

Environmental Parameters Affecting the Concentration of Iodine in New Zealand Pasture

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Abstract

Iodine (I) is an essential trace element commonly deficient in agricultural systems. Whereas there is much information on I in food crops, there is a lacuna of knowledge on the environmental factors that affect pasture I concentrations. We aimed to identify the most important environmental factors affecting the concentration of I in New Zealand pastures, and the consequences to agricultural systems. Soil and pastoral samples were collected throughout the country and analyzed for I and other elements. The soils contained 1.1 to 86 mg I kg⁻¹, with 0.005 to 1.4 mg kg⁻¹ in the pasture. In 26% of pastures, I concentrations were insufficient for sheep nutrition, whereas 87% contained insufficient I for cattle nutrition. Pasture I concentrations were negatively correlated with the distance from the sea, and the concentration of oxalate-extractable amorphous Al, Fe, and Si oxides, which immobilize soil I. Soil organic C and clay increased I retention in soil but did not significantly affect pasture I concentrations. Future work should investigate how soil properties affect pasture I uptake in inland areas.

Core Ideas

- 87% of New Zealand pastures contained insufficient iodine for cattle.
- Pasture I levels are higher close to the sea.
- Pasture I levels are lower in soil that is high in Fe and Al (oxy-hydr)oxides.

MORE than two billion humans are deficient in iodine (I), an essential trace element for animals (Mottiar, 2013), and many agricultural systems suffer deficiencies, thereby reducing productivity. Deficiencies cause mental and physical retardation, goiter, and potentially death (Zimmermann, 2009). Sheep and cattle require feed I concentrations of 0.05 to 0.2 and 0.3 to 0.5 mg kg⁻¹ respectively, for which pasture is the predominant source in many grazing systems (Suttle, 2005; Flachowsky, 2007; Grace and Knowles, 2012). The prevalence of I deficiency is due to low concentrations in inland soils, low soil bioavailability, and nonessentiality for terrestrial plants (Fuge, 1996; Zimmermann, 2009; Medrano-Macías et al., 2016). Iodine deficiency is exacerbated by goitrogens, which are thyroid I-limiting compounds commonly found in brassicas (*Brassica* spp.) and clover (*Trifolium* spp.) (Suttle, 2005). These are predominantly organic glucosinolates (Hurrell, 1997). Iodine deficiency is potentially exacerbated by selenium (Se) deficiencies, which are common worldwide, including in New Zealand (Arthur et al., 1999; Grace and Knowles, 2012). Consequently, iodized salt has been extensively supplied to humans, with iodized salt licks, oil, drenches, and boluses provided to animals (Miller, 1979; Lee et al., 2002; Grace and Knowles, 2012; Ershow et al., 2018).

The background concentrations of I in soil typically range from 0.1 to 100 mg kg⁻¹ (Fuge 1996; Fuge and Johnson 2015), with a mean concentration of 5.1 mg kg⁻¹ (Johnson, 2003). Concentrations in Northern Ireland, which is geographically similar to New Zealand, averaged 10.6 mg kg⁻¹ (Smyth and Johnson, 2011). Concentrations of 56 mg kg⁻¹ were reported in the United Kingdom peat soils, whereas soils in both Norway and the United Kingdom ranged from 4.2 to 14.7 mg I kg⁻¹ (Fuge, 1996). Relatively little soil I originates from the parent material (Whitehead, 2000). In contrast, seawater, which contains 0.06 mg I L⁻¹ (Wright, 1995), is a major reservoir of the element in the biosphere, with seaspray and subsequent wet and dry deposition of volatilized I being important processes for supplying the element to soils. It follows that elevated I concentrations are often found in soils close to the sea (Fuge and Johnson,

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Abbreviations: CRM, certified reference material; DFS, distance from sea; DI, deionized; TMAH, tetramethylammonium hydroxide.

