent with Fe oxides being important adsorbents of Cd. Because organic carbon is known to bind Cd, the incorporation of organic C with a negative sign is unsurprising. The organic carbon contents where medium to high (range, 2.0–15.9 wt%; median, 5.5 wt%). This is characteristic for NZ soils, which typically have higher organic matter contents than soils from arid countries (Cadmium Working Group, 2008). Compared with total Fe, the relative importance of organic C as explanatory variable was minor. Linear regression models to predict $Ca(NO_3)_2$–Zn differed slightly from Cd, with total P, total Mn, and temperature in the set of explanatory variables (Table 5).

Table 2. Descriptive statistics of variables determined in this study.

Parameter	Mean	Median	Range	SD	Skew
pH	5.5	5.6	4.8–6.9	5.21–6.18†	1.01
Clay, %	22.5	21.0	3.0–40.0	9.0	0.07
Sand, %	36.2	35.8	2.6–76.4	16.4	0.21
Organic C, wt%	6.3	5.5	2.0–15.9	2.8	1.09
Total Fe, g kg^{-1}	24.5	25.9	1.8–44.7	10.9	−0.43
Total Mn, g kg^{-1}	0.44	0.39	0.04–2.35	0.32	3.10
Total Al, g kg^{-1}	27.0	26.9	3.9–52.0	11.1	−0.13
Total N, g kg^{-1}	5.3	5.0	2.0–10.5	2.1	0.70
Total P, g kg^{-1}	1.15	1.06	0.36–2.64	0.52	0.81
Total S, g kg^{-1}	0.64	0.58	0.18–1.77	0.29	1.25
Total Ca, g kg^{-1}	6.92	6.85	1.08–18.78	3.99	1.00
Total Zn, mg kg^{-1}	65.0	67.5	11.5–107.8	26.0	−0.28
Total Cd, mg kg^{-1}	0.89	0.88	0.25–1.58	0.31	0.24
Ox-Zn, mg kg^{-1}	13.1	10.7	4.0–57.2	7.83	2.88
Ox-Cd, mg kg^{-1}	7.9×10^{-2}	6.9×10^{-2}	2.8×10^{-2} to 2.1×10^{-1}	3.8×10^{-2}	1.12
$Ca(NO_3)_2$–Zn, mg kg^{-1}	3.0×10^{-2}	2.0×10^{-2}	8.7×10^{-4} to 1.6×10^{-1}	3.1×10^{-2}	2.26
$Ca(NO_3)_2$–Cd, mg kg^{-1}	7.3×10^{-4}	6.8×10^{-4}	4.9×10^{-5} to 3.1×10^{-3}	5.5×10^{-4}	1.22
Radiation, MJ m^{-2} d^{-1}	13.8	14.0	12.1–15.0	0.8	−0.43
Temperature, °C	12.1	12.2	8.6–16.4	1.7	0.24
Rainfall, mm	1995	1838	424–5406	1128	1.46
Pasture N, g kg^{-1}	19.7	17.2	6.3–46.0	8.9	1.18
Pasture P, g kg^{-1}	2.80	2.73	0.77– 6.01	1.15	0.46
Pasture S, g kg^{-1}	2.34	2.08	0.43–4.68	1.03	0.48
Pasture Ca, g kg^{-1}	5.48	5.67	1.75–10.06	1.86	0.20
Pasture Zn, mg kg^{-1}	28.1	26.8	9.0–89.4	10.6	2.90
Pasture Cd, mg kg^{-1}	0.13	0.08	0.01–1.55	0.21	5.44

† Standard deviation range.

Pasture Cadmium and Zinc Concentrations

Most (90%) pasture samples had Cd concentrations <0.2 mg kg^{-1}. The median and mean were 0.08 and 0.13 mg kg^{-1}, respectively (Table 2). Seven samples had concentrations >0.2 mg kg^{-1}, with the highest sample having a Cd concentration of 1.55 mg kg^{-1}. A previous NZ survey reported mean Cd concentrations in pasture grass of 0.1 mg kg^{-1}. However, the pasture soil surveyed by Roberts et al. (1994) and most probably the species composition in this work are not comparable to our study. Although there can be significant differences in Cd uptake among various plant species and cultivars (Crews and Davies, 1985; McLaughlin et al., 1994), there is a lack of information on variations in Cd uptake between various pasture species. The Zn concentrations ranged from 9 to 89 mg kg^{-1}, with a mean of 28 mg kg^{-1}. In terms of soil-to-plant transfer coefficient (plant/soil concentration quotient), the median values for Cd and Zn were 0.09 and 0.42, respectively. Similarly the median plant/$Ca(NO_3)_2$–extractable quotients for Cd and Zn were 157 and 1352, respectively. This indicates that pasture takes up Zn, an essential micronutrient, more effectively than Cd, a nonessential heavy metal. This is probably due to the presence of Zn-specific transporters that are adapted to take up this essential element (Reid and Hayes, 2003).

