Elemental fluxes in *Elaeis guineensis* (oil palm) production: Implications for environmental quality

A thesis
submitted in partial fulfilment
of the requirements for the degree of

Doctor of Philosophy
In
Environmental Science

By
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Notes on the thesis

This thesis was conducted from 2019–2022 during the COVID-19 pandemic. Originally, the intention was to conduct a geographically extensive study of oil palm (*Elaeis guineensis* Jacq.) production systems throughout Indonesia. In August 2019, the Indonesian government introduced a law-change whereby any foreign researchers in Indonesia required government-approved permits, which were not forthcoming for *E. guineensis* research. The export of scientific samples was thus restricted. This, combined with the COVID-19 border closures limited the number of *E. guineensis* plantation sites in this thesis to four. Therefore, the thesis developed a mass-balance model, validated using New Zealand data, to calculate the likely elemental concentrations in a range of environmental conditions in Indonesia. Further investigations on the use of *E. guineensis* products in New Zealand, particularly palm kernel expeller, were undertaken.

The thesis comprises five manuscripts that have been submitted for publication. At the time of writing, two of these manuscripts have been published. As such, there may be changes between the remaining chapters and their respective published manuscripts. As these manuscripts are included here verbatim, there is repetition in parts of the thesis, particularly the introduction sections of the manuscripts. Note that the referencing is in the original journal styles and therefore not consistent between all chapters.

Abstract

Oil palm (*Elaeis guineensis* Jacq.) production currently covers 17,000,000 ha of land in Indonesia and Malaysia, with plantations increasing by 270,000 ha annually. Palm oil, 71,000,000 t per year, has only been a significant crop since the 1980s. Much of the land currently in *E. guineensis* production was formerly tropical rainforest. Rainforest clearing for *E. guineensis* production has been subsidised in Indonesia. These soils require significant agrichemical inputs, both fertiliser and pesticides, to maintain production. Potentially, these agrichemicals may result in the accumulation of some chemical elements in soils. For example, the application of phosphate fertilisers may result in the accumulation F, Zn, Cd and U, while some fungicides contain Cu or Mn as their active ingredients. Potentially, these elements may accumulate in soils and enter *E. guineensis* products. There is a lacuna of information on the fluxes of the chemical elements in the soil – plant system in *E. guineensis* production. This thesis aimed to determine the sustainability of *E. guineensis* production systems with respect to chemical elements and assess whether the accumulation of these chemical elements pose a risk to the safety of *E. guineensis* products. Soils and plants were sampled from four *E. guineensis* plantations in Sumatra, a key production region of Indonesia. Soils were also sampled from a rubber plantation for comparison. The chemical elements in soils, and plants were determined and the results compared with published data on adjacent rainforest soil. Palm oil and palm kernel expeller from several shipments imported into New Zealand were analysed, and the liver samples from animals fed with PKE were compared to non PKE-fed animals. A model was developed and validated to determine the likely accumulation of trace elements in *E. guineensis* production soils.

The soil concentrations of essential plant nutrients, especially N, P, K, Mg and Mo, were below levels required for the optimal production of *E. guineensis*. This indicates that production could be significantly increased with sufficient fertiliser addition. There was evidence that Cu, Zn, As and Pb were accumulating in *E. guineensis* production soils, although none had reached concentrations where they would reduce soil fertility. The concentrations of the chemical elements in *E. guineensis* fronds, rachides, mesocarps and endosperms also indicated deficiencies in the aforementioned chemical elements. Modelling indicated that within eight years under current production systems, F would accumulate to almost double its background concentrations, while Cu would increase approximately 14-fold. After 50 years, modelling indicated that these elements may detrimentally affect productivity, reaching 719 and 289 mg kg^{-1} respectively. While Cd concentrations increased, after 50 years, they would reach 0.76 mg kg^{1}, below the 1 mg kg^{1} threshold used in many jurisdictions.

No elements exceeded food safety standards in palm oil, although Al concentrations (159 mg kg^{-1}) were elevated relative to the *E. guineensis* tissues. Potentially, this Al may pose a risk to children consuming the average consumption volume of 37 g day^{-1} of palm oil. Palm kernel expeller contained P, Mg, Al and Fe concentrations that exceeded threshold concentrations for these elements in cattle fodder, while K, S and Cu were within 90% of threshold concentrations. Given that many of these elements were deficient in the *E. guineensis* production soils tested, the elevated concentrations in PKE may be the result of post-harvest contamination or of elevated concentrations of these elements in the areas where the PKE was produced. Potentially, Mg and Cu may be beneficial in New Zealand dairy systems, where PKE is used extensively as a supplementary feed. Copper is commonly deficient in New Zealand agricultural soils and milking cows require Mg supplementation to prevent milk fever. However, liver analyses indicated no statistically significant differences between PKE-fed animals and non-PKE fed animals with respect to concentrations of Cu.

In the *E. guineensis* production systems tested, it is likely that production would not be economically viable without subsidies. Several adjacent *E. guineensis* plantation areas sampled in this thesis had been abandoned possibly because they were economically unviable. Given the widespread environmental destruction resulting from *E. guineensis* production, there is an urgent need to determine whether removal of the subsidies may lead to better environmental and economic outcomes. Future work should involve widespread sampling of *E. guineensis* production soils, although this is challenging given restrictive policies regarding research in many *E. guineensis* production areas.

Thesis contributions and outputs

My contributions and the contributions of co-authors to the thesis are detailed in co-authorship forms for Chapters 1, 3, 4, 5 and 6, below.

Five manuscripts have been produced from the thesis:

Thompson-Morrison, H., Gaw, S., & Robinson, B. (2022). An assessment of trace element accumulation in palm oil production. *Sustainability, 14*(8), 4553. https://www.mdpi.com/2071-1050/14/8/4553?type=check_update&version=2

Thompson-Morrison, H., Gaw, S., Ariantiningsih, F., Arief, S. M., & Robinson, B. (2022). Chemical elements in plants and oils of current and former palm oil production systems [Submitted to *Scientific Reports*].

Thompson-Morrison, H., Gaw, S., Ariantiningsih, F., Arief, S. M., & Robinson, B. (2022). Chemical elements in soils of current and former palm oil production systems [Submitted to *Soil Use and Management*].

Thompson-Morrison, H., Moltchanova, E., Gaw, S., & Robinson, B. (2022). Elemental composition of palm kernel expeller used as supplementary stock fodder. *Sustainability, 14*(23), 15752; https://doi.org/10.3390/su142315752

Thompson-Morrison, H., Moltchanova, E., Gaw, S., & Robinson, B. (2022). Development and use of a mass-balance model to calculate the likely effects of agrichemicals on trace element accumulation in soils supporting palm oil production [Submitted to *Soil Use and Management*].

Presentations arising from this thesis include:

Thompson-Morrison, H., Moltchanova, E., Gaw, S., Robinson, B. (2022, 28 November–1 December). Validation and use of a mass-balance model for the calculation of trace element accumulation in soils [Conference presentation]. The New Zealand Society of Soil Science Conference, Blenheim, New Zealand.

Thompson-Morrison, H., Arif, S. M., Gaw, S., Robinson, B. (2021, 27 June–2 July). *Trace elements in palm oil production systems: Implications for New Zealand agriculture* [Conference presentation]. The Soil Science Australia and the New Zealand Society of Soil Science Joint Conference, Hamilton, New Zealand.

For which I was awarded the T. W. Walker prize for most meritorious student oral presentation.

Thompson-Morrison, H., Ariantiningsih, F., Arif, S. M., Gaw, S., Robinson, B. (2022, 31 July–5 August). *Trace elements in palm oil production systems: Implications for international agriculture* [Conference presentation]. World Congress of Soil Science, Glasgow, Scotland, United Kingdom.

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During my PhD I was awarded a FoodHQ Strategic Thinking in Agribusiness Management Programme Scholarship and the Evans Fund Scholarship.

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Published as:
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The candidate conducted 95% of the work. She took scientific and editing advice from the co-authors.

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Hadee organised the collection of samples, which was aided by co-authors. Hadee travelled to Indonesia and returned the samples to New Zealand, where she analysed them using appropriate methods. Hadee wrote the paper. Hadee conducted 90% of the work. She took scientific and editing advice from the co-authors.

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Chapter 4: Chemical elements in plants and oils from current and former palm oil production systems
Submitted to *Scientific Reports*

Hadee organised the collection of samples, which was aided by co-authors. Hadee travelled to Indonesia and returned the samples to New Zealand, where she analysed them using appropriate methods. Hadee wrote the paper. Hadee conducted 90% of the work. She took scientific and editing advice from the co-authors.

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Chapter 5: Elemental composition of palm kernel expeller used as supplementary stock fodder
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Please detail the nature and extent (%) of contribution by the candidate:
Hadee used data provided by VetLife for the pasture and animal data. She collected and analysed the PKE samples. Statistical advice — particularly the code used for the power analysis — and visualisations were provided by Elena Moltchanova. Hadee conducted 80% of the work. She took scientific and editing advice from the co-authors.

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Chapter 6: Development and use of a mass-balance model to calculate the likely effects of agrichemicals on trace element accumulation in soils supporting palm oil production
Submitted to *Soil Use and Management*

Please detail the nature and extent (%) of contribution by the candidate:
Hadee wrote the model in R from existing sources with advice and inputs from the co-authors. She validated the model and carried out the simulations for the oil palm plantation scenarios. Hadee wrote the manuscript with inputs from the co-authors. Elena Moltchanova wrote the equations in the manuscript. Hadee's contribution to the paper was 75%.

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To anyone I haven't named personally, know that I'm thankful for your help and support.

Tidak pernah mencoba, tidak pernah tau.

Table of Contents

List of abbreviations

ANOVA	Analysis of variance
BAC	Bioaccumulation coefficient
BS	Base saturation
CEC	Cation exchange capacity
EU	European Union
ICP-MS	Inductively coupled plasma-mass spectrometry
ICP-OES	Inductively coupled plasma-optimal emission spectroscopy
K_D	Soil adsorption coefficient
LUE	Land use efficiency
ME	Mean error
MP-AES	Microwave plasma-atomic emission spectroscopy
MTL	Maximum tolerable level
NPK	Nitrogen phosphorus potassium (fertiliser)
PC1	Principal component 1
PC2	Principal component 2
PCA	Principal component analysis
PKE	Palm kernel expeller
PUE	Phosphorus use efficiency
REE	Rare earth element
RMSE	Root mean square error
RSPO	Roundtable on Sustainable Palm Oil
SDG	Sustainable Development Goal
SGV	Soil guideline value
SRM	Standard reference material
TE	Trace element
US	United States
USDA	United States Department of Agriculture

Chapter 1: An assessment of trace element accumulation in palm oil production (General introduction)

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Abstract

African oil palm (*Elaeis guineensis* Jacq.) is grown on 17,000,000 hectares in Southeast Asia, producing oil and the by-product, palm kernel expeller (PKE), for export. *Elaeis guineensis* is typically produced on weathered acidic soils, with fertilisers and fungicides used to increase production. These amendments can contain elevated concentrations of trace elements (TEs), either as the active ingredient (e.g., Cu-based fungicides) or as contaminants, including F, Zn, As, Cd, Pb and U. TEs may accumulate in soil over time, and be taken up by plants, posing a food-chain transfer risk if allowed to exceed soil guideline values. We reviewed available literature on trace elements in soil, plant material, oil and PKE to evaluate the risk of TE accumulation due to phosphate fertiliser and Cu-fungicide use. TE concentrations of Cu, Zn, and Cd were reported to be up to 69, 107 and 5.2 mg kg^{-1}, respectively, in *E. guineensis* plantation soils, while Cu and As were reported to be up to 28.9 and 3.05 mg kg^{-1}, respectively, in PKE (>50% their permissible limits). Iron, a TE, has also been reported in PKE up to 6130 mg kg^{-1} (>10-fold the permissible limit). TE accumulation is an emerging issue for the palm oil industry, which, if unaddressed, will negatively affect the industry's economic and environmental sustainability. There are critical knowledge gaps concerning TEs in palm oil systems, including a general lack of research from Southeast Asian environments and information concerning key contaminants (Fe, Cu, As and Cd) in soils, plants and PKE.

Keywords

Copper; *Elaeis guineensis*; fertiliser contaminants; heavy metals; palm kernel expeller

1.1 Introduction

The African oil palm (*Elaeis guineensis* Jacq.) is the highest yielding oil crop, with a maximum potential yield of 8 t ha^{-1} [1]. From 2012–2020, *E. guineensis* represented 36–37% of vegetable oil production worldwide [2,3]. Indonesia and Malaysia supply ca. 85% of the world's palm oil, with a combined harvestable area of more than 17,000,000 ha [4,5]. Thailand, Colombia and Nigeria are the third-, fourth- and fifth-highest producing countries of palm oil, respectively [6]. Palm oil is used in products for health care, home care and hygiene, as well as in many processed foods [7]. A dry matter by-product of palm oil production, palm kernel expeller (PKE), is sold as a supplementary stockfeed. In 2020, worldwide PKE production reached 10,214,000 t [8].

Production of *E. guineensis* in Southeast Asia occurs on tropical soils, which are typically weathered and acidic, and, as such, require significant fertiliser inputs to meet nutrient demands. Fungicides are applied to control plant disease [9–11]. Without fertiliser applications of Nitrogen (N), Phosphorus (P), Potassium (K), Magnesium (Mg) and Boron (B), *E. guineensis* trees will become nutrient deficient, yields will decline, and production may become uneconomic, particularly for smallholders in Southeast Asian environments [11,12]. Applications of Copper (Cu) and Zinc (Zn) are also critical when growing *E. guineensis* on Histosol soils [11]. Fertiliser trials have shown that yields may reach 8 t oil ha^{-1} yr^{-1} with appropriate fertiliser management; however, the average yield is 4 t ha^{-1} yr^{-1} [1,13,14]. Fungicidal treatments using Cu as an active ingredient are commonly applied to prevent plant diseases, including Ganoderma spp. [15]. Fertilizers and fungicides can contain significant concentrations of trace elements (TEs), either as part of the active ingredient [16] or asa contaminant [17]. TE contaminants in agrichemicals can accumulate in soil [18], which can result in reduced soil fertility [19] and/or uptake into the (human) food chain [20,21].

A combination of poor environmental controls (in the form of poor monitoring and enforcement of fertiliser-use recommendations) and a lack of knowledge about soil nutrient status and plant requirements often leads to inappropriate fertiliser applications, particularly in Southeast Asian smallholder plantations [22]. As well as having demonstrable negative effects on fruit-bunch yield [12], inefficient nutrient management can lead to the contamination of soils and surface and groundwaters due to N leaching or the run-off of P and TEs [23]. Similarly, TE-containing fungicides may be overapplied, accelerating TE accumulation in soil. Cadmium (Cd) from fertilisers and Cu from fungicides are two of the most likely TE contaminants to accumulate in horticultural soils [24,25]; however, other fertiliser-borne contaminants include Fluorine (F), Chromium (Cr), Zn, Arsenic (As) and Lead (Pb) [17]. While there are disparate reports on TE concentrations in some components of *E. guineensis* production, there is no information available on fluxes throughout the whole system. This review aims to identify key TEs of concern, elemental fluxes, potential risks to ecosystems and human health, and knowledge gaps concerning this production system.

1.2 Trace elements in soils of *Elaeis guineensis* production

1.2.1 Sources of trace elements in *Elaeis guineensis* plantation soils

1.2.1.1 Geogenic trace elements

Soils naturally contain TEs, with concentrations varying geographically due to biogeochemical processes [26]. In tropical regions, weathering and climate are key factors influencing background TE concentrations of soils [27,28]. Food-chain transfer risk may occur when naturally high TE soils with low pH and organic matter content are used to produce food crops, as these conditions favour TE mobility and subsequent plant uptake [29,30]. *Elaeis guineensis* in Southeast Asia is typically cultivated on acidic, weathered soils, including Ultisols, Inceptisols, and Oxisols, as well as Histosols [11] according to the USDA Soil Classification [31]. Ultisols and Oxisols are generally highly leached and contain TEs, including Cobalt (Co), Nickel (Ni), Cu, Zn, Cd, Mercury (Hg) and Pb [32]. General background concentrations of these TEs in soils are 8, 40, 20, 50, 0.06, 0.03 and 10 mg kg^{-1}, respectively [26]. Histosols contain TEs (Titanium (Ti), Manganese (Mn), F and Barium (Ba) at background concentrations of 5000, 850, 200 and 500 mg kg^{-1}, respectively [26,33], which are released as the soil degrades under drainage and cultivation processes [34].

1.2.1.2 Anthropogenic sources of trace elements in soils

Most TEs that enter soils via agrichemical use are rapidly immobilised through the processes of specific absorption, occlusion and precipitation [35]. Therefore, most TEs will accumulate in soil following repeated applications of agrichemicals. TEs may be lost from the soil via surface erosion or run-off, while leaching of TEs is usually negligible [36]. Tropical soils in Southeast Asia typically contain deficient levels of P to support *E. guineensis* production, thus requiring fertiliser inputs to provide plants with the required nutrients [11,37]. A study of smallholder plantations in Indonesia by Woittiez et al. [22] identified that Indonesian farmers relied heavily on subsidised fertilisers, which did not supply soils with appropriate nutrients for *E. guineensis* production. More than 99% of smallholders surveyed used mineral fertilisers, including several phosphatic fertilisers. These fertilisers can contain contaminants including F, Cu, Zn, As, Cd, Pb and Uranium (U), which can result in soil contamination and human exposure risk if inappropriately or excessively applied [17,38]. Rare earth elements (REEs) are also present in phosphate fertilisers and may enter the soil-plant system through regular use of these [39,40]. The rate of use and application of the most commonly used fertilisers reported by Woittiez et al. are presented in Table 1.1, as well as the average mass of key elements added as fertiliser contaminants through such applications.

Table 1.1. Fertiliser use and application rates in smallholder plantations in Indonesia, reported by Woittiez et al. [22] with associated contaminant inputs of F, Cu, Zn, Cd, Pb and U.