2015; Medrano-Macías et al., 2016). Johnson (2003) reported geometric means of 11.6 and 2.6 mg kg⁻¹ <50 and >50 km from the sea, respectively. Iodine may be lost from soil via removal in crops, volatilization, and leaching (Medrano-Macías et al., 2016). Therefore, soil I concentrations are also a function of the capacity of the soil to immobilize I (Fuge and Johnson, 2015).

Speciation, pH, redox potential, organic carbon (C), and clay content influence the mobility of I in soil (Kaplan et al., 2000). The predominate soil forms of I are organic, followed by iodide (I⁻) and iodate (IO₃⁻), which are present at various proportions (Fuge and Johnson 2015; Hu et al., 2009). Labile electrostatic adsorption to positive functional groups is higher under acidic conditions, though as the pH increases, I⁻ oxidizes to IO₃⁻ thus it has more specific adsorption and less volatilization potential—albeit partially hindered by anion repulsion (Yoshida et al., 1992; Fuge, 1996; Zou et al., 2018). Soil sorption decreased as the pH increased from approximately 4.5 to 7 (Yoshida et al., 1992; Söderlund et al., 2017), and liming caused I losses in English soil (Bowley et al., 2017). However, there was no significant relationship between pH and total I in an extensive Northern Irish database (Smyth and Johnson, 2011), with reports of high-I soils formed from heavily weathered limestone and other high pH areas (Fuge, 1996). Solubility increases with waterlogging and decreasing redox potential when iron (Fe) and manganese (Mn) oxides dissociate and IO₃⁻ is reduced to I⁻ (Medrano-Macías et al., 2016; Qian et al., 2017). Iodine is firmly retained by clay-sized particles and particularly organic matter (Shetaya et al., 2012; Medrano-Macías et al., 2016; Humphrey et al., 2018), with Johnson (2003) finding that peat followed by clay soils contained the most I.

Iodine concentrations in pasture are sometimes insufficient to sustain grazing animal health. Much of the international pasture concentration data fall within the range of 0.1 to 1.0 mg I kg⁻¹, which encompasses the ruminant requirement range of 0.05 to 0.5 mg kg⁻¹, with as little as 0.026 and >3 mg I kg⁻¹ recorded (Smith et al., 1999; Grace and Waghorn, 2005; Trávníček et al., 2011). Plant roots take up I⁻ more rapidly than IO₃⁻, as I⁻ likely follows the same pathway as chloride (Cl⁻) (Whitehead, 2000; White and Broadley, 2009), with recent evidence that plants convert IO₃⁻ to I⁻ (Humphrey et al., 2019). Iodine enters the leaves via stomata and cuticular waxes, though it is often fixed by the waxes and hence retained in exposed leaves with reduced translocation throughout the plant (Humphrey et al., 2019; Medrano-Macías et al., 2016). Solubility and atmospheric concentrations affect plant uptake. Pakchoi (*Brassica chinensis* L.) I concentrations across three different soils were negatively correlated with their degree of I immobilization (Hong et al., 2012). In field conditions, *Hylocomium splendens* (Hedw.) Schimp. (moss) and willow (*Salix* spp.) shrub leaves contained significantly more I near the sea in Norway (Steinnes, 2008; Sivertsen et al., 2014). English plot trials demonstrated that pasture I was positively correlated with soil I and negatively correlated with soil pH (Bowley et al., 2017). In Northern Ireland, 20 predominantly pasture vegetation and soil samples from various locations found that pasture I negatively correlates with the distance from the sea, soil pH, and Fe or Mn oxides and is positively correlated with soil I and organic C (Bowley, 2013).