The concentration of Cd in pasture was positively correlated with the total soil concentrations of P and Cd, the ox-Cd, the $Ca(NO_3)_2$–Cd, rainfall, and pasture concentrations of P, S, Ca, and Zn (Table 3). The correlation with $Ca(NO_3)_2$–extractable Cd (R = 0.41) was stronger than with ox-Cd (R = 0.36) and total Cd (R = 0.31). Thus, of the three Cd fractions, $Ca(NO_3)_2$–extractable Cd explained the bioavailable Cd pool best, in line with the findings of Black et al. (2012).

Total Cd and $Ca(NO_3)_2$–extractable Cd in soil correlated significantly with pasture Cd, whereas total Cd and $Ca(NO_3)_2$–extractable Cd did not correlate with each other. This may imply that the readily soluble fraction of Cd in soil was not the only source for pasture Cd (i.e., plants had access to stronger bound Cd, possibly through Cd mobilized by root exudates). Analyses of pasture throughout the growing season may reveal a different relationship between total Cd and plant Cd due to physiological changes during the plants' life cycles.

The uptake and root-to-shoot translocation of micronutrients by plants can be affected by interactions among these elements (Nan et al., 2002a) and vary with the plant's nutrient status (Robinson et al., 2009). Synergistic and antagonistic effects have been reported between Zn and Cd uptake (Kabata-Pendias and Pendias, 2001). Although our results do not reveal any mechanisms of Cd–Zn interactions, they are not consistent with a strong antagonistic interaction between Cd and Zn. The pasture concentrations of the two metals were positively correlated (R = 0.39) but were not correlated with the soil concentrations (total, $Ca(NO_3)_2$–extractable) of each other. Compared with Zn, which is often reported to behave similar to Cd in the soil–plant system (Hooda, 2010), Cd was less dependent on the plant nutrient status but more strongly related to the total soil Cd

Table 3. Spearman coefficient of correlation.

Parameter	Total Zn	Total Cd	Oxalate-extractable		$Ca(NO_3)_2$–extractable		Pasture Zn	Pasture Cd
			Zn	Cd	Zn	Cd		
pH	0.23	0.20	0.11	−0.04	−0.62a†	−0.62a	0.03	0.07
Clay	0.50a	0.35b	0.21	0.07	−0.11	−0.29c	−0.20	−0.12
Sand	−0.45a	−0.28c	−0.12	0.03	0.02	0.05	0.35b	0.01
Organic C	−0.21	0.05	0.21	0.58a	−0.06	0.04	0.05	0.01
Total Fe	0.67a	0.86a	0.09	0.20	−0.20	−0.23	−0.02	0.04
Total Mn	0.54a	0.67a	−0.02	0.06	−0.14	−0.18	−0.02	0.09
Total Al	0.6a	0.66a	0.10	0.25c	−0.09	−0.28c	−0.06	−0.01
Total N	−0.01	0.13	0.30c	0.60a	−0.08	0.04	0.14	0.01
Total P	0.37b	0.51a	0.36b	0.63a	−0.17	0.06	0.22	0.26c
Total S	−0.10	0.13	0.30c	0.55a	−0.03	0.09	0.23	0.21
Total Ca	0.45a	0.36b	0.04	−0.12	−0.37b	−0.40a	0.16	−0.06
Total Zn		0.63a	0.34b	0.05	−0.11	−0.21	0.06	0.00
Total Cd	0.63a		0.13	0.48a	−0.25c	−0.04	0.10	0.31b
Ox-Zn	0.34b	0.13		0.14	0.23	−0.07	0.10	−0.06
Ox-Cd	0.05	0.48a	0.14		−0.13	0.33b	0.06	0.36b
$Ca(NO_3)_2$–Zn	−0.25c	−0.25c	0.23	−0.13		0.51a	0.11	0.00
$Ca(NO_3)_2$–Cd	−0.04	−0.04	−0.07	0.33b	0.51a		0.10	0.41a
Radiation	0.14	0.03	−0.19	−0.32b	0.02	−0.19	−0.02	−0.13
Temperature	−0.02	−0.24c	0.14	−0.16	0.22	0.01	0.11	0.01
Rainfall	−0.09	0.10	−0.02	0.26c	0.03	0.46a	0.16	0.25c
Pasture N	0.24c	0.15	0.19	0.07	−0.28c	−0.16	0.63a	0.16
Pasture P	0.21	0.20	0.17	0.14	−0.25c	−0.03	0.50a	0.29c
Pasture S	0.25c	0.16	0.08	0.02	−0.24c	−0.08	0.55a	0.24c
Pasture Ca	0.09	0.25c	0.10	0.27c	−0.52a	−0.10	0.30c	0.34b
Pasture Zn	0.06	0.10	0.10	0.06	0.11	0.10		0.39b