Fertiliser (NPK ratio)	**Use (% of farmers surveyed)**	**Application rate (kg ha^{-1} yr^{-1})**	**Approximate mass added (g ha^{-1} yr^{-1})**					
			F [a]	**Cu** [a]	**Zn** [a]	**Cd** [b]	**Pb** [a]	**U** [a]
NPK Phonska (15–15–15)	66	692	15,916	64	261	4.8–48	3.5	25
NPK Pelangi (15–15–15)	9	756	17,388	70	285	5.2–52	3.8	27
Single super-phosphate (0–36–0)	21	452	6396	26	94	7.5–75	9.04	24
Triple super-phosphate (0–46–0)	7	400	8400	17	146	8.4–84	6.8	47
Rock phosphate (0–20–0)	1	1000	29,856	22	245	9.2–92	12	92

[a] Calculated using average total contaminant concentrations in phosphate fertilisers from Taylor et al. [38]. [b] Possible range given due to the differing contaminant concentrations of Cd in phosphate rock dependent on geographical origin (20–200 mg Cd kg $P_2O_5^{-1}$ in sedimentary rock [41]).

The majority of smallholders surveyed by Woittiez et al. [22] applied excessive amounts of fertilisers compared with the nutritional requirements of the crop and employed practices likely to lead to nutrient run-off and leaching. Such practices included weed-clearing in plantations using glyphosate and paraquat, which left soil bare and vulnerable to erosion and transfer to kernels. Farmers also manually (un-uniformly) mixed straight fertilisers, applied fertilisers in bulk once per year, and applied fertilisers in a narrow ring around the base of the palm, rather than uniformly to the soil area colonised by palm roots or on top of decomposing palm fronds, as is recommended as good practice [42–44]. Conversely, some farmers applied insufficient fertilisers for *E. guineensis* production (18% applied no K, 20% no P, and 15% no N), leading to decreased yield and soil nutrient deficiencies. Smallholders manage more than 40% of *E. guineensis* plantations in Indonesia (4,700,000 ha) [45], and inefficient nutrient management over this area is both a significant risk to the economic sustainability of this industry as well as a driver of deforestation, as more land area is needed to produce economically viable yields [22].

Elaeis guineensis production can be reduced by diseases; in particular, the fungal pathogen *Ganoderma* spp., which causes trunk rot, is especially problematic in Southeast Asian plantations [9,10,46]. Copper-based fungicides are commonly employed to control this disease, with available products having varying active ingredient (Cu) concentrations from 7.6–75% [10,15,47]. Bivi et al. [15] found that continuous Cu applications totalling 21.45 g Cu ha^{-1} yr^{-1} (combined with Calcium (Ca) and salicylic acid) were effective in controlling basal stem rot in *E. guineensis* plants (This application rate was calculated using data from Bivi et al. [15] with a tree density of 143 trees ha^{-1}, described as optimal by FAO [48]). However, the actual application rates used by farmers may be in excess of this due to the uncontrolled nature of fungicide use in Indonesia [49]. There is a lack of data available on actual Cu application rates utilised in *E. guineensis* plantations. In Europe, past fungicide application rates have been as high as 80 kg Cu ha^{-1} on agricultural soils and have led to soil contamination [50]. Current European Union guidelines allow for applications of 6 kg Cu ha^{-1} on agricultural soils [51]. Only one Indonesian study on fungicide use could be found for comparison [49]. Here, fungicide application rates used in hot pepper cultivation in Indonesia are reported as a mean of 14.3 kg active ingredient ha^{-1} and a maximum of 33.8 kg active ingredient ha^{-1}. Copper was an active ingredient in the fungicides used in this study [52]. The variation in application rates observed in this study fits with that observed throughout other systems: for example, Morgan and Taylor [53] reported median Cu application rates of 2 kg ha^{-1} yr^{-1}, with the highest application rate being 16 kg ha^{-1} yr^{-1} in New Zealand vineyards. Repeated applications of Cu-containing products to horticultural crops can result in Cu accumulation in soils [54,55] through the return of fungicide-applied plant material to the soil surface and through spray drift landing on the soil surface during application. In excessive quantities, Cu in soils can inhibit plant growth and pose risks to human health [56,57].

1.2.2 Measured trace element concentrations in soil

Available data on trace element concentrations in *E. guineensis* plantation soils are presented in Table 1.2. Two soils from Papua New Guinean *E. guineensis* plantations have Cd concentrations <1 mg kg^{-1} [58]. Aini Azura et al. [59] reported Zn and Cd concentrations up to 99 and 5.2 mg kg^{-1}, respectively, in Malaysian *E. guineensis* production soils. These are the only two studies to be identified which were conducted in Southeast Asia, and thus there is a lack of relevant research looking at critical TEs, including F, Cu and Pb, in a Southeast Asian environment.

Table 1.2. Trace element concentrations in *E. guineensis* plantation soils (0–30 cm depth unless otherwise stated) with average concentrations for soil, rocks and Earth's crust.

Element	**Concentration range (mg kg^{-1})**	**Average concentrations of earth materials** [a]				
		Soil (mg kg^{-1})	**Earth's crust (mg kg^{-1})**	**Granite (mg kg^{-1})**	**Sandstones (mg kg^{-1})**	**Igneous rocks (mg kg^{-1})**
V	0.56–4.9 [b]	100	135	17	20	135
Cr	26–132 [c]	100	100	20	35	100
Mn	2.4–20.5 [b] 0.12–29 [c] 20.6–81 [d]	850	950	195	X0 [h]	950
Co	0.80–9.4 [c] 0.03–2.9 [d]	8	25	2.4	0.3	25
Ni	1.5–13 [b] 30.3–59 [c]	40	75	1	2	75
Cu	1.1–9.8 [b] 3.9–6.5 [e] 13–69 [c] 3.06–18 [d]	20	55	13	X h	55
Zn	2.09–25 [b] 4.8–6.2 [e] 12–107 [c] 0.85–6.8 [d] 11–99 [f]	50	70	45	16	70
Mo	<0.01–1.05 [d]	2	1.5	6.5	0.2	1.5
Cd	0.22–2.0 [b] 0.32–4.3 [c] 0.45–5.2 [f] <1 [g]	0.06	0.2	0.03	0.0X [h]	0.08
Pb	6.8–14 [c]	10	13	48	7	13

[a] [26]. [b] Nigeria [60]. [c] Nigeria, range of means [63]. [d] Ghana, 0–40 cm depth [64]. [e] Nigeria [61]. [f] Malaysia [59]. [g] Papua New Guinea [58]. [h] X denotes an order of magnitude estimate.

In Nigeria, TEs have been reported present in concentrations that both exceed and fall short of optimal concentrations for *E. guineensis* cultivation. Orobator et al. [60] reported significant differences in Mn, Iron (Fe), and Cd concentrations across three plantation sites, with Cu and Zn found to be deficient at one site and no excessive TE concentrations observed. In a separate study, *E. guineensis* plantation soil reportedly contained lower concentrations of Cu and Zn than secondary forest soils [61]. Soil Cr and Pb concentrations have been reported at background levels by Uwumarongie-Ilori et al. [62]. In contrast to these studies, Olafisoye et al. [63] reported Cd concentrations exceeding background concentrations (0.5 mg kg^{-1}) in 15 plantation soils, with the highest soil containing 4.3 mg kg^{-1} Cd. This was attributed to the repeated use of phosphate fertilisers. Additionally, the authors reported that total Cr, Co, Ni and Pb concentrations exceeded maximum permissible levels for soils (100, 8, 40, and 10 mg kg^{-1}, respectively). The maximum reported soil copper concentration was 61 mg kg^{-1} and was concluded to pose a toxicity risk to plants and animals. *Elaeis guineensis* production soils in Ghana were reported as Molybdenum (Mo) deficient by Golow et al. [64]. In all studies mentioned here, soil TE levels were found to be either deficient (Cu, Zn, Mo) or excessive (Cr, Co, Ni, Cu, Cd, Pb) relative to the requirement and tolerance of *E. guineensis*. This underlines the need to match on-farm management with soil status, especially in systems with high nutrient demands, such as palm cultivation on weathered tropical soils. Notably, no data could be found on F, As, or U concentrations in soil under *E. guineensis* production.

The TE concentrations in the aforementioned reports will be a function of both the agrichemicals used and the time that the land has been under agricultural production, either as *E. guineensis* or a previous crop(s). Given that many *E. guineensis* plantations have recently been converted from virgin forests, TE concentrations may still be similar to background levels. Therefore we might hypothesise that TE concentrations in African soils, which were converted to *E. guineensis* some 100 years ago [13], will have higher TE concentrations than recently converted soils in Southeast Asia.

1.3 Trace elements in *Elaeis guineensis* and comparable species

There are three varieties of *E. guineensis*: pisifera, dura, and the hybrid variety tenera [65]. Var. pisifera typically has no endocarp and a thick mesocarp, var. dura a thick mesocarp and endocarp, and var. tenera a moderately thick mesocarp and thin endocarp, with a moderately sized endosperm [13]. This hybrid variety was developed to produce higher oil-yielding crops and now dominates most plantations in Southeast Asia [66].

Table 1.3. Trace element concentrations in *E. guineensis* kernels from Malaysia and Nigeria.

Element	**Mean concentration range in Malaysian kernels (mg kg^{-1})**	**Mean concentration range in Nigerian kernels (mg kg^{-1})**	**Mean concentration in terrestrial plants (mg kg^{-1}) [a]**
Mn	82–145 [b]	410–610 [c]	630
Fe	43–52 [b]	110–220 [c]	140
Cu	16–18 [b]	17–26 [c]	14
Zn	25–36 [b] 4–13 [d]	26–43 [c]	100
Cd	0.09–0.31 [d]	—	0.6
Pb	—	≤0.05 [b]	2.7

[a] [26]. [b] Endosperm of seed- and clonally-derived var. tenera [70]. [c] Defatted and undefatted var. tenera and var. dura [71]. [d] Estimated from graph [59].

Table 1.4. Trace element concentrations in materials of other Arecaceae species.

Element	**Mesocarp concentration *P. dactylifera* (mg/kg^{-1})**	**Seed concentration *P. dactylifera* (mg/kg^{-1})**	**Leaf concentration *P. dactylifera* (mg/kg^{-1})**	**Mesocarp concentration *E. oleracea* Mart. (mg/kg^{-1})**	**Mesocarp concentration *E. edulis* Mart. (mg kg^{-1})**
Li	<0.007–0.17 [a]	<0.007–0.017 [a]	—	—	—
Al	48.4 (2.6) [b]	—	—	—	—
Cl	3340 (280) [b]	—	—	—	—
Sc	0.028 (0.004) [b]	—	—	—	—
V	<0.008–0.016 [a]	<0.008–0.021 [a]	—	—	—
Cr	0.49 (0.5) [b]	—	0.18–0.99 [c]	—	—
Mn	1.0–7.0 [a] 7.5 (0.3) [b]	2.4–11.5 [a]	0.35–0.96 [c]	—	—
Fe	2.0–7.0 [a] 197 (10) [b]	3.2–30.9 [a]	1.6–9.4 [c]	—	—
Co	0.026–5.1 [a] 0.025 (0.003) [b]	0.075–3.20 [a]	—	—	—
Ni	0.071–0.70 [a]	0.15–0.69 [a]	0.022–0.083 [c]	—	—
Cu	0.7–7.2 [a]	1.3–8.4 [a]	2.0–9.6 [c]	—	—
Zn	1.4–12.6 [a] 9.5 (0.5) [b]	3.9–28 [a]	0.6–3.5 [c]	—	—
As	<0.04–0.051 [a] 1.9 (0.17) [b]	<0.04–0.089 [a]	—	0.0095 [d]	—
Se	<0.1–0.120 [a] 0.102 (0.013) [b]	<0.1–0.3 [a]	—	—	—
Br	3.2 (0.15) [b]	—	—	—	—
Rb	5.4 (0.5) [b]	—	—	—	—
Sr	1.1–14.8 [a] 13.9 (1.2) [b]	0.21–5.2 [a]	—	—	—
Mo	0.18 (0.05) [b]	—	—	—	—
Cd	<0.002–0.013 [a] 0.08–0.23 [e]	<0.002–0.012 [a]	0.043–0.19 [c]	0.0094 [d]	—
La	0.36 (0.04) [b]	—	—	8.03–230 [f]	17–199 [f]
Ce	0.47 (0.05) [b]	—	—	20.1–575 [f]	36–319 [f]
Hg	0.051 (0.003) [b]	—	—	0.0016 [d]	—
Pb	<0.02–0.14 [a] 0.84–2.3 [e]	<0.02–0.11 [a]	0.22–1.98 [c]	0.037 [d]	—
Th	—	—	—	0.99–179 [f]	15–87 [f]

[a] Spain [72]. [b] Pakistan, number in brackets assumed to be standard deviation of the mean, *n* ≥ 6 [73]. [c] Turkey, range of means [76]. [d] [74]. [e] Saudi Arabia, range of means [75]. [f] [40].

Monocotyledonous plants, including *E. guineensis,* uptake TEs from soil via exchange sites on root surfaces that are then transported to the above-ground portions, especially the leaves, via xylem transport, using the symplastic and apoplastic pathways. Leaf tissue, a water sink, is expected to have higher concentrations of TEs relative to other tissue. To reach the fruit, however, TEs must be transported in the phloem and maybe traverse the placenta, making fruit tissues (mesocarp and seed) likely to contain lower TE concentrations [67]. In *E. guineensis* production systems, leaves (fronds) are often returned to soil as organic matter inputs [68,69]. If high levels of TEs are contained within these leaves, or if TEs are sorbed to palm fronds after fungicide spray applications, this may result in accumulation of TEs on the soil surface following leaf abscission. The study mentioned in Section 2.2 by Aini Azura et al. [56] is the only study that could be identified which investigated TE concentrations in *E. guineensis* fronds, reporting concentrations of ca. 0.18–0.38 mg kg^{-1} for Cd and ca. 15–28 mg kg^{-1} for Zn. Only three relevant studies could be identified which addressed TE concentrations in *E. guineensis* kernels. Nigerian kernels were generally reported to contain higher levels of Mn, Fe, Cu and Zn (up to 610, 220, 26 and 43 mg kg^{-1}, respectively) relative to Malaysian kernels (up to 145, 52, 18 and 36 mg kg^{-1}, respectively) [70,71]. Lead was reported below detection levels in Nigerian kernels, while Cd was reported up to ca. 0.31 mg kg^{-1} in Malaysian kernels [59,71]. Mean concentrations of TEs in *E. guineensis* kernels from these studies are compared with average TE concentrations of land plants in Table 1.3.

While research on TEs in *E. guineensis* plant material is sparse, data have been collected on TEs in other Arecaceae species, including date palm (*Phoenix dactylifera* L.), acai (*Euterpe oleracea* Mart.), and jucara (*Euterpe edulis* Mart.) (Table 1.4). Due to their similar physiologies, TEs in these species may behave similarly to those in *E. guineensis*. As such, it is beneficial here to review the literature concerning TEs in these Arecaceae species to garner insights into whether a wider range of TEs may accumulate in *E. guineensis.*

Accumulation of Cr, Co and Ni has been reported in mesocarp and seed up to concentrations of 0.5, 5.1 and 0.7 mg kg^{-1}, respectively [72]. In a separate study, As was reported in mesocarp samples at 0.17 mg kg^{-1}, although this did not exceed tolerable dietary levels [73]. Comparable concentrations were reported for Cd in *P. dactylifera* and *E. oleracea*, of up to 0.13 and 0.0094 mg kg^{-1}, respectively [72,74]. Both leaves and mesocarp of *P. dactylifera* have been shown to take up TEs from the air in contaminated environments, with TE concentrations increasing in areas with increased anthropogenic air pollution sources [75,76].

Rare earth element concentrations of *E. oleracea* and *E. edulis* were reported by Santos et al. [40]. *Euterpe oleracea* contained the highest concentrations of REEs, up to 230, 575 and 179 mg kg^{-1} for La, Ce and Th, respectively. It was also reported that geographical location affected REE concentrations of both *E. oleracea* and *E. edulis*. This emphasises the variation which can occur in the TE content of plant tissues due to geographical and geological factors. Tables of REE contents in Arecaceae species are provided in Appendix A.

The TE concentrations detailed in Table 1.4 indicate that TE accumulation occurs in palm species when exposure pathways are present. This indicates that TE accumulation in *E. guineensis* tissues and associated products may occur when plants are grown in contaminated environments.

1.4 Trace elements in palm oils

The *E. guineensis* tree produces two types of oil from its fruit: palm kernel oil from the endosperm; and palm oil from the mesocarp. While the two names are often used interchangeably, palm oil is commonly used for food products due to its near-even balance of unsaturated and saturated fatty acids and its alpha- and beta-carotenes, while palm kernel oil is more commonly used in other products such as soaps and cosmetics and is less saturated relative to palm oil [77]. Red palm oil is a further differentiated oil and refers to palm oil with a high carotenoid content giving it a dark red colour.

Trace elements may be present in palm and palm kernel oil as a result of processing (including refining, manufacturing, storage, shipping and packaging) and processing equipment due to *E. guineensis* plant uptake from soils or from soil contamination on kernels (i.e., dust from harvesting on associated soils) [7,78]. Trace elements in palm oils may occur as complexed impurities and contribute to dietary trace nutrient intake, which may provide nutritional benefits or, in excessive quantities, may breach food safety standards and pose risks to human health when ingested [7,79]. Here, research on TEs in palm oils is reviewed. A summary of available data of TEs in palm and palm kernel oils is presented in Table 1.5.