Widespread agricultural I deficiencies have been identified in NZ (Hercus et al., 1925; Grace and Knowles, 2012). Nevertheless,

some recent work has shown that soil I concentrations are not unusually low in some areas, with a mean of 20.9 mg kg⁻¹ found in the Waikato region (McNally, 2011). New Zealand's intensive grazing systems use pasture as the first source of I for ruminants. There has not yet been investigation into the effect of the aforementioned soil properties and proximity to the sea on the concentration of I in New Zealand pastures, how they may be affected by major and trace elements such as Mo, N, P, S, and Se in the soil-plant system, and how they affect the potential for ruminant I deficiencies. We hypothesized that pasture I concentrations decreased with increasing yield, were higher close to the sea, and were lower in organic C and clay-rich soils via reduced availability. We aimed at identifying the environmental factors that significantly influenced the concentration of I in New Zealand pasture and the consequences to agricultural systems.

Materials and Methods

Sampling and Processing

About 0.2 m² of pasture, predominantly perennial ryegrass (*Lolium perenne* L.), was sampled from intensively grazed pastures at 69 locations throughout New Zealand during December 2009 to January 2010. The sampling locations are provided in the Supplemental Table S1 (Reiser et al., 2014). Pasture was cut 2 to 3 cm from the surface to avoid contamination with soil. From the same paddock, 10 to 15 soil samples of 0- to 10-cm depth soil were collected with a soil corer then combined.

Locations were determined via GPS, and minimum distances to the sea (DFS) were estimated using Google Earth. The soil was dried at 105°C for 24 h then sieved to 2 mm and stored. Pasture was rinsed with deionized (DI) water, dried at 60°C, then milled and stored.

Chemical Analysis

The soil particle size composition was determined with the pipette method, and pH measured with a Mettler Toledo pH meter after addition of DI water (Milli-Q at 18.2 MΩ) (Reiser et al., 2014). Soil and vegetation C and N were measured with an Elementar Vario MAX CN element analyzer; in most New Zealand soils, all the measured C is organic if the pH is <7, which was true in all soils, hence the total C will be referred to as organic C.

The concentrations of many elements in the soils were measured: pseudo-total Al, As, B, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Se, and Zn via digestion of 0.5 g soil in 5 mL aqua regia (BDH Aristar nitric acid 69%, BDH Aristar hydrochloric acid 37%), oxalate extractions were completed for Al, As, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, P, Pb, S, Si, and Zn using the method adapted from (Blakemore et al., 1987), and soluble concentrations of Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, P, S, and Zn were measured via extraction and filtration with 0.05 M calcium nitrate (Reiser et al., 2014). Samples were stored at 4°C then measured by inductively coupled plasma optical emission spectroscopy (ICP-OES) with a Varian 720 ES. Certified reference materials (CRMs) Wageningen ISE 921, IPE 100, and NIST 1573a were used to confirm acceptable recovery by the digestion. Recoveries ranged from 89 to 108% of the CRMs.

Soil and pasture were analyzed for total I, following standard methods (Whitehead, 1984; Yamada et al., 1996; Fecher et al.,

1998; Tagami et al., 2010). Subsamples of each were dried at 103°C for 4 h (prepAsh 129 machine, Precisa Instruments). The soil was milled further, then 0.25 g was added to 4 mL of DI water (Milli-Q at 18.2 MΩ) and 1 mL of 25% tetramethylammonium hydroxide (TMAH) in a 25-mL volumetric flask, in 90°C water for 3 h. When cooled, flasks were filled to volume with DI water, transferred and centrifuged at 2620g for 20 min (HiCen21, Herolab), then filtered (0.45-μm syringe filter; Sartorius Stedim). The procedure was repeated for pasture, except 4.5 mL DI water and 0.5 mL 25% TMAH was added to flasks, with a centrifuge speed of 5897g. Samples were measured with a Varian inductively coupled plasma mass spectrometry (ICP-MS) type 820. Iodine calibrations at 0, 1, 5, 10, 25, and 50 μg L⁻¹ were completed. The machine detection limit was 0.032 μg L⁻¹, giving a sample detection limit of 0.0032 mg kg⁻¹. Certified reference materials included five soils (GSS 4, 6, 8, 10, and 14) from the Institute of Geophysical and Geochemical Exploration in Hebei, China, and hay powder (BCR-129) and skim milk powder (BRC-063R) from the Community Bureau of Reference in Brussels, Belgium. Recoveries ranged from 78 to 118% (Supplemental Table S2).