† Significance code: a < 0.001 < b < 0.01 < c < 0.05.

Table 4. Multiple linear regression models for $Ca(NO_3)_2$–extractable Cd.

No. of variables	Regression model† log[$Ca(NO_3)_2$–Cd] =	Relative importance of explanatory variables‡	Model validation		
			R^2§	R^2_{adj}¶	*P*
1	0.9670 – 0.75881 (pH)		0.52	0.51	$<10^{-10}$
2	1.5466 – 0.7183 (pH) + 0.7057 log(ox-Cd)#	78%, 22%	0.63	0.62	$<10^{-13}$
3	0.9920 – 0.7916 (pH) – 0.0351 (total Fe) + 1.1450 (total Cd)	71%, 18%, 11%	0.73	0.71	$<10^{-15}$
4	3.0451 – 0.6966 (pH) – 0.8185 log(org. C) – 0.0134 (total Fe) + 1.2986 log(ox-Cd)	60%, 5%, 11%, 24%	0.78	0.77	$<10^{-15}$
5	−0.579406 – 0.7369 (pH) – 0.6607 log(org. C) – 0.0440 (total Fe) + 1.4215 (total Cd) + 0.5340 log(rainfall)	51%, 3%, 17%, 10%, 19%	0.84	0.82	$<10^{-15}$
6	0.4121 – 0.6758 (pH) – 0.8890 log(org. C) – 0.0326 (total Fe) + 0.8289 (total Cd) + 0.7241 log(ox-Cd) + 0.5072 (rainfall)	45%, 5%, 12%, 6%, 16%, 16%	0.87	0.86	$<10^{-15}$

† All coefficient significant at $\alpha = 0.001$ (F-test using type III sums of square).

‡ LMG/R^2, in the order of explanatory variables in the model.

§ Model with the complete set of explanatory soil and climate variables (# 19, Table 3) determined $R^2 = 0.90$.

¶ Maximum adjusted R^2 for model size of 12 explanatory variables ($R^2_{adj} = 0.87$).

ox-Cd, oxalate-extractable Cd.

Table 5. Multiple linear regression models for $Ca(NO_3)_2$–extractable Zn. Best model with a size of five explanatory variables not listed because significance of all explanatory variables (F-test using type III sums of square) was not given.

No. of variables	Regression model† log[$Ca(NO_3)_2$–Zn] =	Relative importance of explanatory variables‡	Model validation		
			R^2§	R^2_{adj}¶	*P*
1	2.4748 – 0.7480 $(pH)_a$		0.51	0.50	$<10^{-10}$
2	1.9550 – 0.7747 $(pH)_a$ + 0.6348 log$(ox\text{-}Zn)_a$#	86%, 14%	0.61	0.60	$<10^{-12}$
3	0.3208 – 0.8373 $(pH)_a$ + 0.7135 log$(ox\text{-}Zn)_a$ + 0.1383$(radiation)_b$	82%, 14%, 4%	0.67	0.66	$<10^{-14}$
4	0.4261 – 0.8187 $(pH)_a$ – 0.1610 $(total\ P)_c$ + 0.8335 log$(ox\text{-}Zn)_a$ + 0.1272 $(radiation)_b$	76%, 5,% 16%, 4%	0.71	0.69	$<10^{-14}$
6	1.7950 – 0.8023 $(pH)_a$ – 0.0086 $(total\ Fe)_c$ + 0.4216 log$(total\ Mn)_b$ – 0.1477 $(total\ P)_c$ + 0.7642 log$(ox\text{-}Zn)_a$ + 0.0623 $(temp)_b$	74%, 4%, 13%, 2%, 6%	0.74	0.71	$<10^{-14}$

† Significance code: a < 0.001 < b < 0.01 < c < 0.05 (F-test using type III sums of square).