Crude palm oil is extracted from the mesocarp of *E. guineensis* either by solvent (chemical) or physical (manual crushing of palm kernels) extraction [80]. It may then be further refined by rinsing with hot demineralised water; degumming with citric acid; adsorptive cleaning, bleaching, and filtering to remove TEs and other impurities; and deodorising to remove volatile compounds [7]. Non-uniform manufacturing processes can affect TE concentrations in palm oil. In an analysis of oil at all stages of the above refining process, Szydłowska-Czerniak et al. [7] found that refining removed 72%, 94% and 63% of Fe, Cu and Pb from crude palm oil, respectively. A more than three-fold increase in Pb was noted between the filtering and deodorising steps and attributed to contamination from the equipment used in this step of the refining process. Copper concentrations in crude palm oil (0.46 mg kg^{-1}) exceeded Polish standards of 0.40 mg kg^{-1}. However, the final product fell within the acceptable range of <0.10 mg kg^{-1}. No other exceedances of standards were noted by the authors. A similar study of the same TEs was conducted on Indonesian crude palm oil by Rossi et al. [81]. They concluded that using phosphoric rather than citric acid as a degumming agent increased Pb concentrations in degummed oil; however, concentrations of Cu and Pb in the fully refined product were less than the suggested limit of 0.1 mg kg^{-1} [82]. These two studies demonstrate how non-uniform manufacturing processes impact the TE content of marketed palm oils.

Two studies by Chen et al. [83] and Chen et al. [84] determined As concentrations in refined and fractionated palm oils from Taiwan, with both studies reporting low As concentrations (≤0.025 mg kg^{-1}). Limits for As in oils have not yet been determined, and the toxicity risk depends on the type of As present (organic or inorganic) [85]. Australasian food standards set a limit of 1 mg kg^{-1} total As for cereals and 0.5 mg kg^{-1} for salt; however, limits are often set only for inorganic As due to its higher toxicity [86]. Consequently, research on As speciation in palm oils may be beneficial for the purpose of addressing the risk of As-toxicity.

Table 1.5. Elemental concentrations in palm and palm kernel oils.

Element	**Concentration in palm kernel oil ($mg\ kg^{-1}$)**	**Concentration in palm oil ($mg\ kg^{-1}$)**
Al	38.30 (0.58) [a]	31.00 (0.56) [a] 1.9 [b]
Cl	22.21 (0.79) [a]	29.60 (0.74) [a]
V	0.055 (0.009) [a]	0.065 (0.007) [a]
Cr	—	2.3 [b] 0.101–0.298 [c] 0.021–0.033 [d]
Mn	1.45 (0.03) [a]	0.94 (0.02) [a] 0.24–1.1 [e] 6.55–12.05 [c]
Fe	20.04 (0.20) [f]	11 [b] 15–35 [e] 65–232 [c] 38.3–78.3 [g] 0.12 (0.0058) [h] 0.27–2.40 [i]
Co	—	0.000–0.064 [e]
Ni	—	0.000–0.79 [e] 0.15–0.81 [c] 0.044–0.068 [d]
Cu	6.0 (0.35) [a]	1.4 (0.09) [a] 0.071 [b] 0.000–0.25 [e] 0.56–2.09 [c] 0.030 (0.001) [h] 0.03–0.05 [i]
Zn	2.82 (0.30) [f]	0.45–1.6 [e] 3.6–14.6 [c] 0.05–0.24 [g]
As	—	0.001–0.0025 [d] 0.025 [j] <0.015 [k]
Cd	—	0.022 [b] 0.024–0.094 [c] 0.025–0.065 [d]
Hg	—	0–0.055 [d]
Pb	—	0.018 [b] 0.024–0.067 [c] 0.023–0.038 [d] 0.0060 (0.0003) [h] <0.005 [i]

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[a] Nigeria, neutron activation analysis, number in brackets assumed to be standard deviation of the mean, number of samples not indicated [93]. [b] Nigeria [88]. [c] Nigeria [90]. [d] Nigeria, range of means, red palm oil [94]. [e] Nigeria, range of means [87]. [f] Nigeria, number in brackets assumed to be standard deviation of the mean, number of samples not indicated [92]. [g] Nigeria [89]. [h] Poland, number in brackets is standard deviation of the mean (n = 3) of final palm oil product after deodorising, analysed by inductively coupled plasma [7]. [i] Indonesia, steam refined oil [81]. [j] Taiwan [83]. [k] Taiwan [84].

Various studies have investigated elemental concentrations of Nigerian palm oil [87–90]. Collectively, Al, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg and Pb have been analysed using atomic absorption spectrometry methods. All elements were reported present at concentrations that posed no risk to human health according to standards set by Nigerian regulatory bodies; however, atomic absorption spectrometry may not be a sufficiently sensitive method for this analysis [91]. Further research analysing TEs in Nigerian palm oil using more analytically sensitive methods, e.g., optical emission spectrometry or mass spectrometry, would be beneficial.

Two studies were identified which investigated TE concentrations in palm kernel oil [92,93]. Here, reported Al, Cl, V, Mn, Fe, Cu and Zn concentrations were comparable to those of palm oils in other studies (Table 1.5).

While multiple studies have addressed TEs in Nigerian palm oils, there is a scarcity of research on palm kernel oil and oils from other countries, particularly Malaysia and Indonesia. This is significant since these countries produce almost 85% of the world's traded palm oil supply [95]. Malaysian and Indonesian production soils differ from each other and from other palm-producing countries, and there are various methods of oil extraction used across different plantation types. There is little current knowledge on the risks or benefits of TEs in palm oil and palm kernel oil as a result of these differing soils, plantation types, and production practices in Southeast Asia.

1.5 Trace elements in palm kernel expeller

Palm kernel expeller is the dry matter by-product of palm oil production, which is exported by manufacturing countries—primarily Indonesia and Malaysia—and utilised as a stockfeed in many Asian, European and Oceanic nations. Of these, New Zealand imports the largest amount of PKE annually, at 2,300,000 t in 2019 [96]. Although studies (and indeed importers) rarely differentiate between palm oils produced by physical and solvent extraction, Alimon [80] asserts that the latter leaves less oil in the PKE by-product (1–2%) compared to physical extraction (4–8%). Oil contents ranging up to 17.3% have been reported elsewhere [97]. Physical extraction is most commonly used as it is a more economical option [80]. As with palm oils, TEs may occur in PKE as a result of plant TE-uptake from soil, machinery and equipment used in manufacturing processes, and from soil contamination [7,78]. The TE content of PKE as a supplementary stockfeed has implications for animal and human health. Accordingly, Grace and Knowles [98] acknowledged that attention must be paid to the cumulative intake of minerals by animals from non-pasture-based feeds to ensure TE toxicity does not occur. Available information on PKE TE concentrations is presented in Table 1.6.

Three studies could be identified which have investigated TE concentrations of PKE [80,99,100]. Of these, two reported TE concentrations within acceptable ranges, or below Maximum Tolerable Levels (MTLs), for animal feed [80,100]. These include Mn, Zn, Se and Mo at concentrations below 2000, 500, 5 and 5 mg kg^{-1}, respectively [101].

Copper and As have been reported at concentrations that exceed 50% of their MTLs of 40 (This MTL is for animals fed a diet of 1–2 mg kg^{-1} Mo and 1500–2500 mg kg^{-1} S: Cu may become toxic at lower concentrations if dietary Mo and S are below these levels) and 4 mg kg^{-1}, respectively [80,99,100]. Alimon [80] noted that Cu concentrations of PKE up to 28.9 mg kg^{-1} exceeded ruminant requirements, and animals fed a diet of >50% PKE may develop Cu-toxicity symptoms. An ovine feed experiment on the occurrence of Cu-toxicity by Hair Bejo and Alimon [102] corroborates this point: all animals in PKE-fed groups not supplemented with Zn died before the study was completed due to Cu-toxicity from PKE, while those in Zn-supplemented and control groups survived. Thus, while high Cu concentrations in PKE can be a source of risk for livestock in receiving environments, this may be mitigated with appropriate TE management. Conversely, if managed correctly, the Cu content of PKE may provide a benefit to countries such as New Zealand, where background soil Cu concentrations are low, and farmers must supplement livestock at an expense [103].

Iron has been reported in PKE at concentrations up to 6130 mg kg^{-1} [80]. This upper concentration range is >10-fold higher than is permissible in cattle feed (500 mg kg^{-1} [101]). It is likely that this Fe is a result of contamination during manufacturing processes, as this concentration is far in excess of upper range kernel concentrations reported by Akpanabiatu et al. [71] (220 mg kg^{-1}) and average land plant Fe concentrations (140 mg kg^{-1} [26]). Excessive Fe intake in bovines can decrease milk production and body weight, with minimum deleterious doses estimated below 30 mg kg^{-1} day^{-1} [105]. Thus, there is an apparent risk of TE-toxicity for animals fed PKE containing TEs of these concentrations.

Table 1.6. Trace element concentrations in PKE from Malaysia and MTLs for cattle feed.

Element	**Reported concentration (mg kg^{-1})**	**MTL for cattle feed (mg kg^{-1})**
Mn	225 [a] 132–340 [b]	2000 [d]
Fe	4.05 [a] 835–6130 [b]	500 [d]
Cu	28.5 [a] 20.5–28.9 [b]	40 d,e
Zn	77 [a] 40.5–50.0 [b]	500 [d]
As	0.18–3.05 [c]	4 f
Se	0.23–0.30 [b]	5 d
Mo	0.70–0.79 [b]	5 d

[a] [100]. [b] [80]. [c] [99]. [d] [101]. [e] This value assumes a dietary Mo concentration of 1–2 mg kg^{-1} and S concentration of 1500–2500 mg kg^{-1}. With less Mo and S in animals' diets, Cu toxicity may occur at lower levels. [f] [104].

While attention has been given to Cu-toxicity risk in animals consuming PKE, other elements identified in the above-mentioned publications may also be of concern. In particular, Fe may be present in concentrations that pose risks to milk yield and animal health. Further research on the overall mineral composition of PKE would be beneficial, as there is a lack of data available on the concentrations of other non-essential biotoxic TEs, including Pb and Cd, which may be present in PKE due to their presence as impurities in phosphate fertilisers used in *E. guineensis* production [17,37]. Furthermore, there is a lack of research on PKE sourced from outside Malaysia and from different plantation types (including smallholder, community managed, government- and corporate-owned), whose management practices result in non-uniform application and accumulation of TEs in soil, plants and end-products [37]. Overall, there are scarce data available on the TE content of PKE and filling this knowledge gap would be beneficial for all countries utilising this by-product in their agricultural systems by way of maintaining livestock well-being and soil nutrient status.

1.6 Conclusions

This review has identified the elements of concern in *E. guineensis* production systems as Fe, Cu, Zn, Cd and As. Copper, Zn and Cd have been reported in production soils at elevated concentrations and may pose risks to production if allowed to accumulate past soil guideline values. These TEs are present and accumulate in *E. guineensis* production soils, likely as a result of phosphate fertiliser and Cu-fungicide use in plantations. Iron may be present in PKE in concentrations >10-fold higher than its MTL—likely as a result of contamination during PKE processing—while Cu and As have been reported to be in excess of 50% of their MTLs. Using PKE for animal fodder may present risks or a benefit to the agricultural systems of importing countries depending on what TEs are present and in what concentrations. There is a disparity between regions where *E. guineensis* is produced— namely Indonesia and Malaysia, and regions producing research on TEs in *E. guineensis* systems—most often Nigeria. The critical knowledge gaps concerning TEs in *E. guineensis* production systems are: data on the elemental composition of PKE; the concentrations of Cu, Zn, As and Cd in Southeast Asian production soils; and data on mercury across the entire production system, from soils to products. Future research should focus on a whole-systems analysis to determine the viability of *E. guineensis* production systems and identify appreciable risks to allow the development of mitigation strategies that may be applied at the plantation level and address the emerging issue of TE contamination.

1.7 Statements and declarations

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su14084553/s1, Table S1: Rare earth element concentrations in Aceraceae species.

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1.9 Summary of knowledge gaps

While there is much scientific literature on the ecological effects of *E. guineensis* production, there is a lacuna of knowledge on soils under *E. guineensis*. Specifically, Chapter 1 indicated that it there is likely accumulation of TEs in *E. guineensis* production soils. However, reports on soil elemental concentrations are inconsistent and unverified. There is an urgent need to quantify the rate of elemental accumulation in *E. guineensis* systems to determine the sustainability of the system. Specific knowledge gaps are:

1. There is no whole-system analysis to determine the viability of *E. guineensis* production systems and identify appreciable risks which would allow the development of mitigation strategies that can be applied at the plantation level.
2. Copper, Zn and Cd have been reported in production soils at elevated concentrations and may pose risks to production if allowed to accumulate past soil guideline values. However, the chemical status of *E. guineensis* production soils, particularly those from Southeast Asia, is unclear and there is no information on other elements, such as F and As, which may be contaminants.
3. There is a lack of data available on the overall elemental composition of PKE, which may affect animal health if elements breach maximum tolerable levels for animal fodder. Chapter 1 indicates that elements including Fe, Cu and As concentrations in PKE may approach or exceed maximum tolerable levels for animal fodder.
4. There is a lack of research on PKE sourced from outside Malaysia and from different plantation types with varying management practices that may result in non-uniform application and accumulation of TEs in soil, plants, and end-products.

1.10 Research hypotheses

The hypotheses in this thesis are that:

1. The concentrations of some chemical elements, in particular Cu, Zn and Cd, will exceed background concentrations in *E. guineensis* soils due to the repeated application of fertiliser and pesticides containing elevated concentrations of these elements.

2. *Elaeis guineensis* production may not be sustainable due to the accumulation of some of the elements listed in Hypothesis 1.

3. The *E. guineensis* product PKE will also contain elevated concentrations of the aforementioned elements, as well as elements that may be introduced post-harvest.

4. Palm kernel expeller will introduce significant quantities of both essential and non-essential elements into the diets of New Zealand dairy cattle and that this may be detected in liver samples.

1.11 Research aims and objectives

This research aims to determine the fluxes of elements in *E. guineensis* plantations and the implications for environmental quality in producing agroecosystems as well as agroecosystems importing *E. guineensis* products.

Specifically, this research aims to determine:

1. The concentrations of chemical elements in selected *E. guineensis* production soils.

2. Elemental uptake by *E. guineensis* trees in terms of plant nutrition, contamination and food safety standards.

3. Elemental fluxes associated with export of PKE and implications for its use to support dairy platforms

4. To develop a validated mass-balance model to calculate the likely rate of accumulation of TEs in *E. guineensis* production systems.

1.12 Outline of Chapters 2–7

Chapter 2 provides background information relevant to this thesis. This includes the context in which palm oil has come to dominate global vegetable oil production, palm oil production and processing in an Indonesian context, PKE use in New Zealand, relevant *E. guineensis* biology and properties of TEs likely to be accumulating in *E. guineensis* production soils.

Chapter 3 explores elemental concentrations of soils from four *E. guineensis* plantations in Indonesia. This chapter assesses the nutrient status of these soils and determines whether TE accumulation is occurring as a result of production.

Chapter 4 explores elemental concentrations of plant tissue — including pinnae, rachides, mesocarps and endosperms — sampled from three *E. guineensis* plantations in Indonesia. The elemental concentrations in palm oil processed in Indonesia and sourced in New Zealand are measured. These concentrations are discussed in the context of plant nutrition, contamination and food and fodder safety standards.

Chapter 5 assesses the elemental composition of five shipments of PKE imported into New Zealand. The implications of this work are discussed in the context of animal health and farm management in New Zealand. A pilot study using secondary data on Cu concentrations in dairy cattle livers is undertaken to determine whether PKE may be affecting the liver status of dairy cows in New Zealand.

Chapter 6 develops and validates a mass-balance model for the calculation of likely TE accumulation in agricultural soils as a result of agrichemical use. This model is used to calculate the rates of accumulation of F, Cu, Cd and U likely to accumulate in *E. guineensis* production soils into the future.

Chapter 7 provides a discussion of information presented in the preceding chapters. This includes phosphorus use efficiency in *E. guineensis* plantations, the potential for correcting soil nutrient deficiencies, the sustainability of oil palm production and implications of this production system of environmental quality both in Indonesia and New Zealand.

Chapter 2: Background information to the thesis

2.1 The rise of palm oil production

Global consumption of vegetable oil has grown exponentially from the mid-1900s (Figure 2.1). As the establishment of global trade routes and expanding crop cultivation allowed vegetable oils to replace animal fats as the dominantly consumed fat-source, production grew into the 20th century (Robins, 2018). Initially, the prevailing use for vegetable oils was in the manufacture of margarine and soap (Gruen & Poggendorff, 1948). However, due to advances in chemistry and food technology, the use of vegetable oil has broadened to food, hygiene and personal care products (Aguiar et al., 2018; Matthäus, 2007). Consequently, the current global supply of vegetable oil is substantially higher, relative to population density, than it was up until the 1960s: while population density has doubled, global vegetable oil production has increased approximately ten-fold (Figure 2.1). Vegetable oils have become prolific ingredients in household products to the extent that supply has surpassed global population growth: oil production is driven primarily by per capita increases in consumption. Palm oil, produced from the African oil palm (*Elaeis guineensis* Jacq.) is the most produced and consumed vegetable oil, making up 35% of global production from 2021-2022 (Shahbandeh, 2022). In 2018, 71,000,000 t of palm oil was produced (Ritchie & Roser, 2021).