Data Analysis

Several authors have reported that soil and dust particles become incorporated into the leaves of field sampled plants, thereby changing the measured concentration (Robinson et al., 2008). Iron was used as an indicator for soil contamination of the sample. Although Fe is an essential plant micronutrient, plant uptake is strongly regulated, hence high values can indicate some contribution from soil. In our survey, pasture Fe concentrations ranged from 44 to 740 mg kg⁻¹. There was a strong correlation ($r = 0.790$, $p < 0.001$) with chromium (Cr), which is usually found at low concentrations in vegetation. Corrections for soil contamination were completed using Eq. [1] and [2], adapted from Robinson et al. (2008). First, the concentration of contaminating soil, $[s]$ (kg kg⁻¹), was estimated, using Eq. [1]:

$$[s] = ([Fe]^1 - [Fe]) / ([Fe]_s - [Fe]) \quad [1]$$

where $[Fe]^1$ is the concentration of Fe measured in pasture (mg Fe kg⁻¹ dry wt.), $[Fe]$ is the regulated concentration of 40 mg Fe kg⁻¹ dry wt. assumed in pasture, and $[Fe]_s$ is the concentration of Fe measured in soil (mg kg⁻¹). After this, the actual concentration of the element in vegetation, $[x]$, was calculated using Eq. [2]:

$$[x] = ([x]^1 - [x]_s[s]) / (1 - [s]) \quad [2]$$

where $[x]^1$ and $[x]_s$ are the concentrations of the elements in soil and vegetation (mg kg⁻¹), respectively.

Data were tabulated using Microsoft Excel (Office 365). Correlation analyses were performed after the data were tested for normality and log-transformed as appropriate.

Results and Discussion

Concentrations of Iodine in Soil and Pasture

Soil I concentrations ranged from 1.1 to 86 mg kg⁻¹, with a mean of 9.0 mg kg⁻¹ and median of 4.8 mg kg⁻¹. Supplemental Fig. S1 shows a map of the soil I concentrations. Our soil I data are higher than the international mean of 5.1 mg kg⁻¹

(Johnson, 2003), but within the range of 4.2 to 14.7 mg kg⁻¹ in single-country studies from Northern Ireland, Norway, and the United Kingdom (Fuge, 1996; Smyth and Johnson, 2011). Our mean soil concentration was significantly lower than the mean (20.9 mg kg⁻¹) reported for the Waikato region of New Zealand (McNally, 2011).

Pasture I concentrations ranged from 0.005 to 1.4 mg kg⁻¹, with a mean of 0.27 mg kg⁻¹ and median of 0.21 mg kg⁻¹. Our mean fell within the common pasture and grass range of 0.1 to 1.0 mg kg⁻¹, and values were of similar scale to prior studies in New Zealand and overseas (Whitehead, 1984; Trávníček et al., 2011; Sivertsen et al., 2014; Bowley et al., 2017). Some 74% of the pasture concentrations were sufficient for sheep, which require 0.05 to 0.20 mg I kg⁻¹ (Suttle, 2005), but just 13% contained sufficient I for cattle, which require 0.3 to 0.5 mg I kg⁻¹ (Flachowsky, 2007). Goitrogenic white clover (*Trifolium repens* L.) accompanies perennial ryegrass in pastures (Suttle, 2005), likely raising animal I requirements. Other feed is supplemented when pasture growth is low in winter. The predominant I deficiency scenario is sheep eating goitrogen-containing brassicas during winter (Grace and Knowles, 2012).