‡ LMG/R^2, in the order of explanatory variables in the model.

§ Model with the complete set of explanatory soil and climate variables (# 19, Table 3) determined $R^2 = 0.80$.

¶ Maximum adjusted R^2 for model size of 12 explanatory variables ($R^2_{adj} = 0.74$).

ox-Zn, oxalate-extractable Zn.

concentration and soil factors affecting Cd solubility. Pasture Cd concentration was only weakly correlated with pasture P ($R = 0.29$) and S ($R = 0.24$), was not correlated with pasture N, but was strongly correlated with total Cd, $Ca(NO_3)_2$–Cd and ox-Cd in soil. In contrast, pasture Zn concentration strongly correlated with pasture N ($R = 0.63$), P ($R = 0.50$), and S ($R = 0.55$) but did not correlate with total Zn, $Ca(NO_3)_2$–extractable Zn, or ox-Zn. Similar observations were made by other researchers. Nan et al. (2002b) used stepwise regression analysis to produce a meaningful model relating wheat grain Cd concentration with the explanatory variables total Cd in soil, pH, and available P. Starting with the same set of potential explanatory variables (pH, OM, available P, and total soil metal concentrations), they found no meaningful multiple regression model for grain Zn concentration of wheat. They suggested that, in contrast to Cd, under field conditions the uptake of Zn was largely affected by the crop's physiological characteristics.

Tables 6 and 7 show the best multiple linear regression models we derived to predict pasture concentrations of Cd and Zn, respectively. Generally, edaphic and climate variables only poorly accounted for pasture Cd concentration, with a six-factor regression model having an R^2_{adj} value of just 0.38. The $Ca(NO_3)_2$–extractable, ox-Cd, and total Cd concentrations were heavily weighted in the regression models (Table 6).

The multiple regression models for Zn had more explanatory power, with just four terms providing a fit with an R^2_{adj} of 0.51. Besides $Ca(NO_3)_2$–Zn, these models were heavily weighted in pasture N and include pasture Cd (with a positive sign), strengthening the above-mentioned dominant role of plant factors in Zn uptake.

Daily Intake of Cadmium for Animals Grazing on Sampled Pasture

Assuming that animals graze in the field all the time and always with the same Cd concentration in the forage, the daily intake of Cd (DI_{Cd}) (mg d^{-1}), can be estimated according to Eq. [1] (Rodrigues et al., 2012). Using daily consumption parameters I_{feed} (kg d^{-1} dw) and I_{soil} (kg d^{-1} dw) from the literature (Rodrigues et al., 2012) and the measured soil and pasture Cd concentrations, $[Cd]_{soil}$ and $[Cd]_{feed}$ (mg kg^{-1}), we calculated a DI_{Cd} for all sites for cow and sheep. We used food standards of Australasia (ANZFSC, 2012) and the European Union (Rodrigues et al., 2012), respectively, and bioaccumulation factors (BAFs) from the literature (Rodrigues et al., 2012) to calculate an acceptable daily intake for Cd (ADI_{Cd}) using Eq. [2] (Rodrigues et al., 2012). The BAF is defined as the Cd concentration in the organ (mg kg^{-1} fresh weight) divided by the concentration in the feed (mg kg^{-1} dw).

$$DI_{Cd} = [Cd]_{feed} \times I_{feed} + [Cd]_{soil} \times I_{soil} \qquad [1]$$

$$\mathrm{ADI_{Cd}} = \frac{[\mathrm{Cd}]_{\text{limit in animal organ}} \times (\mathrm{I_{feed}} + \mathrm{I_{soil}})}{\mathrm{BAF}_{\text{feed - animal organ}}} \qquad [2]$$

Table 8 shows that all calculated $\mathrm{DI_{Cd}}$ values were lower than $\mathrm{ADI_{Cd}}$ when calculated considering muscle tissue food standards. Considering offal food standards (kidney, liver), some calculated $\mathrm{DI_{Cd}}$ values exceeded the $\mathrm{ADI_{Cd}}$. The few exceptions are consistent with the reported exceedence of food safety standards for offal products of gracing animals in NZ, which led to the current policy of the NZ meat industry not to use offal of animals over 2.5 yr of age for human consumption (Roberts et al., 1994). Although continued application of Cd-containing phosphate fertilizers will increase Cd concentrations in animal products, this effect may be lessened by the Tiered Fertilizer Management System (Rys, 2011). Using this system, soils with <0.6 mg kg^{-1} Cd (19% of our samples) have no restriction placed on the application of P fertilizers. Soils with concentrations >0.6 and <1.0 (51% of our samples) are permitted restricted use of P to ensure that the soil does not exceed 1.8 mg kg^{-1} Cd after 100 yr. More intensive monitoring and mod-

Table 6. Multiple linear regression models for pasture Cd.