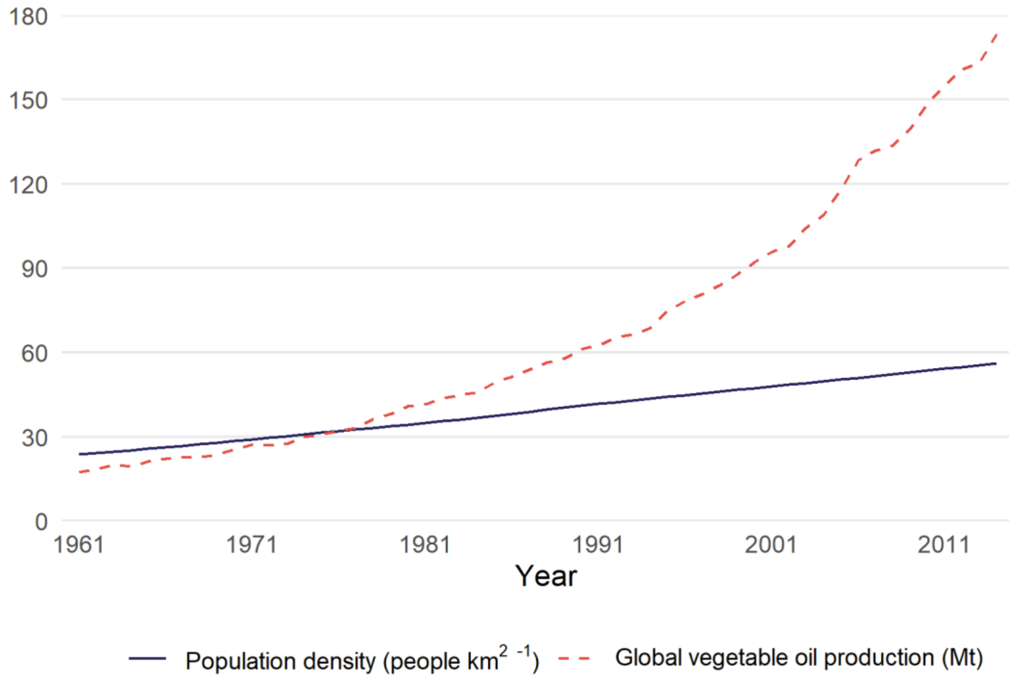

Figure 2.1. Global population density and global vegetable oil production from 1961–2014. Produced using data from Roser et al. (2013) and Our World in Data (2019).

The first commercial uses for palm and palm kernel oils outside of Africa (the natural range of *E. guineensis*) were in non-food products including soap and axle grease (Robins, 2018). Historically, palm oil was viewed as undesirable for human consumption due to its odour, taste and high free fatty acid content (Robins, 2018). However, as food chemistry advanced, it became increasingly viable to hydrogenate, fractionate and bleach palm oil for use in food products. Oleochemicals and surfactants are easily manufactured from palm oil, and it has a stable oxidative state (Parsons et al., 2020). Palm oil has proved to be one of the most versatile oils in terms of fractionation and this, combined with its high smoke point, has made it an attractive ingredient in a range of food and non-food products (Matthäus, 2007; Parsons et al., 2020). As such, palm oil has become a component in products which were traditionally palm oil-free, such as soaps, make up and biofuels (Aguiar et al., 2018). Palm oil has been produced in West African countries including Nigeria for more than 100 years (FAO, 2002a). Since the 1980s when palm plantations began to expand into Southeast Asia, *E. guineensis* has become the world's dominant vegetable oil crop, surpassing soybean (*Glycine max*) in 2001 (Our World in Data, 2019). Today, as well as food, it is used in health care, home care and hygiene products. The biggest palm oil-using manufacturer, Unilever, used 1,544,142 t of palm oil in its products in 2014 (Consumer NZ, 2016). Rainforest Action Network (2011) estimated that around 50% of household items contain palm oil or its derivatives. Palm oil is also heavily used in the biofuel sector, included as an ingredient in aviation fuel and biodiesel (Padfield et al., 2019). The European Union (EU) has been one of the biggest supporters for the use of palm oil in biofuels and imports both palm oil for processing into biofuel and pre-processed palm oil-based biofuel (Gerasimchuk & Koh, 2013). Approximately 1,900,000 t of palm oil was imported into the EU in 2013 for biodiesel production (Gerasimchuk & Koh, 2013).

2.2 Palm oil production in Indonesia

Commercial production of palm oil expanded outside of Africa and became established in Southeast Asia – primarily Indonesia and Malaysia – in the 1980s (Corley & Tinker, 2015). Following this, production volumes increased and costs dropped below other vegetable oils (Corley & Tinker, 2015; Lai et al., 2015). The establishment of plantations in Southeast Asia largely occurred on soils which, up until that point, had supported virgin forest (Vijay et al., 2016). Conversion to *E. guineensis* agriculture was financially incentivised, particularly in Indonesia, where both the trees and maintenance fertilisers were subsidised, and funds were allocated for plantation establishment and land clearing (Afriyanti et al., 2019; Kissinger, 2016; Sari et al., 2019; Woittiez et al., 2018). Other Southeast Asian countries including Thailand have followed suit with subsidised for *E. guineensis* production (Arunmas, 2022). In 1994, Indonesia was deemed to have approximately 60,000,000 ha of land suitable for agricultural production, of which half was already under cultivation (UNEP & FAO, 1994). Today, more than 55,000,000 ha in Indonesia (29% of total land area) is under agricultural production, with 12,500,000 ha (6.6% of total area, or 23% of all agricultural land) in *E. guineensis* cultivation (Index Mundi, 2022b; Quincieu, 2015). National policies and international investment accelerated production: from 2013-2019, Indonesia received foreign direct investment into agriculture, forestry and fishing at rates of more than 15 times the world average (FAO, 2022). This was due primarily to palm oil. In 2014, foreign direct investment in Indonesian *E. guineensis* agriculture peaked at USD$13,9000,000,000 (Pasaribu et al., 2021). Palm oil plantations exist as state-owned enterprises (as well as private enterprises and smallholders) (Chalil, 2008), which are subsidised and incentivised by internal tax policies (Pasaribu et al., 2021). Furthermore, the use of palm oil in biofuels is heavily subsidised by the EU, resulting in increased export of palm oil to EU countries, driving production volume (Gerasimchuk & Koh, 2013). These factors have supported the expansion of *E. guineensis* agriculture in Indonesia to the extent that they currently supply >50% of the world's palm oil (Index Mundi, 2022a). Barriers to entry in the Indonesian palm oil sector are high as large amounts of both capital and land are required, and up until 1996 entry into this sector was incentivised with government concessions and loans (Chalil, 2008). These concessions have since been removed, however, subsidies persist in the Indonesian palm oil sector. When barriers to entry are high and subsidies are present, an industry can become devoid of competition, and production may become inefficient (Chalil, 2008). Inefficient production necessitates more land area to maintain production volume. This results in greater encroachment of *E. guineensis* into surrounding land, that may contain ecologically valuable ecosystems (Xu et al., 2020).

Since 1989, some 45% of land converted to *E. guineensis* production in Southeast Asia was transitioned from primary forest (Vijay et al., 2016). In major producing countries, during the period 2000-2011, an average of 270,000 ha of forest was converted to *E. guineensis* production annually (Henders et al., 2015). Most scientific attention has focused on the impact of *E. guineensis* monocultures and plantations on rainforest habitats of threatened megafauna. In contrast, there is a lacuna of knowledge on the impact of *E. guineensis* agriculture on soil resources (Padfield et al., 2019).

Whether palm oil would remain competitive without subsidies is unclear. Furthermore, whether the existence of distorting subsidies has been a driver of the global supply of vegetable oil, to the point where it is produced in quantities which far exceed human population growth (Figure 2.1), is a question concerning both economies and environments.

2.3 The political climate in Indonesia, as it relates to *Elaeis guineensis* agriculture and soil management

The political climate in Indonesia is not conducive to sustainable management of agricultural systems. The country scores 38 out of 100 on Transparency International’s global corruption scale (Merkle, 2018), meaning corruption exists in Indonesia throughout bureaucratic, judiciary and police systems, from central government down to local levels. The TRACE International Bribery Index (Trace International, 2017) found Indonesia to have low-medium levels of anti-bribery enforcement and transparency concerning financial interests, while Bertelsmann Stiftung (2018) concluded that along with poor public infrastructure, low education levels and poverty, corruption severely limits the Indonesian government’s capacity to effectively govern (Merkle, 2018). Furthermore, the World Bank (2017) Control of Corruption Indicator scores Indonesia at -0.25 on a scale of -2.5 – 2.5, indicating that corruption remains rampant throughout governance systems.

This national context has allowed the development of the palm oil industry to grow largely unregulated, in part because this industry has played a key role in Indonesia’s economic development. Specifically, licenses for logging concessions to establish plantations on certain land may be given out multiple times in overlapping fashion to different companies, and officials running in elections have been known to give concessions in return for financial campaign support (Mathieson, 2016; Merkle, 2018). Indonesia is known for its ‘powerful domestic vested interests’ (Merkle, 2018, p. 5) which have come to the forefront of international media attention in varying forms, including criticism over forest fires and subsequent haze affecting neighbouring countries in 2015 and 2019 as a result of slash-and-burn forest clearance to make space for *E. guineensis* plantations ("Indonesia haze: Why do forests keep burning?," 2019; "Indonesia province shuts schools as haze from fires returns," 2019; Jong, 2019a, 2019c). An Indonesian government audit found 81% of *E.* guineensis plantations breached regulations, including failing to obtain permits, non-compliance with sustainability standards, and encroachment into protected forests (Jong, 2019b). Indonesia's anti-corruption commission produced a similar audit in 2016, which concluded the palm oil industry suffered from a lack of accountability and credibility to prevent corruption and violations of regulations (Komisi Pemberantasan Korupsi, 2016).

Unregulated production, combined with subsidised fertiliser products has resulted in fertiliser application without consideration of the nutrient status of the soil, particularly in smallholder plantations (Woittiez et al., 2015; Wortmann et al., 2019). This has resulted in both nutrient deficiencies and the application of excess fertiliser, particularly in smallholder plantations (Woittiez et al., 2018). *Elaeis guineensis* requires a tropical climate with substantial water and nutrient inputs: key nutrients for production are N (recommended maintenance inputs 214.5–286 kg ha^{-1}), P (143–214.5 kg ha^{-1}), K (357.5–429 kg ha^{-1}), Mg (214.5–286 kg ha^{-1}) and B (14.3–28.6 kg ha^{-1}) (Woittiez et al., 2015). Subsidised fertilisers are relied on, which are not always agronomic options for production. In a survey of smallholder plantations, 73% had soil K deficiencies with mean concentrations below 0.15 cmol K kg^{-1} (Woittiez et al., 2015). Approximately 80% of smallholders surveyed used subsidised NPK (Phonska, NPK 15-15-15), a blend which contains unsuitable nutrient ratios for *E. guineensis* growth (Woittiez et al., 2015). As well as fertilisers and pesticides used in production, inexpensive biowaste products, such as empty *E. guineensis* fruit bunches (returned to the plantation once the kernels have been removed at a palm oil processing mills) and palm oil mill effluent (a waste product from palm oil processing at mills) are sometimes returned to plantation soil as amendments (Figure 2.2). While these may aid soil fertility, they may also carry TE contaminants which accumulate in soils.

Figure 2.2 a (left) *Elaeis guineensis* empty fruit bunches returned to plantations and laid in a ring around the base of palms as organic matter inputs; and b (right) *E. guineensis* empty fruit bunches and fronds returned to plantation soils as organic amendments.

2.4 The Roundtable on Sustainable Palm Oil

In response to mounting criticism around environmental impacts of *E. guineensis* production, the Roundtable on Sustainable Palm Oil (RSPO) was founded by the World Wide Fund for Nature, Malaysian Palm Oil Association, Unilever, Migros and AAK (RSPO, 2022) as an initiative to promote sustainable management practices in plantations. The RSPO encourages companies at any stage of the production chain using palm oil to apply for accreditation to become members. These members then agree to adhere to guidelines that aim to increase the social, economic and environmental sustainability of the palm oil industry (RSPO, 2019). A key focus of the RSPO is to stop deforestation due to increased land use conversion to *E. guineensis* cultivation. The RSPO (2019) claim that due to their efforts, 19% of palm oil and palm oil products worldwide come from sustainable sources, however, they have attracted criticism for their lack of rigid environmental regulations, the continued orangutan fatalities that occur in *E. guineensis* plantations and the continuation of deforestation and burning of peatlands to clear land for *E. guineensis* conversion (Environmental Investigation Agency, 2018; Jong, 2018; Kilvert, 2018). A 2018 study (Morgans et al., 2018) determined that there was no significant difference between plantations that were RSPO certified and those that were not, in terms of the following social, environmental and economic metrics: availability of rural health facilities, number of households receiving government assisted income, orangutan presence in areas with substantial palm plantations (orangutans are a key species threatened by habitat loss due to *E. guineensis* expansion), number of fire incidents to clear land, fresh fruit bunch yield and share price. Similarly, Carlson et al. (2018) reported that sustainable certification did not affect deforestation in peatlands. Morgans et al. (2018) called for a more rigid implementation of the RSPO's criteria to improve sustainability of the industry and more research to be conducted in this area. The RSPO does not consider soil or any aspect of its management in its Code of Conduct or Membership Rules (RSPO, 2022). Without assessing soil quality and management, RSPO certification does not necessarily indicate the sustainability of *E. guneensis* production.

2.5 New Zealand's role in the palm oil industry

Annually, New Zealand imports approximately 2,000,000 t of palm kernel expeller (PKE), a by-product of palm oil production, as a supplementary stockfeed on many dairy farms (Index Mundi, 2022c). Palm kernel expeller is protein and fibre rich, and is moderately palatable (PGG Wrightsons, 2019). New Zealand imports refined palm oil and products containing its associated fractionations. The two-year rolling average value of New Zealand PKE imports from 2020-2022 was approximately US$450-500,000,000 (G. Aspinall, personal communication, July 8th 2022). Therefore, New Zealand has a significant economic investment in the palm oil industry.

Common deficiencies in New Zealand agricultural soils include Cu, Co, Se and I, which affects farm management practices as farmers must supplement stockfeed to ensure the continued health and well-being of livestock (Morton & Roberts, 2018). Supplements of Mg and Zn are also used on-farm to prevent milk fever and facial eczema in dairy cattle, respectively (Cuttance et al., 2016; Thilsing-Hansen et al., 2002). The TE composition of PKE is currently unknown and investigating this will elucidate the potential of this by-product to be used as a cost-effective supplement stockfeed in place of traditional supplements, or conversely, whether any TE contamination is likely to pose risks to New Zealand systems. Such risks include impacts on animal nutrition, food-chain transfer impacting human health, and impacts on soil nutrient status through nutrients returned to soil in animal excretions from TEs entering New Zealand systems from imported stockfeed.

Palm kernel expeller is imported into New Zealand by several companies including Agrifeeds and ADM from Indonesia and Malaysia. Agrifeeds was initially a joint venture between Fonterra, New Zealand's largest dairy co-operative, and Wilmar International, the world's largest palm oil production company, however, in 2020 Fonterra sold its shares to Wilmar International (Kissun, 2020). Agrifeeds sources all of its PKE from Wilmar's processing facilities, 88% of which are in Indonesia (Agrifeeds, 2020a; 2020b). Agrifeeds reports that during 2020, 99% of its imported PKE was traceable to individual palm oil mills however, only 12% was traceable to individual plantations (Agrifeeds, 2020). Since 2018, Fonterra has penalised farmers for feeding PKE using their Fat Evaluation Index Grading System (Fonterra, 2017), as PKE changes the composition of cows' milk, increasing the fat content (Van Wyngaard & Meeske, 2017). Excess fat content results in milk that is deemed unsuitable for manufacturing products to customer specifications (Fonterra, 2017). Synlait, another New Zealand dairy company, disincentivises the use of PKE by paying farmers a premium for milk that is produced without the use of PKE (Synlait, 2018). Thus, while still widely used, PKE is not actively encouraged in all New Zealand dairy systems.

2.6 Biology of *Elaeis guineensis*

Elaeis guineensis (Figures 2.3, 2.4) is a monocotyledonous plant of the Arecaceae family in the order Arecales. It is an angiosperm, native to West Africa and prefers a tropical, humid climate (FAO, 2002b). It grows optimally in regions up to with a latitude of <10°, primarily in lowland areas. The ideal temperature range is 24-28°C, with limited growth <15°C (Corley & Tinker, 2015). The plant is tolerant to intermittent waterlogging and grows well in moist soils (Corley & Tinker, 2015). If left unpruned, trees can reach 20 m tall with a crown of 30-50 leaves (Corley & Tinker, 2015), however, when cultivated, their older leaves are pruned and trees are typically felled and replanted after 20-30 years as they become too tall to harvest (Figure 2.3). Plants are monoecious, with male and female inflorescences and are pollinated largely by insects, in particular *Elaeidobius kamerunicu*, the African oil palm weevil (Yousefi et al., 2020). Plants begin producing after approximately three years and produce year-round (MPOC, 2022). Palms are typically planted in a hexagonal grid pattern in plantations at a density of 143 trees ha^{-1} to maximise the use of space (FAO, 1990).

Elaeis guineensis trees have four root classes: primary roots (6-10 mm in diameter) of which there are thousands, secondary roots (2-4 mm in diameter), tertiary roots (0.7-1.2 mm in diameter) and quaternary roots (0.1-0.3 mm in diameter) (Corley & Tinker, 2015). Roots do not have hairs, and surface roots down to 30 cm are used for water and nutrient uptake while deeper roots are thought to be primarily for stability (Corley & Tinker, 2015). The processes of elemental uptake by plants are introduced in Chapter 4.

Elaeis guineensis has three varieties: pisifera, dura and the hybrid variety tenera (Lim, 2012). Var. *dura* is the most common naturally occurring variety in Africa and has a thick endocarp (2-8 mm) and 35-65% mesocarp (Corley & Tinker, 2015). Var. *pisifera* has no endocarp and 92-97% mesocarp, while var. *tenera* is a cross between the two with a thin endocarp (0.5-4 mm), small endosperm (3-15%), and 55-96% mesocarp (Corley & Tinker, 2015). This variety was developed to produce higher oil-yielding crops.

Figure 2.3. Fully grown *E. guineensis* tree. Note that this photo was taken in an abandoned plantation and under normal production practices, no understorey would be present.

Figure 2.4. *Elaeis guineensis.* A. Whole plant. B. Male inflorescence. C. Single branch of male inflorescence. D. Part of a female inflorescence. E. Infructescence. 1 bud; 2 male flowers; 3 male flowers in longitudinal section; 4 steel tube; 5 female flower; 6 female flower in longitudinal section; 7 ovaries in cross section; 8 fruit; 9 fruit after the upper half of the pulp has been removed; 10 stone core in cross section; 11 stone cores in longitudinal section; 12 a two-seeded stone core in cross-section; 13 seeds; 14 seeds in cross section; 15 seeds in longitudinal section. Image created by Franz Eugen Köhler, Köhler's Medizinal-Pflanzen 1897, used under Creative Commons (CC 1.0) Universal Public Domain Dedication.