Single outliers with inordinately high I concentrations (soil > 86 mg kg⁻¹, pasture > 1.4 mg kg⁻¹) were present in both soil and pasture samples. These may have resulted from external I inputs such as salt licks, or animal supplements that have been deposited onto soil via urine and feces (Grace and Waghorn, 2005). Iodine fertilizers are not often used in New Zealand agriculture (Grace and Knowles, 2012), though superphosphate can contain I, depending on the source material (McNally, 2011). Seaweed fertilizers, which can contain up to 5000 mg I kg⁻¹, may be used (Yeh et al., 2014). A few herbicides and pesticides contain I (McNally, 2011).

There is a low risk of Se deficiency exacerbating I deficiency. Sheep and cattle require at least 0.03 mg kg⁻¹ in feed (Ullrey et al., 1977), and pasture had a mean of 0.090 mg Se kg⁻¹, with just six samples (11%) below the deficiency threshold. Of those, four contained less than the dairy cow requirement of 0.49 mg I kg⁻¹.

Factors Affecting Soil Iodine Concentrations

There was no significant correlation between soil I and the DFS (Table 1). This may be because soil I also depends on the capacity of the soil to retain the I that it has received (Fuge and Johnson, 2015). Our data showed significant correlations between soil I and clay ($p < 0.01$) and organic matter (total C) ($p < 0.005$) (Table 2), which is consistent with strong retention on the solid phase (Medrano-Macías et al., 2016). The maximum soil I concentration decreased as the distance from the sea increased (Fig. 1), and the mean annual rainfall was high, at 1838 mm (Reiser et al., 2014); thus, it is likely that weakly and nonbound material was leached beyond the 0- to 10-cm sampling depth, producing the range of concentrations. The four sites that did not fit this trend were N04 and the three locations between 46.5 and 62 km with the second to fourth highest concentrations of I. Supplement contamination is a possible cause; soil sodium (Na) concentrations (a constituent of salt licks) at three of the locations were within the highest 20%. There was a significant positive correlation between soil I and soil Na (Table 2). Our findings were similar to those of other authors (Johnson, 2003; Smyth

and Johnson, 2011; Bowley, 2013), who reported the highest soil I concentrations close to the sea, but with a large variation. McNally (2011) verified that leaching is likely, with significantly more I at the 10- to 20- than the 0- to 10-cm depth in soils from the Waikato region of New Zealand. Leaching is reduced in soils where I is retained by organic C and clay (Whitehead, 2000; Johnson, 2003; Smyth and Johnson, 2011; Bowley, 2013). Most of our samples were influenced by seawater as they were within 50 km of the sea. The sea has a negligible influence on soil I concentrations >50 km inland (Fuge, 1996; Johnson, 2003). Only 12 of our sampled locations (17% of the samples) were >50 km from the sea.

The amorphous Fe and aluminum (Al)-silicate (oxyhydr) oxide fraction sorbed and retained I in soil (Shetaya et al., 2012). Soil I was positively correlated with Al, Fe, and silicon (Si) oxalate ($r = 0.652-0.843$) (Table 2). There was no significant correlation between soil pH and soil I, which is consistent with the findings of other authors measuring I in field soils (Smyth and Johnson, 2011; Bowley, 2013). However laboratory-based studies have reported a positive correlation between pH and I sorption (Yoshida et al., 1992; Zou et al., 2018).

Factors Affecting Pasture Iodine Concentrations

Total soil I did not significantly affect pasture I, with an r value of -0.207 . Our findings stand in contrast with other studies (Bowley, 2013; Bowley et al., 2017) that have reported significant positive correlations between soil I and plant I. Our study had 69 locations throughout New Zealand, whereas Bowley et al. (2017) used just one location and Bowley (2013) used 20 locations. Therefore, it is likely that our study incorporated many more soil types and climatic zones (Tait and Zheng, 2007) (NIWA), which are critical determinants of plant I uptake (Hong et al., 2012).

Table 1. Concentrations of predominant elements in seawater, and correlations between the distance from sea and their soil or plant concentrations ($n = 44-69$).