No. of variables	Regression models† log(pasture Cd) =	Relative importance of explanatory variables‡	Model validation		
			R^2§	R^2_{adj}¶	*P* value
1	$-0.3118 + 0.6562\ \log(\text{ox-Cd})_b$#		0.14	0.13	<0.01
2	$-1.7490 + 0.4841\ (\text{pH})_a + 0.6170\ \log[\text{Ca(NO}_3)_2\text{–Cd}]_a$	28%, 72%	0.26	0.24	<0.01
3	$-1.7465 + 0.4234\ (\text{pH})_b + 0.2931\ (\text{total Cd})_c + 0.5935\ \log[\text{Ca(NO}_3)_2\text{–Cd}]_a$	19%, 25%, 57%	0.33	0.29	$<10^{-4}$
4	$-2.4780 + 0.3808\ (\text{pH})_b + 0.2672\ (\text{total Cd})_c + 0.5308\ \log[\text{Ca(NO}_3)_2\text{–Cd}]_a + 0.5518\ \log(\text{pasture Zn})_c$	14%, 19%, 43%, 24%	0.37	0.33	$<10^{-4}$
5	$-0.7926 - 0.0224\ (\text{total Fe})_b - 0.0440\ (\text{total N})_c + 1.0233\ (\text{total Cd})_a + 0.2120\ \log[\text{Ca(NO}_3)_2\text{–Cd}]_c + 0.0532\ (\text{pasture Ca})_c$	14%, 5%, 31%, 26%, 24%	0.40	0.35	$<10^{-5}$
6	$1.8828 - 1.4610\ \log(\text{org. C})_a - 0.0303\ (\text{total Fe})_a + 1.0404\ \log(\text{total S})_c + 1.2863\ (\text{total Cd})_a - 0.4255\ \log(\text{ox-Zn})_c$†† $- 0.1125\ (\text{radiation})_c$	16%, 19%, 17%, 36%, 7%, 4%	0.44	0.38	$<10^{-5}$

† Significance code: a < 0.001 < b < 0.01 < c < 0.05 (F-test using type III sums of square).

‡ LMG/R^2, in the order of explanatory variables in the model.

§ Model with the complete set of explanatory variables (# 25, Table 3) determined $R^2 = 0.55$.

¶ Maximum adjusted R^2 for model size of 10 explanatory variables ($R^2_{adj} = 0.42$).

ox-Cd, oxalate-extractable Cd.

†† ox-Zn, oxalate-extractable Zn.

Table 7. Multiple linear regression models for pasture Zn.

No. of variables	Regression models† log(pasture Zn) =	Relative importance of explanatory variables‡	Model validation		
			R^2§	R^2_{adj}¶	*P* value
1	$0.8439 + 0.4632\ \log(\text{pasture N})_a$		0.37	0.36	$<10^{-7}$
2	$0.9325 + 0.0968\ \log(\text{Ca(NO}_3)_2\text{–Zn})_b + 0.5261\ \log(\text{pasture N})_a$	10%, 90%	0.44	0.43	$<10^{-7}$
3	$1.0693 + 0.0936\ \log(\text{Ca(NO}_3)_2\text{–Zn})_b + 0.4874\ \log(\text{pasture N})_a + 0.0882\ \log(\text{pasture Cd})_c$	9%, 73%, 18%	0.49	0.47	$<10^{-8}$
4	$1.0197 + 0.0019\ (\text{sand})_c + 0.0871\ \log(\text{Ca(NO}_3)_2\text{–Zn})_b + 0.4674\ \log(\text{pasture N})_a + 0.0937\ \log(\text{pasture Cd})_c$	12%, 7%, 64%, 17%	0.54	0.51	$<10^{-8}$

† Significance code: a < 0.001 < b < 0.01 < c < 0.05 (F-test using type III sums of square).

‡ LMG/R^2, in the order of explanatory variables in the model.

§ Model with the complete set of explanatory variables (# 25, Table 3) determined $R^2 = 0.67$.