2.7 The palm oil process: From plantation to product

Elaeis guineensis trees are typically grown from seedlings (Figure 2.5) that can be harvested after three years (MPOC, 2022). Fresh fruit bunches (Figures 2.6 and 2.7), each comprised of 1000–3000 fruitlets (Figure 2.8), grow near the base of the fronds (FAO, 2002a) (Figure 2.9). Fresh fruit bunches are harvested by hand using a blade on the end of a long pole, which cuts the bunch off the tree. Fresh fruit bunches typically reach weights of 25 kg (FAO, 2002a). These are collected from plantations and transported to palm oil mills where they are processed into crude palm oil.

Figure 2.5. *Elaeis guineensis* seedlings in a nursery plantation before planting for production

Figure 2.6. A harvested *E. guineensis* fresh fruit bunch

Figure 2.7. *Elaeis guineensis* fruit bunches stacked after harvest, before transportation to a palm oil processing mill.

Figure 2.8. Individual *E. guineensis* fruitlets.

Figure 2.9. Fronds, containing pinnae and rachides, of *E. guineensis.*

Once at the mills, fresh fruit bunches are cleaned with steam and stripped of fruitlets (MPI, 2015). The remaining empty fruit bunches are sometimes returned to plantations as organic inputs. Fruits are then separated into mesocarp and endosperm (kernel) fractions. Crude palm oil is expressed from the mesocarp, while palm kernel oil is expressed from the endosperm, however, sometimes literature (and palm oil products) does not differentiate, labelling both kinds as ‘palm oil’. Crude oil goes through several refining processes to produce edible palm oil (explained in Section 1.4). The remaining mesocarp fraction is referred to as fibre and may be used as fuel stock for mill boilers (MPI, 2015). The remaining dry matter material from the endosperm is heat treated and ground to produce PKE (Figure 2.10) (MPI, 2015). Water is used at various stages throughout this process (Figure 2.11) and the resultant wet waste product is known as palm oil mill effluent (Baharuddin et al., 2010). Approximately 3 t of palm oil mill effluent are produced for every t of crude palm oil processed (Osman et al., 2020). This liquid waste has a high biological and chemical oxygen demand and is typically discharged to water (Corley & Tinker, 2015; Osman et al., 2020). Thus, the inputs to the palm processing system are water (as water and steam) and fresh fruit bunches and the outputs are empty fruit bunches, palm oil, palm kernel oil, PKE, mesocarp fibre and palm oil mill effluent (Figure 2.11). Due to the many steps in this production process and the range of machinery used, there are multiple opportunities for contamination to enter this system post-harvest. Metal contaminants may come from machinery while water used in mills may carry elemental contaminants. Once processed, products are shipped vast distances and further opportunities for contamination exist here from materials used for packaging and shipping. The potential for contamination exists at multiple stages throughout the palm oil growth, harvest and pressing process. Maximum tolerable levels for TEs in animal fodder are available in Appendix B.

Figure 2.10 Palm kernel expeller a (left) stored after processing; and b (right) fed to cows as supplementary stockfeed.

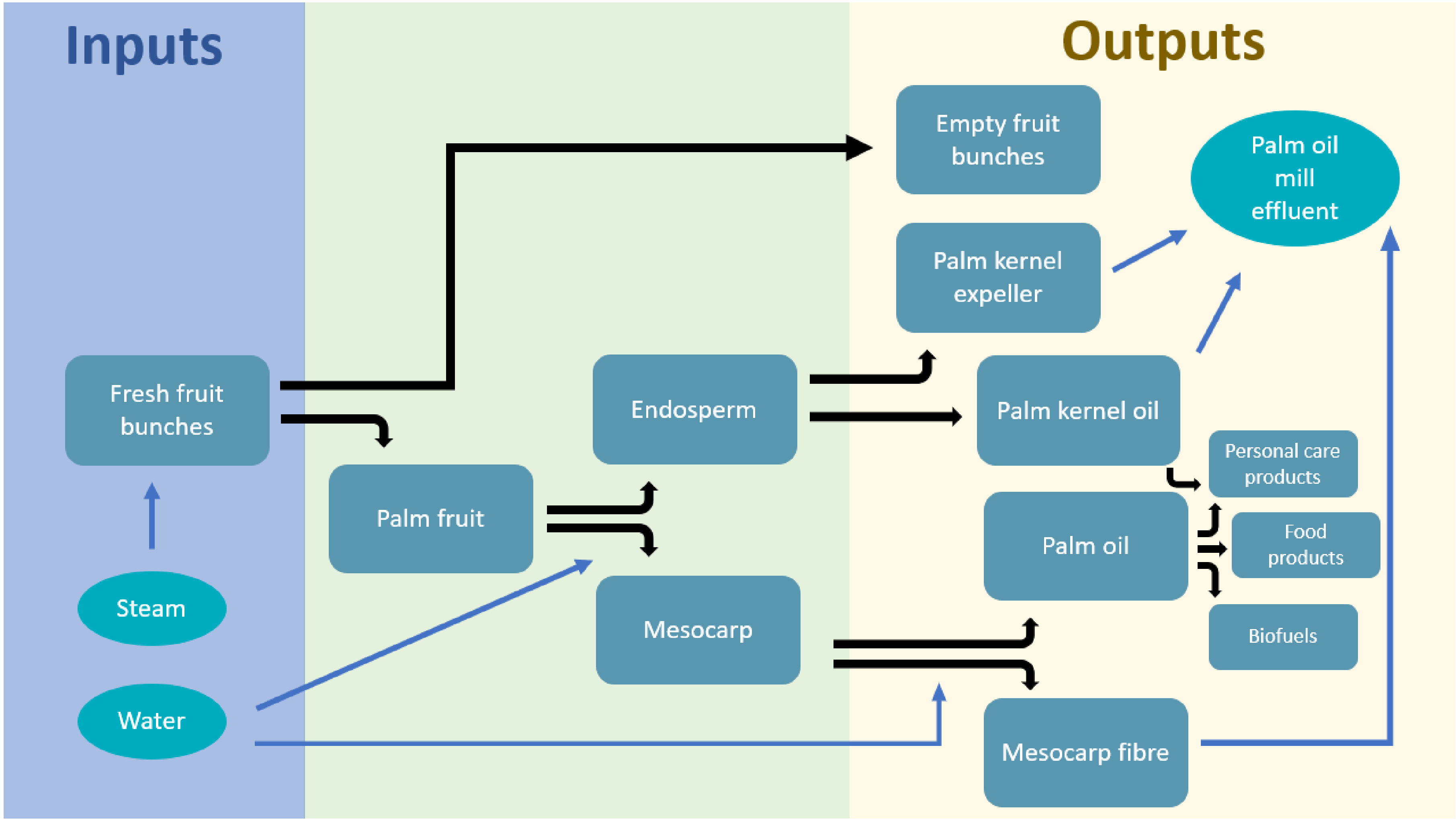

Figure 2.11. The palm oil production process at the palm oil mill, detailing inputs and outputs.

2.8 Properties of trace elements that are likely to occur at elevated concentrations in *Elaeis guineensis* production systems and their movement through the environment

Note: this thesis focuses on a whole-system approach to determining TE fluxes within oil E. guineensis production systems, rather than the mechanisms of TE uptake that occur in the rhizosphere. Information on the latter can be found in Gobran and Huang (2011).

Production of *E. guineensis* involves the use of fertiliser, fungicides and organic amendments that are often unregulated and applied at un-agronomic rates (Woittiez et al., 2015; Wortmann et al., 2019). As a result, deficiency or contamination of certain TEs may go unchecked within production systems. Current research on TE fluxes within these systems is lacking.

Copper, Zn and Cd are of particular interest regarding *E. guineensis* agroecosystems. Zinc and Cd are common contaminants in agricultural fertilisers (Taylor et al., 2016), while Cu is a key component to many fungicidal products (Copper Development Association, 2013). Accumulation of these elements in soils, plant material and *E. guineensis* products is possible considering the lack of stringent management guidelines in *E. guineensis* plantations, and the unregulated nature of smallholder production in many Global South countries (Wortmann et al., 2019). Accumulation of Cu, Zn and Cd may occur under *E. guineensis* plantations that require high levels of fertiliser amendments to maintain productivity. Accumulation of key elements can have negative effects on soil microorganisms, animal and human health, land use flexibility and international trade (Rys, 2011). Furthermore, the lack of consistent data on these TEs in *E. guineensis* systems calls for further investigation in this area.

Fluorine and U are also present in superphosphate and thus may accumulate under fertilised soils (Taylor et al., 2016), however, due to the analytical methods required to measure F in environmental samples, this thesis does not investigate this TE. Uranium is not explored here as there is no evidence to suggest that this is a TE of interest to *E. guineensis* agriculture (Chapter 1).

2.8.1 Copper

Copper is an essential TE, necessary for photosynthesis as well as carbohydrate and protein metabolism in plants (Kabata-Pendias & Pendias, 2001). The mean range for Cu in soils is 13-24 mg kg^{-1}, and it is likely to accumulate in surface horizons from anthropogenic sources (Kabata-Pendias & Pendias, 2001). Copper is commonly used as a base for fungicides, and often accumulates in soils under horticultural land use due to this. It is present in various fungicide products used on a variety of economic crops grown in tropical regions including rubber (*Hevea brasiliensis* (Willd.ex A. Juss.) Mull. Arg.), cacao (*Theobroma cacao* L.), citrus (*Citrus spp.*), avocado (*Persea americana* Mill.), palm (*Elaeis spp.*) and rice (*Oryza spp.*) to control a range of plant pathogens (Copper Development Association, 2013). For this reason, it may accumulate in soils under *E. guineensis* plantations, either as a result of fungicide use on *E. guineensis* or from past land use activities. Furthermore, in areas where Cu-fungicides have been applied for long periods of time, microorganism resistance to Cu as well as heavy Cu accumulation in edible plant tissues has been observed (Kabata-Pendias & Pendias, 2001). For example, Cu concentrations of 20 mg kg^{-1} and 80 mg kg^{-1} have been found in tea leaves form Kenya and Japan respectively, while viticultural soils in France have been found to contain 100-1500 mg kg^{-1} Cu as a result of long-term historical use of a copper sulphate fungicide (Besnard et al., 1999; Wong et al., 1998).

2.8.1.1 Copper in Soils

Copper occurs in relatively similar concentrations throughout the soil profile and is largely immobile. Copper may form a range of ionic species, bonds and compounds in different soil conditions (Mengel et al., 2001). Both organic and inorganic exchange sites in soil may fixate Cu ions, through adsorption, organic complexation and chelating, microbial fixation or occlusion and coprecipitation (Kabata-Pendias & Pendias, 2001). Copper ion adsorption is a function of pH, as the surface charge of soil mineral adsorbents controls adsorption, which is dependent on pH (Mengel et al., 2001).

2.8.1.2 Plant uptake of copper

Copper absorption into plant roots may occur both actively and passively likely through dissociation, however, within plant roots, Cu most often occurs in a complexed state (Kumar et al., 2021). Like Cd, there is a clear relationship between Cu concentrations of soils and plants, however, the uptake rate of Cu is highly variant and dependent on ionic species (Kabata-Pendias & Gondek, 1978; Kabata-Pendias & Pendias, 2001). For example, anthropogenic CuO more readily undergoes plant uptake than lithogenic Cu to barley plants grown on contaminated sites (Grupe & Kuntze, 1988). Copper accumulates most commonly in plant roots, leaves and reproductive organs, and as in soil, Cu in plant tissues has low mobility. Copper deficiency in plants generally occurs at dry weight concentrations of 2-5 mg kg^{-1} (Appendix B), and is expressed by wilting, darkening of tissues, and disturbed pollen development (Kabata-Pendias & Pendias, 2001). Copper toxicity symptoms appear mainly in plant roots and leaves at concentrations of 20-100 mg kg^{-1}, as roots become shorter or thicker, and leaves darken before undergoing Fe chlorosis (Kabata-Pendias & Pendias, 2001).

2.8.2 Zinc

Zinc is an essential TE that commonly accumulates in topsoils from agricultural activities including fertiliser application as well as geogenic inputs. Worldwide, mean background Zn levels in soils are 64 mg kg^{-1} (Kabata-Pendias & Pendias, 2001). It is utilised by plants for processes of lipid, carbohydrate and nucleic acid metabolism, and both deficiencies and phytotoxic levels of Zn will cause interveinal chlorosis, especially in monocots (Brown et al., 1993; Kabata-Pendias & Pendias, 2001). In productive crops such as *E. guineensis,* this can effect a disturbance to the development of inflorescences, which may decrease yield (Broeshart et al., 1957). For this reason, Zn concentrations of soils and plant uptake from *E. guineensis* plantations are of interest when investigating TE fluxes in *E. guineensis* systems.

2.8.2.1 Zinc in soils

Zinc is readily soluble in soils, mobilising into Zn^{2+} as well as complexed ionic forms, specifically and non-specifically adsorbed cations (Alloway, 2008). In general, Zn solubility – particularly where adsorption and precipitation occur – negatively correlates with the presence of P compounds and Ca-saturation of soils (Kabata-Pendias & Pendias, 2001). In high pH soils however, Zn solubility may be affected by the presence of soluble complex anionic forms and Zn-organic complexes (Bloomfield, 1981). Where soils are rich in P and Ca, Zn plant deficiency may be an issue due to Zn immobilisation (Kabata-Pendias & Pendias, 2001). This most often occurs as a result of land use management. Clay and soil organic matter (OM) strongly bind Zn in soils with the clay fraction holding up to 60% of soil

Zn (Kabata-Pendias & Krakowiak, 1995). This generally gives higher total Zn concentrations to organic soil horizons, while mobile Zn^{2+} is readily formed in oxidising and acidic soil environments (Kabata-Pendias & Pendias, 2001). Following OM, hydrous Fe and Al oxides hold between 14-38% of total soil Zn, leaving organic complexes and mobile fractions with 1.5-2.3% and 1-20% respectively (Kabata-Pendias & Pendias, 2001; Zyrin et al., 1976).

2.8.2.2 Plant uptake of zinc

Plants take up soluble Zn forms from soil solution through processes of absorption, with varying rates that depend on plant species and soil conditions (Kabata-Pendias & Pendias, 2001). As with Cd and Cu, plant concentrations of Zn correlate with total soil-Zn (Diez & Rosopulo, 1976; Thoresby & Thornton, 1979). Zinc forms associated with Fe and Mn oxides in soils are typically more plant-available than other Zn forms in soils (Norrish, 1975). In soils high in Zn, this TE will accumulate in the roots of plants as well as in mature leaves and tops (Kabata-Pendias & Pendias, 2001). Zn concentrations in grain however, have not been observed to increase with higher Zn soil concentrations above 50 mg kg^{-1}. Kabata-Pendias and Pendias (2001) suggest this may be due to a biological barrier, restricting the transport of Zn to grain tissue. This may impact the Zn concentrations of *E. guineensis* endosperms, and products derived from this including palm kernel oil and PKE.

2.8.3 Cadmium

Cadmium is a non-essential TE, which has harmful effects on soil organisms, plants, animals and humans at high concentrations (Loganathan et al., 2012). The maximum tolerable level in animal feed is 10 mg kg^{-1} (National Research Council, 2005). Cadmium is bioaccumulative, residing primarily in the kidneys and liver of animals and humans, and is a known nephrotoxin and human carcinogen (Golia et al., 2008; World Health Organisation, 2010). Geogenic soil-Cd concentrations average 0.1-0.2 mg kg^{-1}, however, anthropogenic activities such as phosphate fertiliser application, mining, smelting (particularly Pb and Zn) and industrial power generation may lead to increased Cd accumulation in soils (Bešter et al., 2013). Cadmium may be present in soils, plant tissues and products of *E. guineensis* plantations, due to Cd's natural presence in phosphate rock and thus phosphate fertilisers that are applied to plantation soils (Loganathan et al., 2012; Woittiez et al., 2015). It is important to note that in areas where soils have developed in a humid climate (including tropical regions most suited to *E. guineensis* production), Cd is likely to move downwards through the soil profile rather than to accumulate in topsoils, and thus high topsoil-Cd concentrations in such regions are likely due to contamination rather than geogenic inputs (Kabata-Pendias & Pendias, 2001).

Conversely, in New Zealand pasturelands where soils have become contaminated with Cd through repeated application of Cd-rich phosphate fertilisers (Gray et al., 1999a), PKE concentrations of Cd may prove lower than pasture concentrations. The mean Cd concentration for New Zealand soils under dairying is 0.59 mg kg^{-1}, with some areas having concentrations of up to 2.14 mg kg^{-1} in regions with long histories of repeated phosphate fertiliser application such as Taranaki (Cavanagh, 2014). Here, PKE as a supplement stock-feed may be an economic solution to plant uptake of Cd and the subsequent risks to human health through food-chain transfer.

2.8.3.1 Risk to human health as a function of cadmium speciation in soils

Human health risk (that is, pollution) exists where exposure pathways make possible the transfer of contaminants from soil to humans – thus the presence of a contaminant above geogenic levels represents contamination and not necessarily risk (Chapman, 2007). Cadmium and other TEs enter animal and human food chains through plant uptake from topsoils, however, the incidence of plant uptake depends on several factors (Bešter et al., 2013). Most of the total Cd in soils (>98%) remains bound in soil colloids, with the remaining phytoavailable Cd in soil solution as free metal ions (Cd^{2+}) that are commonly quantified as exchangeable Cd (Al Mamun et al., 2016; Gray & Mclaren, 2006). In soil solution, Cd^{2+} may however, take the form of several complex ions including $CdOH^{+}$, $Cd(OH)^{3-}$, $Cd(OH)_4^{2-}$, $CdHCO_3^{+}$, $CdCl^{+}$, $CdCl_3^{-}$, $CdCl_4^{2-}$ as well as organic chelates (Kabata-Pendias & Pendias, 2001). On average, 20% of total soil-Cd exists as free Cd^{2+} ions in soil solution, however, this ranges from 0-60% across different soil types (Sébastien Sauvé et al., 2000). The fractions of this exchangeable Cd organically complexed, inorganically ion-paired, or present as divalent Cd^{2+} varies dependant on soil properties including soil solution pH: at pH of 4-7, the majority of Cd in soil solution is organically complexed, while at pH<4, the majority of Cd present is Cd^{2+} (Sébastien Sauvé et al., 2000). Plant-availability of Cd^{2+} is lowest at a soil solution pH of 6-7 (Simmler et al., 2013). Oxidation potential is a key factor influencing Cd^{2+} mobility and minerals CdO and $CdCO_3$ may form where there are strong oxidation conditions (Kabata-Pendias & Pendias, 2001).