Element	Concentration in seawater†	Correlation between soil concentration and distance‡			Correlation between pasture concentration and distance
		Total	Oxalate	Soluble	
	mg L ⁻¹				
Na	10,770	-0.080			-0.256**
Mg	1,290	0.129			-0.240*
S	905	-0.063	0.219	-0.367**	-0.245*
Ca	412	0.231			0.200
K	380	0.140			0.022
C	28	0.072			0.357**
N	11.5	0.076			0.046
B	4.4	0.010			-0.257*
Li	0.18	-0.017			0.056
P	0.06	0.157	0.221	-0.056	-0.051
I	0.06	0.123			-0.546***
Mo	0.01	-0.154	0.060		-0.082

*, **, *** Significant at the 0.05, 0.01, and 0.001 probability levels, respectively.

† Wright (1995).

‡ Total, pseudo-total soil concentration; oxalate, oxalate-extractable soil concentration; soluble, soluble/exchangeable soil concentration.

Table 2. Correlation coefficients of variables significantly correlated ($P < 0.05$, minimum $r > 0.240$) with I concentrations in pasture and soil ($n = 55-69$).

Medium	Variable†	Coefficient
Pasture	Zn soluble	0.344**
	S soluble	0.253*
	Na total	0.253*
	Mn pasture	0.246*
	B pasture	0.242*
	Si oxalate	-0.241*
	P pasture	-0.244*
	P total	-0.259*
	K pasture	-0.263*
	P oxalate	-0.267*
	Ca total	-0.269*
	S oxalate	-0.279*
	Pb oxalate	-0.281*
	N pasture	-0.288*
	Al oxalate	-0.306*
	Ca pasture	-0.310*
	Cr oxalate	-0.333**
	Fe oxalate	-0.372**
	DFS	-0.546***
	Soil	Al oxalate
S oxalate		0.821***
Si oxalate		0.675***
Cd oxalate		0.667***
Fe oxalate		0.652***
C total		0.638***
Cr oxalate		0.636***
Se total		0.589***
N total		0.578***
P total		0.553***
S total		0.525***
Cd total		0.523***
Al total		0.485***
Mo oxalate		0.426***
P oxalate		0.422***
Cu total		0.394***
Mo total		0.391***
Cr soluble		0.333*
Fe total		0.322**
Na total		0.296*
Cu oxalate	0.276*	
Cr total	0.271*	
Clay	0.265*	
Pb oxalate	0.264*	
P soluble	-0.247*	
Silt	-0.267*	
Se pasture	-0.293*	
Co soluble	-0.352**	

*, **, *** Significant at the 0.05, 0.01, and 0.001 probability levels, respectively.

† Total, pseudo-total soil concentration; oxalate, oxalate-extractable soil concentration; soluble, soluble/exchangeable 0.05 M Ca(NO₃)₂-extractable soil concentration, and C total assumed to be all organic. DFS, minimum distance to the sea.

Distance from the sea had the strongest effect on the concentration of I in pasture. Analysis (Table 2) found a strong correlation between these two factors ($p < 0.001$, $r = 0.546$). Pasture concentrations within 15 km from the sea were highly variable, ranging from 0.10 to 0.85 mg kg⁻¹, whereas at DFS >40 km, most samples were <0.12 mg kg⁻¹ except for four sites that may have received anthropogenic I (Fig. 2). Similar trends were observed with barley (*Hordeum vulgare* L.), pasture, wheat (*Triticum aestivum* L.), and willow shrub from Northern Ireland and Norway (Haug et al., 2012; Sivertsen et al., 2014; Bowley et al., 2017).