¶ Maximum adjusted R^2 for model size of 12 explanatory variables ($R^2_{adj} = 0.57$).

Table 8. Daily intake and acceptable daily intake based on food standards of New Zealand (2.50 mg kg^{-1} kidney, 1.25 mg kg^{-1} liver, 0.050 mg kg^{-1} muscle) and the European Union (1.0 mg kg^{-1} kidney, 0.50 mg kg^{-1} liver, 0.050 mg kg^{-1} muscle).†

Animal	$\mathrm{DI_{Cd}}$‡ range (median)	Ingested as plant matter range (median)	Standards from§	$\mathrm{ADI_{Cd}}$¶			DI > $\mathrm{ADI_{Cd}}$		
				Kidney	Liver	Muscle	Kidney	Liver	Muscle
	mg	%			mg			% cases	
Cow	0.3–26.6 (1.8)	47–99 (80)	NZ	15	39	262	2.9	0	0
			EU	5.8	16	262	5.8	1.4	0
Sheep	0.1–4.0 (0.3)	35–98 (70)	NZ	3.1	1.8	45	1.4	2.9	0
			EU	1.3	0.7	45	2.9	7.2	0

† Calculation according to Eq. [1] and Eq. [2] with parameters $\mathrm{I_{feed}} = 16.9$ kg d^{-1} dw; $\mathrm{I_{soil}} = 0.41$ kg d^{-1} dw; $\mathrm{BAF_{feed\text{-}animal\ organ}} = 2.99$ (kidney), = 0.554 (liver), 0.0033 (muscle) for cow; and parameters $\mathrm{I_{feed}} = 2.5$ kg d^{-1} dw, $\mathrm{I_{soil}} = 0.10$ kg d^{-1} dw, and $\mathrm{BAF_{feed\text{-}animal\ organ}} = 2.08$ (kidney), = 1.85 (liver), = 0.0029 (muscle) for sheep (Rodrigues et al., 2012).

‡ Daily intake of Cd.

§ EU, European Union; NZ, New Zealand.

¶ Acceptable daily intake of Cd.

eling is required for soils with Cd concentrations >1 and <1.8 mg kg^{-1} (30% of our samples). No further Cd accumulation is permitted in soils with Cd concentrations >1.8 mg kg^{-1} (0% of our samples) (Rys, 2011).

Intake from pasture was more important than from soil ingestion (80 and 70% of total intake for cow and sheep, respectively). As discussed above, there are numerable variables that affect concentration of Cd in pasture. Therefore, it is unsurprising that there was no significant correlation between the DI_{Cd} and the total soil Cd concentration. The soils where DI_{Cd} exceeded ADI_{Cd} did not have significantly higher Cd than the remaining samples. However, this does not indicate that total soil Cd is unimportant. Other things being equal, increasing soil Cd will invariably increase pasture Cd uptake, as indicated by the positive correlation between the two (Table 3). Soil samples >0.88 mg kg^{-1} Cd (50% of the samples) had a significantly ($p < 0.001$) higher DI_{Cd} (1.32 and 1.50 mg for cow and sheep, respectively) compared with soils with <0.88 mg kg^{-1} Cd (0.74 and 0.82 mg for cow and sheep, respectively). Increasing soil Cd concentrations may result in a greater number of breaches of food safety standards.

Conclusions

Soil Cd and pasture Cd were positively correlated with soil P and soil concentrations of other elements found in phosphate fertilizers. Our study indicated that no single environmental variable could be used to adequately predict pasture Cd uptake. Nevertheless, total soil Cd and $Ca(NO_3)_2$–extractable soil Cd were positively correlated with pasture Cd. Although soil pH, soil Fe, and soil Cd provided an excellent predictor of the $Ca(NO_3)_2$–extractable soil Cd fraction, regression models explained just 38% of the variation of the Cd concentration in pasture grasses. Incorporating the effect of pasture species composition is a crucial next step in improving these models. Our calculation predicted no exceedences of food safety standards for muscle tissue but predicted some exceedences for offal products. Although continued application of phosphate fertilizers containing Cd will increase the concentration of Cd in animal products, the Tiered Fertilizer Management System may lessen this effect. Future work should involve determining the relative accumulation of various pasture species and examining the rhizosphere chemistry in a range of edaphic conditions. This will allow a better understanding of when the acceptable daily intake may be exceeded and permit the development of management tools, such as pasture species selection, that will reduce the exposure of stock to Cd.

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