2.8.3.2 Plant uptake of cadmium

Cadmium is readily absorbed by plants through both roots and leaves, from lithogenic and atmospheric sources. Transport of Cd through plant tissues can occur rapidly, likely as metallo-organic complexes (Kabata-Pendias & Pendias, 2001). This results in uneven accumulation of Cd within different plant tissues, with higher concentrations typically in the older leaves, followed by stem, grain, roots and fruit, and in lower concentrations in tubers (Cavanagh et al., 2018; Golia et al., 2008; Hu et al., 2013; Loganathan et al., 2003). When plants are grown on contaminated soils however, higher concentrations of Cd may also be found in tubers and roots (Cunningham et al., 1975). This may occur as a result of Cd being held in active exchange sites in cell-walls, restricting transport to stems, leaves and fruit (Kabata-Pendias & Pendias, 2001).

Several key factors influence the rate of Cd uptake to plants. Multiple studies have identified total soil-Cd concentrations as a key influencer of plant-Cd concentrations (Alloway et al., 1990; Bešter et al., 2013; Gray & Mclaren, 2006; Gray et al., 1999b; Hornburg & Brümmer, 1986; McBride, 2002; McBride et al., 1997; S. Sauvé et al., 2000; Sillanpää & Jansson, 1992; Zupan, 2003). Activities that increase soil-Cd concentrations, such as phosphate fertiliser application, are therefore likely to lead to increases in plant-Cd concentrations.

Soil pH also has a strong influence on Cd uptake: as soil acidity increases, plant uptake of Cd typically increases (Kabata-Pendias & Mukherjee, 2007). Cadmium, like Zn, does not require an environment of high acidity to undergo dissolution like metals such as Co and Ni, but will ionise in weakly acidic environments (Loganathan et al., 2003). In acidic soils, less negative bonding sites exist thus Cd ions are more likely to undergo plant uptake through plant root surfaces, while in alkaline soils, Ca ions compete directly with Cd ions for uptake sites, reducing Cd uptake (Adriano, 1986; Evans, 1989). $CdOH^{+}$ species more readily undergo plant uptake than Cd^{2+} and form in soils with pH>8, while $Cd(OH)_2$ precipitates when soil pH>10 (Bolan et al., 2003; Naidu et al., 1994). Plants grown in saline soils have been shown to take up greater concentrations of Cd, due to strong complexation with Cl^- forming phytoavailable $CdCl^+$, $CdCl_2$, $CdCl_3^-$ and $CdCl_4^{2-}$ (McLaughlin et al., 1994).

2.9 References

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Chapter 3: Chemical elements in selected soils of current and former palm oil production systems

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Abstract

Palm oil production in Southeast Asia often occurs on nutrient-poor, acidic soils converted from primary forest. Both the agricultural conversion and the production of oil palm (*Elaeis guineensis* Jacq.) are subsidised in Indonesia. As well as depleting soil organic C and plant nutrients, agricultural production on these soils can result in the accumulation of trace elements (TEs) from the use of TE-containing agrichemicals including phosphate fertilisers and Cu-fungicides. We aimed to test the hypothesis that *E. guineensis* soils will have lower C concentrations than forest soils, as well as accumulation of TEs including Cu, Zn and Cd. Soil samples from active and abandoned *E. guineensis* plantations in Sumatra, Indonesia were analysed for general soil properties as well as TE concentrations. Soils were acidic and nutrient-poor, with production likely to be limited by deficiencies of N, P, K, Mg and Mo, present in concentrations as low as 0.021%, 118 mg kg^{-1}, 778 mg kg^{-1}, 1023 mg kg^{-1} and 0.095 mg kg^{-1}, respectively. Mean organic C was lower (2.0-3.3%) than in nearby forest soils (7.7%). Soils under palm production contained elevated levels of Cu, Zn, As and Pb up to 38, 91, 9.0 and 28 mg kg^{-1} respectively, likely due to agrichemical use. Correction of nutrient deficiencies in *E. guineensis* production would require significant fertiliser inputs, which would exacerbate TE accumulation and reduce the net economic revenue from oil production. Future work should determine the economic viability of *E. guineensis* production on soils converted from primary forest with adequate capital fertiliser application and should adequately consider the resulting TE-loading to soils.

Keywords

Elaeis guineensis, palm oil, fertilisers, trace elements, contaminants

3.1 Introduction

Since 1990, most tropical soils used for agricultural production (90%) were formerly under indigenous forest (Gibbs et al., 2010), of which 86% were converted to oil palm (*Elaeis guineensis* Jacq.) and rubber (*Hevea brasiliensis* (Willd. ex A.Juss.) Müll.Arg.) plantations (Vancutsem et al., 2021). Indonesia and Malaysia are the largest palm oil producers (Corley & Tinker, 2015). Indonesia provides financial incentives to convert forest to *E. guineensis* plantations and subsidises *E. guineensis* production (Kissinger, 2016). There has been criticism of the destruction of forest ecosystems for agriculture in Southeast Asia, particularly due to the threats to megafauna habitat and biodiversity (Bateman et al., 2010; Fitzherbert et al., 2008; Morgans et al., 2018; Wilcove & Koh, 2010). However, little attention has been given to the effects of *E. guineensis* production on tropical soils (Padfield et al., 2019).

Typically, indigenous forests overlie acidic soils with low concentrations of plant nutrients that require capital lime and fertilisers to enable production (Foy, 1984; Hartemink, 2002). Following conversion, the soil organic matter usually decreases (Woomer et al., 1994). Both the fertilisers and composts that are used to increase soil fertility as well as applied pesticides may contain trace elements (TEs) as contaminants, such as Zn, As, Cd and Pb in NPK fertilisers (Taylor et al., 2016), or active ingredients such as Cu in Cu-based fungicides (Bivi et al., 2016). Trace elements may accumulate in soils over time (Chaney, 1989) and increase the risk of food-chain transfer of toxic elements, particularly if Soil Guideline Values (SGVs) for TEs are exceeded (Chaney, 1989). In different agricultural systems requiring high input rates of TE-containing agrichemicals, the accumulation of TEs in soil has posed risks for production: in New Zealand dairy farms that have historically used high rates of phosphate fertilisers, Cd now limits production in some regions (Gray & Cavanagh, 2022), while in some regions of France, viticultural production is limited by the accumulation of Cu due to repeated Cu-pesticide use (Besnard et al., 1999). If similar inputs and accumulation has occurred under *E. guineensis* agriculture, TEs may come to limit production in *E. guineensis* producing soils.

Elaeis guineensis is an intensive crop, requiring maintenance nutrient inputs of N (215–286 kg ha^{-1}), P (143–215 kg ha^{-1}), K (358–429 kg ha^{-1}), Mg (215–286 kg ha^{-1}) and B (14–29 kg ha^{-1}) (Woittiez et al., 2015). These input requirements may increase when *E. guineensis* is cultivated on nutrient-poor soils (Corley & Tinker, 2015; Woittiez et al., 2015). Cultivation on Histosols also requires inputs of Cu and Zn to meet plant nutrient requirements (Ng, 2002), and the use of Cu-based fungicides is recommended to treat *E. guineensis* pests including *Ganoderma* spp. (Bivi et al., 2016; MPOB, 2015). In a survey of farmers, the most frequently used fertilisers in smallholder *E. guineensis* production in Indonesia were subsidised NPK (15-15-15) and urea, with farmers citing financial limitations as a key factor in fertiliser choice (Woittiez et al., 2018). *Elaeis guineensis* soils containing concentrations of Cu, Zn and Cd up to 69, 107 and 5.2 mg kg^{-1} respectively have been reported (Thompson-Morrison et al., 2022), while soil organic matter has been found to decrease by up to 42% when converted from forest to *E. guineensis* (Rahman et al., 2018). Maximum potential yields for palm oil have been calculated at between 10-18 t oil ha^{-1} yr^{-1}, however, commercial yields are in the range of 5-7 t oil ha^{-1} yr^{-1} (Wahid et al., 2005), and smallholder yields as low as 4 t oil ha^{-1} yr^{-1} (Woittiez et al., 2017). Without subsidies, *E. guineensis* production on low-fertility tropical soils may not be viable (Afriyanti et al., 2019; Pradipta, 2018; Sari et al., 2019).

We hypothesise that conversion of forest soils to *E. guineensis* or *H. brasiliensis* plantations will result in the accumulation of TEs, particularly Cu, Zn and Cd as well as the depletion of organic C. We aim to determine the chemical composition and plant nutrient status of soils under active production and those that have been abandoned in Indonesia, and compare these with published data from forest soils, international SGVs, *E. guineensis* production systems in other regions, as well as soils under *H. brasiliensis* production. We further hypothesise that soils under current production will have higher N concentrations relative to sites at which production has been abandoned. This research investigates whether there is evidence of accumulation of TEs in *E. guineensis* and *H. brasiliensis* production soils compared to literature values of tropical rainforest soils and compares soil organic matter contents with published data on with forest soils. This will be discussed in the context of land use history to gain a perspective on the potential effects of *E. guineensis* agriculture on tropical soils.

3.2 Methods

3.2.1 Site descriptions

Soil samples were collected from four *E. guineensis* plantation sites on the Indonesian island of Sumatra in May 2019 (Figure 3.1). Samples were obtained under the conditions of anonymity of land ownership and following this, Figure 3.1 identifies the locality where samples were collected from. The sampling region has mean annual rainfall of 2950 mm (Rochiyati et al., 2010). All *E. guineensis* sites had soils classed as Acrisols under the FAO soil classification system (FAO & UNESCO, 1976), representing some 90% of soils in *E. guineensis* growing regions of Indonesia. Acrisols are taxonomically related to Oxisols under the USDA soil classification system (Encyclopedia Britannica, 2016). These soils typically form under humid, tropical climates and old undulating landscapes, and are characterised by a subsurface accumulation of kaolinitic clays. They are typically highly leached, low in plant nutrients, highly erodible and contain high levels of Al. As such, agricultural production on these soils can be challenging. The sites were chosen to give an indication of the range of soil conditions present under both current and former *E. guineensis* plantations with varying management practices and landscape features.

Sites A and B were both abandoned *E. guineensis* plantations on the forest-edge of Gunung Leuser National Park, which were formerly under corporate management. For the previous three years, the sites had been unmanaged. These sites were under production for ca. 15 years. These sites were adjacent to each other and were separated northeast (Site A)/southwest (Site B). Future reforestation was planned at these sites. As such, the sites were over-grown with understory plants and no amendments had been applied to the soil for at least three years. These sites were both hilly and steep in places with slopes up to approximately 12°, and samples were taken from both in areas in which *E. guineensis* trees remained, based on accessibility and practicality.

Site C was an active smallholder *E. guineensis* plantation site on the forest-edge, near to Gunung Leuser National Park. This site was adjacent to a small village and river and had been under *E. guineensis* production for ca. 20 years. The site had a slope of <4°.

Site D was an ex-*E. guineensis* plantation that also bordered Gunung Leuser National Park. Few *E. guineensis* trees remained, as production at the site had been abandoned and the area had transitioned into a mix of permaculture and reforestation. No amendments had been applied to the soil for at least three years. This site had likely been under production for ca. 20 years. This site was hilly with slopes up to 9° and samples were collected here based on accessibility and practicality.

Control samples outside of plantations and in natural forest could not be collected, as permits were not granted to take soil samples from within the National Park. As such, we used published data from soils under nearby Gunung Leuser National Park (Asnur et al., 2019; Wasis, 2012) as well as from a *H. brasiliensis* (Willd. ex A.Juss.) Müll.Arg. (rubber) plantation (Site E) in North Sumatra as a comparison site. *Hevea brasiliensis* cultivation often occurs adjacent to *E. guineensis* cultivation, however, typically uses lower inputs of agrichemicals, particularly fertilisers (Schwarze et al., 2015). The *H. brasiliensis* trees at this site were ca. 10 years old at the time of sampling. The soils from this site were Ultisols under the USDA classification system. Ultisols are similar to Acrisols/Oxisols, but feature a translocated clay layer below the surface horizon (*Ultisol*, 2012).

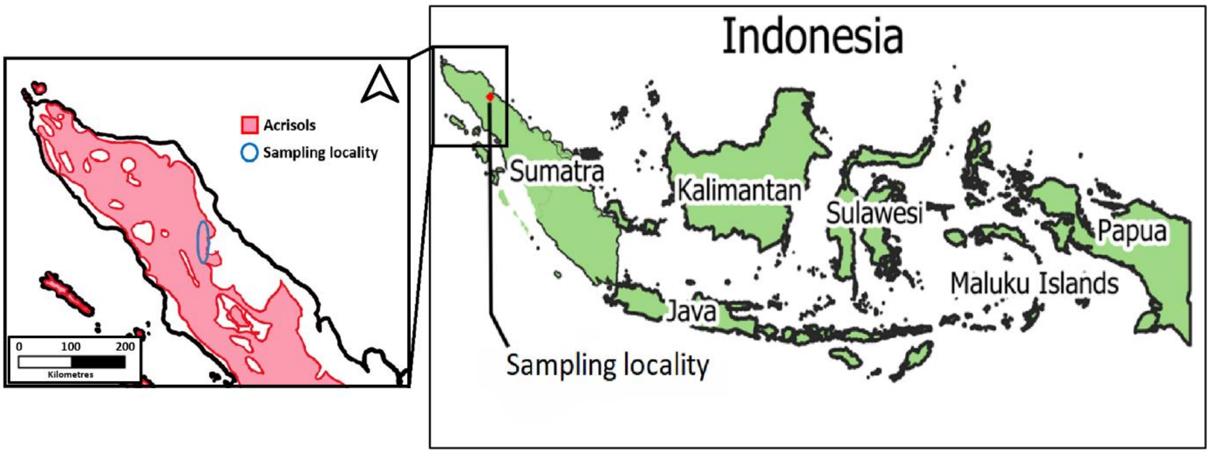

Figure 3.1 Map of Indonesia showing sampling locality in Sumatra with the extent of Acrisol soils over the north of the island. Adapted from FAO & UNESCO (1976)

3.2.2 Analytical methods

3.2.2.1 Sample collection and preparation

After clearing away the surface little, soil samples were collected using a spade to 15 cm depth and placed in zip lock plastic bags. Samples were taken at least 20 m apart, from inter rows between trees. As each site varied in area, more samples were taken from larger sites to achieve representativity of soils at each site. We took 25 samples from Sites A, B and D, and ten samples from Site C as this site was smaller in area. Samples were oven dried at 105°C until a constant weight was achieved. Samples were then crushed using a mortar and pestle and passed through a 2 mm stainless steel sieve for all analyses excluding PSA.

3.2.2.2 Pseudo-total element concentrations

Microwave digestion was used to analyse pseudo-total elemental concentrations (hereafter referred to as total concentrations) in all samples following EPA Method 3051A (US EPA, 2007). The microwave used was an Ultrawave Single Reaction Chamber Microwave Digestion Unit. For each sample, 0.20 g (±0.01 g) was weighed into a digestion tube and 5 mL 70% HNO_3 added. Samples were left to pre-digest overnight, before being digested for 25 minutes at 230°C (Milestone, n.d.). Two reagent blanks and two standard reference material (SRM) samples (NIST, 2003, 2018) were included with every 11 samples for quality assurance.

3.2.2.3 $Ca(NO_3)_2$-extractable element concentrations

A $Ca(NO_3)_2$ extraction was used to measure phytoavailable concentrations of TEs in *E. guineensis* soil samples. This analysis used 0.05 M $Ca(NO_3)_2$ (Gray et al., 1999). Here, 5.00 g (±0.05 g) of soil was weighed into a 50 mL centrifuge tube, and 30 mL of 0.05 M $Ca(NO_3)_2$ added. Tubes were placed on an oscillating shaker for two hours, then centrifuged at 4559 g for ten minutes. Four reagent blanks were included in every analysis.

Soil adsorption coefficients (K_D) were calculated for TEs soils at Sites A, B, C and D based on total and $Ca(NO_3)_2$ extractable (soluble) concentrations using the sorbed/soluble concentration quotient.

3.2.2.4 pH

The pH of soil was measured using a pH electrode according to the pH in H_2O method (Blakemore et al., 1987). Five g of soil was measured into a beaker, and 12.5 mL of Milli-Q water added (soil water ratio of 1:2.5). Samples were then stirred vigorously for at least 1 minute and left to stand overnight. A Hach HQ440d multi pH probe was then used to take the pH measurement of each sample.

3.2.2.5 Cation exchange capacity and base saturation

The method described by Hendershot and Duquette Hendershot and Duquette (1986) was used to analyse samples for cation exchange capacity (CEC). For each sample, 2.00 g (±0.01 g) of soil was weighed into a 50 mL centrifuge tube and 20.00 mL 0.1 M $BaCl_2{:}2H_2O$ added. Samples were shaken on an oscillating shaker for 2 hours, before being centrifuged at 4559 g for 10 mins. Following this, samples were analysed by microwave plasma-atomic emission spectroscopy (MP-AES) for Na, Mg, Al, K, Ca, Mn and Fe. Effective CEC was calculated from this. Base saturation (BS) was calculated as the percentage of CEC made up of base cations, i.e. Na, Mg, K and Ca. At least two reagent blanks were included in every analysis.