Iodine and other elements present in seawater may have been intercepted and retained by leaves or taken up by the roots, thus reducing their accumulation in the underlying soils. Several elements present at ≥ 0.01 mg L⁻¹ in seawater were measured for in soil and pasture, but only soluble S had a negative DFS–soil correlation, with B, I, Mg, Na, and S having negative DFS–pasture correlations (Table 1). The critical leaf area index of pasture is high, at approximately 4 to 6 for perennial ryegrass and 3 for white clover (Martin et al., 2017), hence high rates of sea-derived I interception is likely. Bowley (2013) found that washing did not significantly decrease vegetation I concentrations, which indicates that I was

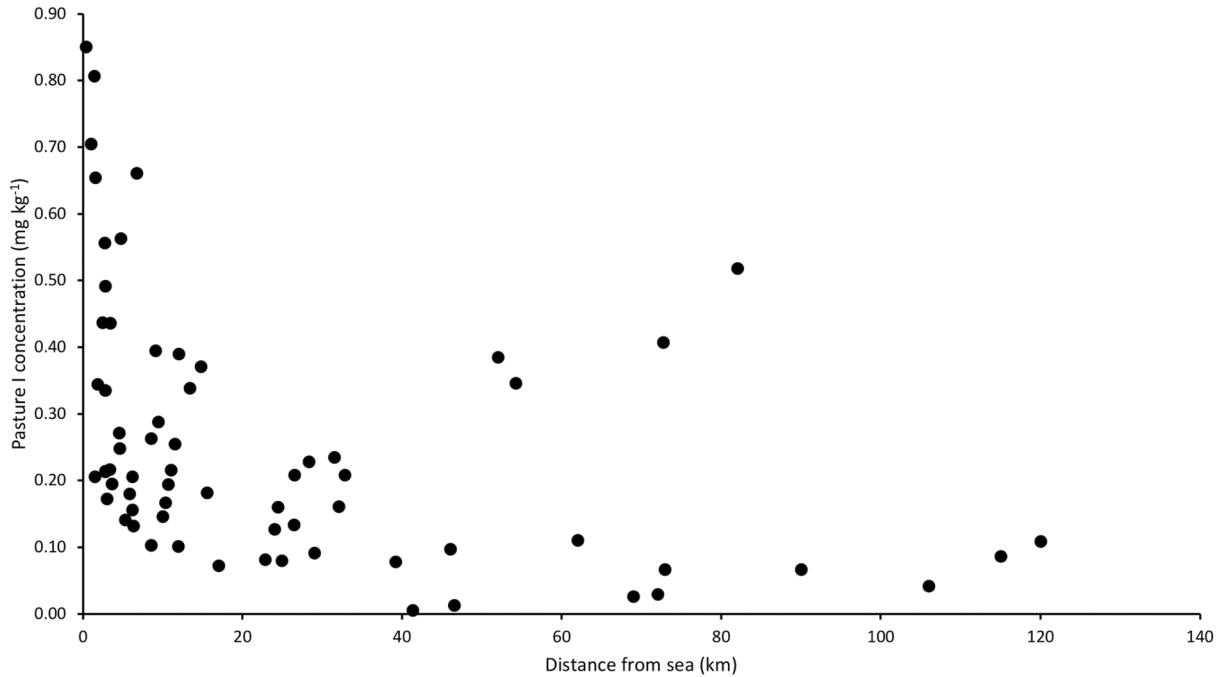


Fig. 1. Distance from the sea versus soil I concentrations ($n = 68$). Note that a single point containing 86 mg kg⁻¹ is not visible on this graph.