3.2.2.6 Total carbon and nitrogen

Total C and N of soils was analysed on a CN828 Elemental Analysis by Combustion C and N analyser. Blanks and SRMs (LECO, 2021) were analysed with samples.

3.2.2.7 Particle size analysis

A laser diffraction method was used to analyse soils for particle size distribution. The method used for PSA was developed in collaboration by University of Canterbury sedimentology and engineering departments, and follows the overview given by Bohren and Huffman (1998) and Pansu and Gautheyrou (2006). Samples were deflocculated in 50 g L^{-1} sodium hexametaphosphate (Calgon) on an oscillating shaker for 24 hours. They were then vortexed and placed on a magnetic stirrer plate to ensure all particles were in suspension. The sample was then analysed using a Saturn DigiSizer 5200, using the Fraunhofer method (Bohren & Huffman, 1998). Drops of the deflocculated sample were dropped into the liquid sample handling unit of the machine until the optimum obscuration was achieved (12-17%), and sample analysis run. A Wentworth size chart was used to determine silt, sand and clay soil fractions, and soil categorised according to USDA classification (USDA, 1987).

3.2.2.8 Elemental determination

Macroelements (P, K, S, Ca, Mg, Al and Fe) were analysed by inductively coupled plasma-mass spectrometry (ICP-MS) and MP-AES. Recoveries of SRMs ranged from 30-169% of certified values.

Appendix C gives a complete list of the elements and their respective recoveries. Trace elements (Cr, Mn, Co, Ni, Cu, Zn, Ge, As, Sr, Mo, Pd, Ag, Cd, Te, Cs, Ce, Re, Ir, Hg and Pb) were analysed by ICP-MS, inductively coupled plasma-optical emission spectroscopy (ICP-OES) or MP-AES. Recoveries ranged from 37-164% of certified values. The limit of quantification for analyses was 0.01 mg kg^{-1}.

3.2.3 Statistical analysis

R (version 4.1.0) (R Core Team, 2021) was used for all statistical analyses. One-way parametric Analysis of Variance (ANOVA) was done to compare means between sites using the package multcomp (Hothorn et al., 2008). Where assumptions of normality were not met, data was log-transformed (natural log). When data did not meet assumptions of homoscedacity, a non-parametric permutation ANOVA was used, however, no difference was found between parametric and non-parametric tests with regard to significance of results, and the parametric ANOVA was concluded to be robust. Tukey's Honest Significant Difference test was used to determine significant differences in soil variables between sites.

A principal component analysis (PCA) was used to determine which soil factors were the most important variants between sites and whether agrichemical-borne elements were important. Only variables that could be measured across all sites were included, so the PCA did not include $Ca(NO_3)_2$ extractable element concentrations, CEC, BS or particle size data. The PCA was run for total elemental concentrations and soil variables in R using the package factoextra (Kassambara & Mundt, 2020). Incomplete sets of elemental data were removed prior to running the PCA.

Log-transformed data from each site was used to create correlation matrices and correlograms using the R packages Hmisc (Harrell, 2021) and corrplot (Wei & Simko, 2021). Any soil variables with missing or below detection values were excluded from this analysis. A probability value of 0.01 (chosen over 0.05) was used to determine statistically significant correlations in this analysis, as there were multiple comparisons made and thus a higher chance of incorrectly identifying significant relationships between variables (Type 1 error). Significance codes were used to identify statistically significant correlations: S* indicates $p < 0.01$, while S** indicates $p < 0.001$.

3.3 Results and discussion

3.3.1 General soil properties and soil carbon

Soil samples from all sites were acidic, with pH ranging from 4.6-5.4 (Table 3.1). This is within the range of 4.5-5.5 and comparable to the mean of 5.1 previously reported in *E. guineensis* plantations in the Gunung Leuser National Park (Asnur et al., 2019; Wasis, 2012). Site E (pH 5.2) was within the preferred pH range for *H. brasiliensis* of 5.0-6.0 (Asnur et al., 2019). Under acidic soil conditions, Al becomes increasingly soluble as Al^{3+}: this is supported by a strong negative correlation between $Ca(NO_3)_2$ extractable Al and pH ($r = -0.86$ S**) for all soils. In kaolinitic soils at pH<5.5, Al^{3+} and Mn^{2+} toxicity may inhibit plant growth, making agricultural production challenging (Foy, 1984; Rao et al., 1993).

There were significant differences in soil texture between sites: Soils at Site C were silt loams while soils at Site D ranged from silt loam through to sandy loam (Figure 3.2). Soils at Sites A and B contained significantly more clay than other sites. The sand, silt and clay fractions of soils ranged from 15-40%.

Table 3.1. General properties of soils at each site (standard error of the mean in parentheses). Sites that share the same letter(s) for a single variable are not significantly different from each other.

Site		**A (n=25)**	**B (n=25)**	**C (n=10)**	**D (n=25)**	**E (n=32)**
Particle size distribution	**Sand (%)**	8.3(1.4) ^{a}	5.6(1.0) ^{a}	27(1.3) ^{b}	50(2.5) ^{b}	—
	Silt (%)	58(1.4) ^{b}	55(0.99) ^{b}	57(1.3) ^{b}	35(1.8) ^{a}	—
	Clay (%)	33(2.2) ^{b}	40(1.7) ^{b}	17(1.3) ^{a}	15(0.92) ^{a}	—
pH		5.4(0.11) ^{c}	4.8(0.061) ^{ab}	5.1(0.097) ^{bc}	4.6(0.047) ^{a}	5.2(0.076) ^{c}
CEC (cmol (+) kg^{-1})		25(1.5) ^{c}	30(0.80) ^{d}	4.3(0.63) ^{a}	10(0.67) ^{b}	—
BS (%)		70(5.5) ^{c}	33(4.7) ^{b}	43(8.5) ^{bc}	21(3.4) ^{a}	—
Al	**Total (mg kg^{-1})**	29,207(868) ^{b}	31,022(1144) ^{b}	32,307(1241) ^{b}	25,174(1855) ^{a}	67,371(2379) ^{c}
	$Ca(NO_3)_2$ extractable (mg kg^{-1})	110(24) ^{ab}	308(30) ^{c}	36(7.1) ^{a}	194(16) ^{b}	—
Fe	**Total (mg kg^{-1})**	28,530(696) ^{c}	31,149(1017) ^{c}	23,084(420) ^{b}	16,847(1007) ^{a}	23,921(334) ^{b}
	$Ca(NO_3)_2$ extractable (mg kg^{-1})	0.42(0.095) ^{a}	0.64(0.18) ^{a}	0.045(0.015) ^{a}	4.2(0.95) b	—

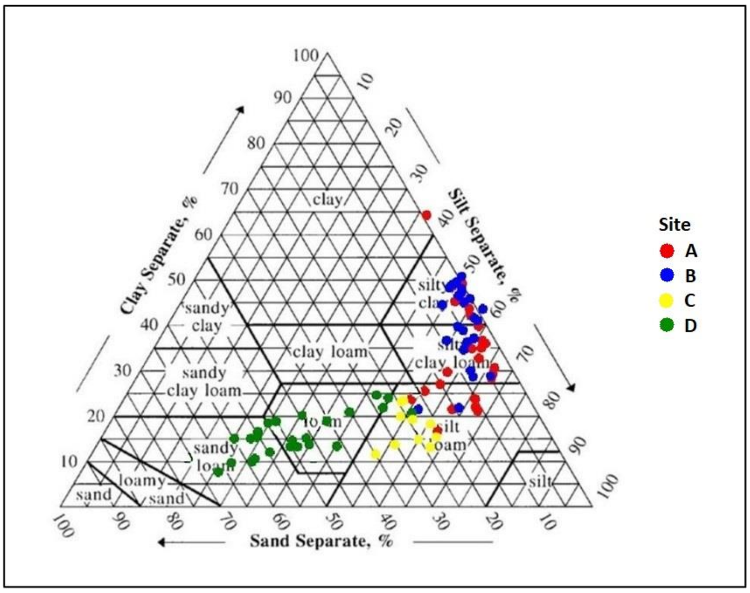

Figure 3.2 Soil texture classes of soil samples from *E. guineensis* sites, according to the USDA classification.

The C concentrations in our soils ranged from 1.3-3.3% with the site under *H. brasiliensis* having significantly less C than the *E. guineensis* sites. The C concentrations for soils of the four *E. guineensis* sites presented in Table 3.2 were comparable to the mean C concentration of 2.1% reported for tropical Oxisols (Shofiyati et al., 2010). The significantly lower C concentration at Site E may be a result of soil type, as tropical Ultisols have lower mean C (1.9%) (Shofiyati et al., 2010). The C concentrations at Sites A–D were similar to C concentrations of Indonesian *E. guineensis* soils in the Gunung Leuser National Park area: Asnur et al. (2019) reported the majority of soils in a *E. guineensis* production area within Leuser were within the range of 1-2%, while Wasis (2012) reported a mean organic C concentration of 0.57%, decreased from 7.7% following conversion from forest to *E. guineensis*. This loss in organic C is typical of soils following conversion to agriculture (Sanderman et al., 2017), particularly in tropical regions and in specifically in Sumatra (van Noordwijk et al., 1997). However, the optimal soil C range for *E. guineensis* production is 1.5-2.0% according to Corley and Tinker (2015), and 1.2-1.5% according to Goh and Po (2005). In this case, any reduction in soil C following agricultural conversion may be beneficial for *E. guineensis* production, while having unfavourable implications for a changing climate (Sanderman et al., 2017).

Cation exchange capacity was significantly different between all four *E. guineensis* sites, in the order Site C < Site D < Site A < Site B. The CEC of the Site D site (10 cmol (+) kg^{-1}) was comparable to published *E. guineensis* soil data of 8.2 cmol (+) kg^{-1} (Wasis, 2012), while Site A and Site B (25 and 30 cmol (+) kg^{-1}, respectively) were closer to published CEC data for natural forest (26 cmol (+) kg^{-1}). All values were higher than those reported from an *E. guineensis* plantation in Western Ghana of 3.0 and 2.3 cmol (+) kg^{-1} (Obeng et al., 2020). There is disagreement in the literature regarding the optimal soil CEC for *E. guineensis* production. Recommended ranges vary from 12-15 cmol (+) kg^{-1} (Goh & Po, 2005; Obeng et al., 2020) to >24 cmol (+) kg^{-1} (Corley & Tinker, 2015).

Both the clay content and the C concentration of soil samples increased in the order Site D < Site C < Site A < Site B (Table 3.2 and Figure 3.2). There was a positive correlation between the specific surface area of soils and C, which is consistent with clay minerals protecting organic matter from degradation (Kahle et al., 2002; Matus et al., 2014), effectively affording soils with a higher clay content a higher C-holding capacity (Feng et al., 2013). Across all soils, there were positive correlations between C and clay content ($r = 0.43$ *S***), C and CEC ($r = 0.51$ S**), and clay content and CEC ($r = 0.68$ S**).

3.3.2 Plant macronutrients

The average soil N ranged from 0.021% (Site D) to 0.23% (Sites A and B) (Table 3.2). The optimal soil N concentrations for *E. guineensis* production range from 0.12% (Goh & Po, 2005) to >0.15% (Woittiez et al., 2015). The active site (Site C) did not contain higher N concentrations relative to the ex-*E. guineensis* site (Site D) and contained significantly lower N than the abandoned sites A and B. This falsified our hypothesis that the active site would contain higher N concentrations than the abandoned sites. Soils at both Sites C and D were deficient in N, which would have contributed to reduced yields of palm oil. This is supported by the high C:N ratios at these sites of 60 and 95 respectively, which indicate that N may undergo rapid immobilisation in these soils [1] (Bengtsson et al., 2003). In comparison, *E. guineensis* plantation soils in Nigeria had C:N ratios of 5 (Aweto & Enaruvbe, 2010). Nitrogen deficiency has been reported in smallholder plantations in Indonesia similar to Site C and attributed to sub-optimal fertiliser applications (Woittiez et al., 2015). It is likely that the soils of the Site C and Site D sites may have experienced N depletion following a change to *E. guineensis* cultivation, and subsequent inappropriate nutrient management. Wasis (2012) reported a decrease in soil N from 0.32% to 0.06% following agricultural conversion in the Leuser region.

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[1] Mineral N (NH_4^+ and NO_3^-) could not be measured due to the inability to import fresh soil into our laboratory.

Table 3.2. Mean soil characteristics at each site (standard error of the mean in parentheses). Sites that share the same letter(s) for a single variable are not significantly different from each other.

Site		**A (n=25)**	**B (n=25)**	**C (n=10)**	**D (n=25)**	**E (n=32)**
Total C (%)		3.2(0.23) ^{c}	3.3(0.23) ^{c}	2.1(0.11) ^{b}	2.0(0.14) ^{b}	1.3(0.027) ^{a}
Total N (%)		0.23(0.023) ^{c}	0.23(0.020) ^{c}	0.035(0.0090) ^{a}	0.021(0.0057) ^{a}	0.12(0.0032) ^{b}
C:N ratio		14	14	60	95	11
P	**Total (mg kg^{-1})**	296(21) ^{b}	305(17) ^{b}	299(13) ^{b}	118(30) ^{a}	323(14) ^{b}
	Ca(NO$_3$)$_2$ extractable (mg kg^{-1})	0.030(0.0040) ^{b}	0.021(0.0029) ^{ab}	0.037(0.0031) ^{b}	0.013(0.0033) ^{a}	—
K	**Total (mg kg^{-1})**	4252(303)^{d}	2316(106)^{c}	1883(176)^{bc}	778(67)^{a}	1612(25)^{b}
S	**Total (mg kg^{-1})**	266(16) ^{c}	358(139) ^{bc}	170(9.5) ^{b}	116(7.6) ^{a}	306(6.5) ^{c}
	Ca(NO$_3$)$_2$ extractable (mg kg^{-1})	2.6(1.3) ^{a}	1.8(0.85) ^{a}	0.45(0.39) ^{a}	1.1(0.42) ^{a}	—
Ca	**Total (mg kg^{-1})**	9422(1400) ^{d}	5468(996) ^{cd}	2439(564) ^{bc}	1286(416) ^{a}	1202(117) ^{ab}
	Ca(NO$_3$)$_2$ extractable (mg kg^{-1})	65(5.6) ^{c}	6.6(2.4) ^{b}	0.0001(<0.001) ^{ab}	1.9(1.9) ^{a}	—
Mg	**Total (mg kg^{-1})**	3005(156)^{c}	2637(101)^{c}	2747(405)^{c}	1023(102)^{a}	1185(49)^{b}

Average total P in soils ranged from 118 mg kg^{-1} (Site D) to 323 mg kg^{-1} (Site E) (Table 3.2). The range of mean P concentrations in all soils was low compared to the world average of 650 mg kg^{-1} (Mason & Moore, 1982) and range of 200-1500 mg kg^{-1} (McLaren & Cameron, 1996). Deficiencies in soil P exist in smallholder plantations in Indonesia (Woittiez et al., 2015), and P deficiency is common in crops grown on weathered acidic soils including Acrisols/Oxisols and Ultisols (Fageria & Nascente, 2014). These soils typically have capacity for high P fixation, resulting in reduced plant uptake of P. Fertilisers are usually scattered on the soil surface in *E. guineensis* plantations around the base of palms. This, combined with management practices such as weed-clearing and growing on landscapes with >6° slope has high potential to result in soil and fertiliser run-off during rainfall events and may limit production (Corley & Tinker, 2015; Foster & Tayeb, 1986; Goh et al., 2003; Maene et al., 1979; Wasis, 2012; Woittiez et al., 2018). As Sumatra experiences ca. 3000 mm of rainfall annually (*Climate: North Sumatra*, n.d.), high erosion rates are common in this region (Aflizar et al., 2018). Soil erosion in *E. guineensis* plantations removes topsoil as well as soil P and may lead to eutrophication of near-by surface water. The low P concentrations in the *E. guineensis* soils may be limiting yields at these sites.

Potassium and Mg at all sites were below global average soil concentrations of 14,000 and 5000 mg kg^{-1} respectively (Mason & Moore, 1982). Low K and Mg concentrations are typical of weathered, acidic soils with low-K parent materials and high K and Mg-leaching, due to high precipitation (Fageria & Nascente, 2014). Woittiez et al. (2015) reported K deficiencies in >80% of smallholder plantation soils surveyed in Indonesia, as well as visual signs of Mg deficiency. Globally, K outputs from soil exceed K inputs, and this is especially pronounced in Global South countries where access to K-fertilisers is more limited than in the Global North (Manning, 2015). Woittiez et al. (2015) identified K deficiency as a potential leading cause of sub-optimal yields in smallholder *E. guineensis* plantations and attributed this to the use of subsidised NPK fertilisers with poor nutrient balances for *E. guineensis* requirements. It is possible that a lack of access to K fertilisers adequate for *E. guineensis* and *H. brasiliensis* production is responsible for the low K concentrations in soils from the *E. guineensis* and rubber sites.

3.3.2 Total trace element concentrations

Except for elevated concentrations of Zn and Pb, TE concentrations (Table 3.3) in *E. guineensis* soils were generally similar to background concentrations. No concentrations of TEs in *E. guineensis* soils exceeded SGVs for agricultural soils.

Table 3.3. Mean TE concentrations of soils of each site (standard error in parentheses). Mean concentrations in mg kg^{-1}. Sites that share the same letter(s) for a single variable are not significantly different from each other.