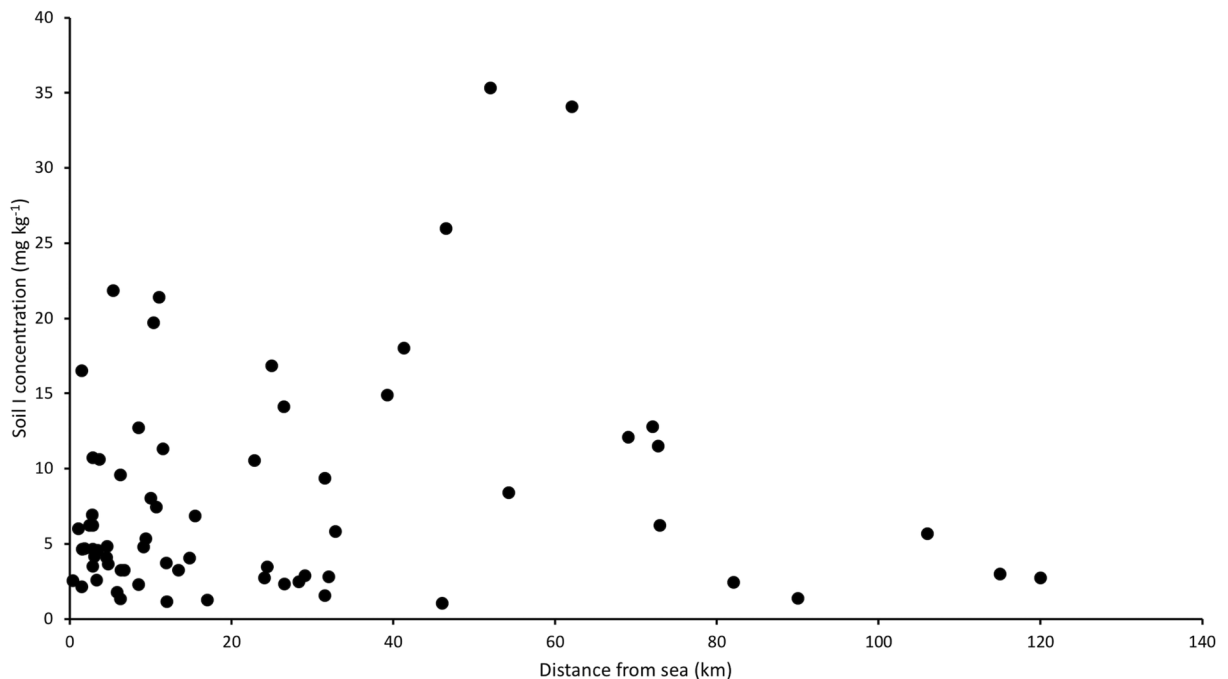


Fig. 2. Pasture I concentration (mg kg⁻¹) as a function of distance from the sea ($n = 67$). Note that a single point containing 1.4 mg kg⁻¹ is not visible on this graph.

not superficially retained, but inside the leaves, a finding replicated by Humphrey et al. (2019). Karunakara et al. (2018) showed that pasture can potentially intercept large quantities of I, with mass interception factors of 0.25 to 7.7 m² kg⁻¹, although decreasing with rainfall and total I.

Competition for soil sorption sites from other seawater anions, particularly halogens, may increase the potential for plant uptake and leaching (Sheppard et al., 1995). The increased solubility of I resulting from these competitive effects may contribute to the observed differences in the relationships between soil and pasture concentrations and the DFS (Table 1). The Fe and Al-silicate (oxyhydr)oxides fractions were negatively correlated with pasture I ($r = -0.241$ to -0.372) (Table 2). Hence, though this fraction retained I in soil, it appeared to also reduce I availability to plants.

Conclusions

This study indicates that without supplementation, sheep and cattle will become deficient in I on 26 and 87% of the pastures tested. There was no correlation between soil I and DFS, nor was there a correlation between soil I and pasture I. However, the critical factors affecting I in pasture were the DFS and the presence of amorphous oxides, with the highest pasture concentrations occurring next to the sea in soils with low amorphous oxides. Therefore, the risk of I deficiency in stock is higher away from the sea and in amorphous-oxide-rich soils. Future research should test the effect of soil properties including organic C and clay on pasture I >50 km away from the sea, include the soil soluble I fraction in similar datasets, and replicate studies of this nature, to determine whether our conclusions are applicable in contrasting environments, in particular tropical soils and arid soils.

Supplemental Material

The supplemental material provides the latitude and longitude of the sampling sites (Supplemental Table S1), a graphical depiction of the sampling sites (Supplemental Fig. S1), and the results of the analyses of the Certified Reference Materials.

Conflict of Interest

The authors declare no conflict of interest.

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