Site	**A (n=25)**	**B (n=25)**	**C (n=10)**	**D (n=25)**	**E (n=32)**
Cr	34(1.1) ^{c}	25(0.90) ^{b}	27(1.4) ^{bc}	50(3.7) ^{d}	5.03(0.20) ^{a}
Mn	281(56) ^{b}	519(87) ^{c}	876(82) ^{d}	28(3.7) ^{a}	2995(52) ^{e}
Co	19(2.3) ^{c}	19(1.5) ^{c}	16(0.36) ^{c}	3.4(0.42) ^{a}	8.9(0.12) ^{b}
Ni	29(2.0) ^{d}	30(1.5) ^{d}	13(0.61) ^{c}	7.9(0.58) ^{b}	4.5(0.40) ^{a}
Cu	19(1.4) ^{c}	38(0.86) ^{d}	5.6(0.69) ^{b}	1.1(0.22) ^{a}	6.6(0.49) ^{b}
Zn	91(3.7) ^{b}	91(3.8) ^{b}	75(5.2) ^{b}	19(1.4) ^{a}	213(5.7) ^{c}
Ge	0.86(0.051) ^{b}	1.0(0.79) ^{b}	1.05(0.36) ^{b}	0.54(0.028) ^{a}	1.7(0.034) ^{c}
As	3.8(0.14) ^{bc}	4.3(0.16) ^{c}	9.0(0.59) ^{d}	3.7(0.28) ^{ab}	3.1(0.12) ^{a}
Sr	24(2.2) ^{c}	14(1.4) ^{b}	48(13) ^{d}	6.1(0.67) ^{a}	4.3(0.16) ^{a}
Mo	1.5(0.17) ^{b}	2.0(0.15) ^{c}	0.13(0.0068) ^{a}	0.30(0.080) ^{a}	0.095(0.0038) ^{a}
Cd	0.14(0.026) ^{bc}	0.087(0.024) ^{ac}	0.085(0.029) ^{ab}	0.026(0.0073) ^{a}	0.16(0.012) ^{b}
Ce	22(1.9) ^{a}	23(2.5) ^{a}	55(1.6) ^{b}	17(0.83) ^{a}	139(1.9) ^{c}
Pb	16(0.70) ^{b}	17(1.2) ^{b}	28(1.8) ^{c}	5.8(1.2) ^{a}	51(0.53) ^{d}

Sites A, B and C all contained soil concentrations of Zn and Pb above background levels of 50 and 10 mg kg^{-1} respectively (Mason & Moore, 1982). Although elevated, concentrations of Zn and Pb did not surpass SGVs. However, Zn concentrations were present at these three sites at >50% of the limits (a benchmark used in environmental monitoring) (Nokes, 2008) for Denmark and the EU, indicating that accumulation should be monitored at the remaining active site (Site C) and further Zn inputs to soils limited to prevent accumulation that could impede production. Our concentrations were comparable to published values of 99 mg kg^{-1} Zn in Malaysian *E. guineensis* soils (Aini Azura et al., 2012) and 14 mg kg^{-1} Pb in Nigerian *E. guineensis* plantation soils (Olafisoye et al., 2016). Soils from Site C also contained As above the background level of 5 mg kg^{-1} (Hooda, 2010), however, below SGVs for the United States (US) (18 mg kg^{-1}), Canada (17 mg kg^{-1}) and New Zealand (20 mg kg^{-1}) (Cavanagh & Munir, 2016). Again, as As in Site C soil was present at >50% SGVs, inputs should be monitored here. No reports could be identified for comparison of As in *E. guineensis* plantation soils. The natural background concentration of Cu in soil is 20 mg kg^{-1} (Mason & Moore, 1982), which was surpassed in soils at Site B. The concentrations here breached 50% of SGV levels for Canada (63 mg kg^{-1}) and the US (70 mg kg^{-1}) (Cavanagh & Munir, 2016). Copper was reported up to 69 mg kg^{-1} in Nigerian *E. guineensis* plantations (Olafisoye et al., 2016). The elevated Cu, Zn, As and Pb were likely a result of agrichemical use during *E. guineensis* production at these sites. Zinc, As and Pb are all common contaminants of phosphate fertilisers (Taylor et al., 2016), while Cu is a common active ingredient in pesticides used in horticultural production (Kiwifruit Vine Health, 2019), and in particular the production of *E. guineensis* (Bivi et al., 2016; MPOB, 2015). Site B may have had higher rates of fungicide application leading to its significantly higher soil Cu concentrations.

Deficiencies in Mo, defined as <2 mg kg^{-1} (Hooda, 2010), were present at all sites except Site B. Our total Mo concentrations were comparable to those of Golow et al. (2010) from *E. guineensis* plantations in Ghana. Several TEs measured in our soils were present at concentrations less than or comparable to natural background levels in soils. These included Cr (5.03-50 mg kg^{-1}), Co (3.4-19 mg kg^{-1}), Ni (4.5-30 mg kg^{-1}), Ge (0.54-1.7 mg kg^{-1}) and Cd (0.026-0.16 mg kg^{-1}) — all non-essential plant TEs — with average backgrounds of 100, 40, 22, 2 and 0.06 mg kg^{-1} respectively (Iyaka, 2011; Kaiser et al., 2020; Mason & Moore, 1982; Sheffer et al., 2016). These measured TE concentrations were comparable to those reported in *E. guineensis* plantation soils in Nigeria, Ghana and Papua New Guinea (Aini Azura et al., 2012; Cole & Craven, 2006; Olafisoye et al., 2016; Orobator et al., 2018).

For all TEs except As, Mo, Cd and Ce, soils from Site D had significantly lower mean total concentrations compared to Sites A, B and C. Despite being present in higher concentrations compared to some other sites, these TEs were below background concentrations in Site D (6, 2, 0.06 and 49 mg kg^{-1} for As, Mo, Cd and Ce, respectively) (Mason & Moore, 1982; Perelomov, 2007). In general, Site D site had no elevated concentrations of TEs above expected background concentrations. Trace elements at Site D are lower compared to the other sites, as it is no longer in production and has had the trees cleared — a process that may have resulted in the erosion and run-off of topsoil (Suryatmojo et al., 2014) and any associated TEs contained there (Aflizar et al., 2018).

There were significant differences between *H. brasiliensis* and *E. guineensis* sites for total Cr, Mn, Co, Ni, Zn, Ce and Pb, presented in Table 3.3. Concentrations of Cr and Ni were lower in Site E compared to all four *E. guineensis* soils, while Mn, Zn, Ce and Pb were higher in Site E. The mean Mn, Zn, Ce and Pb concentrations in Site E (2995, 213 and 139 mg kg^{-1} respectively) were higher than global background levels (850, 50, 49 and 10 mg kg^{-1} respectively) (Mason & Moore, 1982; Perelomov, 2007). The Mn concentration present here was higher than would be expected from geogenic sources (Maynard, 2003), while Zn concentrations at this site exceeded the SGVs for agricultural land from Canada (200 mg kg^{-1}), the EU (94 mg kg^{-1} added Zn [2]), the US (160 mg kg^{-1}) and Denmark (100 mg kg^{-1}) (Cavanagh & Munir, 2016). These Zn concentrations may be phytotoxic, and this may limit production at the *H. brasiliensis* site (Chaney, 1993). The elevated Mn and Zn in Site E may be a result of fungicides — namely mancozeb — used to treat pests including *Corynespora cassiicola* (Berk. & Curt.), which causes leaf fall disease in *H. brasiliensis* trees (Fernando et al., 2010). Mancozeb is an ethylenebisdithiocarbamate compound containing Mn and Zn as active ingredients (Hurt et al., 2010; Mancino et al., 1999). Dithiocarbamate compounds (including Zn dithiocarbamate) (Shi et al., 2021) are used in *H. brasiliensis* production to accelerate vulcanisation (hardening) (Gullino et al., 2010), and thus if used in Site E, may have led to Mn and Zn inputs to soils.

While Ce has no specified toxicity thresholds in soil (Savichev & Vodyanitskii, 2009), it has been found to decrease germination speed and reduce plant biomass, particularly in tropical acidic soils (Moreira et al., 2019), and to be increasingly taken up into the roots and shoots of plants as soil pH decreases (Thomas et al., 2014). A HC_5 (hazardous concentration for 5% of plant species) value of 281.6 mg kg^{-1} was calculated for tropical Oxisols and Inceptisols (Moreira et al., 2019). As such, Ce concentrations in soils at Site E may not yet impact production, however, they have potential to do so in the future if accumulation continues. Lead concentrations at Site E, while elevated — likely as a result of phosphate fertiliser use (Setyorini et al., 2002; Taylor et al., 2016) — did not breach international SGVs [3] (70 mg kg^{-1} in Canada, 120 mg kg^{-1} in US, 166 mg kg^{-1} added Pb in EU and 300 mg

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[2] This SGV applies to soil with pH 5.5, CEC 15 cmol (+) kg^{-1} and 15% clay content

[3] SGVs specific to Indonesia were not identified

kg^{-1} in New Zealand) (Cavanagh & Munir, 2016) and are unlikely to affect production, however, may pose issues in the future if allowed to accumulate past SGVs.

Given the low pH of the soils, both non-essential and essential cations will be relatively soluble (Sheppard et al., 2009). $Ca(NO_3)_2$ extractable concentrations of TEs, used to calculate K_D values, are presented in Appendix C. When calculated K_D values were compared to K_D values for arable and pasture soils, Mn, Ni, Zn, Sr, Cd, Ce and Pb (arable/pasture soil K_D: 22,000, 3000, 6000, 190, 3000, 88,000 and 25,000, respectively) (Reiser et al., 2014; Sheppard et al., 2009) were more mobile in *E. guineensis* soils at all sites (Table 3.4). Cobalt was more mobile in Sites A, B and D (arable soil K_D 1800) (Sheppard et al., 2009), while Mo was more mobile at Sites C and D and less mobile at Sites A and B compared with arable soils (K_D 240) (Sheppard et al., 2009). Toxicity of these TEs may occur at lower concentrations than SGVs suggest in these *E. guineensis* soils, due to the acidic soil environment and associated increase in mobility and plant-uptake. Additionally, P may become even less plant-available in these soils due to its strong specific adsorption onto oxyhydroxides that occurs in acidic environments (McLaren & Cameron, 1996).

Table 3.4. K_D of TEs at each site under *E. guineensis* production. K_D values for Cd and Mn unable to be quantified.

Site	**A**	**B**	**C**	**D**
Cr	3179	2651	2900	5849
Co	279	195	2344	156
Ni	46	26	1389	25
Cu	904	358	5643	933
Zn	72	42	172	52
Ge	649	128	481	148
As	127	46	150	61
Sr	23	26	83	35
Mo	420	1615	43	28
Ce	658	102	615	106
Pb	175	76	257	37

Chromium was less mobile in the *E. guineensis* soils compared with arable soils (K_D 270) (Sheppard et al., 2009). Compared to other TEs, Cu was less mobile in *E. guineensis* soils, particularly at Site C. This may be due to Cu association with organic matter and Fe and Al oxides in these soils, which strongly bind Cu, decreasing its mobility (Oorts, 2013). Therefore, Cu may be less available to plants and exceedance of SGVs at Site B may not result in any negative effect on *E. guineensis* yield, despite high total concentrations.

Total and $Ca(NO_3)_2$ extractable concentrations of Pd, Te, Re, Ir and Hg are presented in Appendix C. These TEs were present in concentrations <0.1 mg kg^{-1} and considered to have no environmental consequences. Total Ag and Cs for Site E soils are also presented in Appendix C.

3.3.3 Factors controlling the variation between sites

Principal component one (PC1) explained 53.9% of variation between soils, and was dominated by Mn, Zn, Pb, Ce and Cr in descending order (Figure 3.3a). Principal component two (PC2) explained 31% of variance and was dominated (in descending order) by Ca, Cu, Ni, Sr, Co and Ce. The five sites formed distinct groups in the PCA plot (Figure 3.3b), with some overlap occurring between Sites A and B. Soils from the *H. brasiliensis* plantation had distinct chemistry separating it from the four sites under *E. guineensis* production. This is attributed to the history of land use and production.

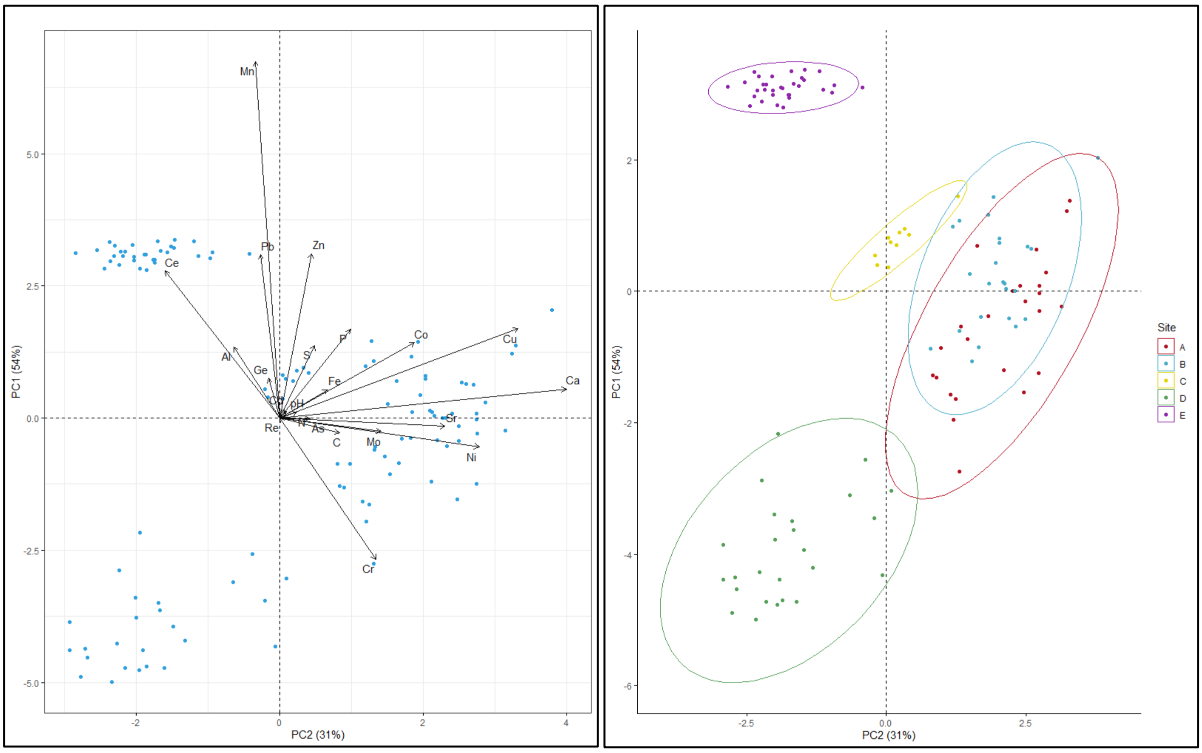

Figure 3.3 a (left) Loading plot showing variables and their influence on principal components 1 and 2; and 3.3b (right) PCA plot with sites identified by colour. Ellipses represent 95% confidence around the mean.

The chemistry of Sites A, B, C and D separated out according to a combination of edaphic and anthropogenic factors when a PCA was done using only soils from the four *E. guineensis* sites (see Appendix C for PCA results of *E. guineensis* sites including general soil parameters).

3.3.4 Intra-site correlations

Correlograms for all five sites are presented in Figure 3. 4. All correlations presented are statistically significant ($p < 0.01$). Soil variables are grouped by hierarchical clustering, whereby similarly behaving variables that share common correlations are grouped closer together.

At Sites A, B and C there was a common positive correlation between P and C ($r = 0.66$ S**, 0.64 S** and 0.79 S* respectively). At Sites A and B, P was also positively correlated with N ($r = 0.62$ S** and 0.56 S* respectively). As organic P in soils positively correlates with organic matter, the correlations at Sites A, B and C indicate that soil P at these sites was mostly associated with organic matter in the soil, and unavailable to plants (Wierzbowska et al., 2020). Carbon, N and S were mutually positively correlated at all *E. guineensis* sites ($0.63 < r < 0.94$ S*-S**), indicating that the majority of N and S in soils at these sites were also organically associated. At Site E, C and N positively correlated ($r = 0.61$ S**) however, there were no other significant correlations between C, N, P and S. This indicates that in contrast to the *E. guineensis* sites, N, P and S in soils from the *H. brasiliensis* plantation were associated with the mineral phase of the soil and may have been more plant-available.

There is some evidence for agricultural correlation with TEs in the *E. guineensis* plantation soils when P is used as a proxy for agriculture. Phosphorus correlated positively with Co (r = 0.52 S*), Mn (r = 0.58 S*), Zn (r = 0.51 S*) and Cd (r = 0.72 S**) at Site A, with Mo at Site C (r = 0.81 S*) and with Sr (r = 0.67 S**) at Site D. All of these TEs are present within fertilisers and thus these correlations are typical of past or present agricultural sites (Németh et al., 2006; Taylor et al., 2016). Where P does not correlate with TEs, we reject the hypothesis that TEs are fertiliser-derived and conclude that any present are likely geogenic.

Correlations existed between variables that would typically be expected in soils: BS positively correlated with pH at all four *E. guineensis* sites ($0.59 < r < 0.91$ S*-S**). Although not shown in Figure 3.4, soil pH also correlated negatively with $Ca(NO_3)_2$ extractable Al at Site A (r = -0.93 S**), Site B (r = -0.89 S**) and Site C (r = -0.95 S**), and negatively with extractable Fe at Site A (r = -0.74 S**) and Site B (r = -0.66 S**). Calcium positively correlated with both BS ($0.88 < r < 0.96$ S**) and pH ($0.71 < r < 0.99$ S**) at all *E. guineensis* sites.

Concentrations of $Ca(NO_3)_2$ extractable cationic TEs including Cr, Ni, Cu, Zn and Cd (see Appendix C) typically negatively correlate with pH, as increasing soil pH can lower the availability of TEs in soil solution (Kim et al., 2012; Kim et al., 2016). While $Ca(NO_3)_2$ extractable TEs were not included in correlograms, this was the case at Site A, where pH negatively correlated with $Ca(NO_3)_2$ extractable Cr (r = -0.51 S*), Ni (r = -0.65 S**), Zn (r = -0.85 S**) and Ce (r = -0.55 S*). At Site B, pH negatively correlated with $Ca(NO_3)_2$ extractable Cu (r = -0.55 S*) and Zn (r = -0.58 S**), while at Site C, pH negatively correlated with $Ca(NO_3)_2$ extractable Ge (r = -0.86 S*) and Ce (r = -0.87 S*).

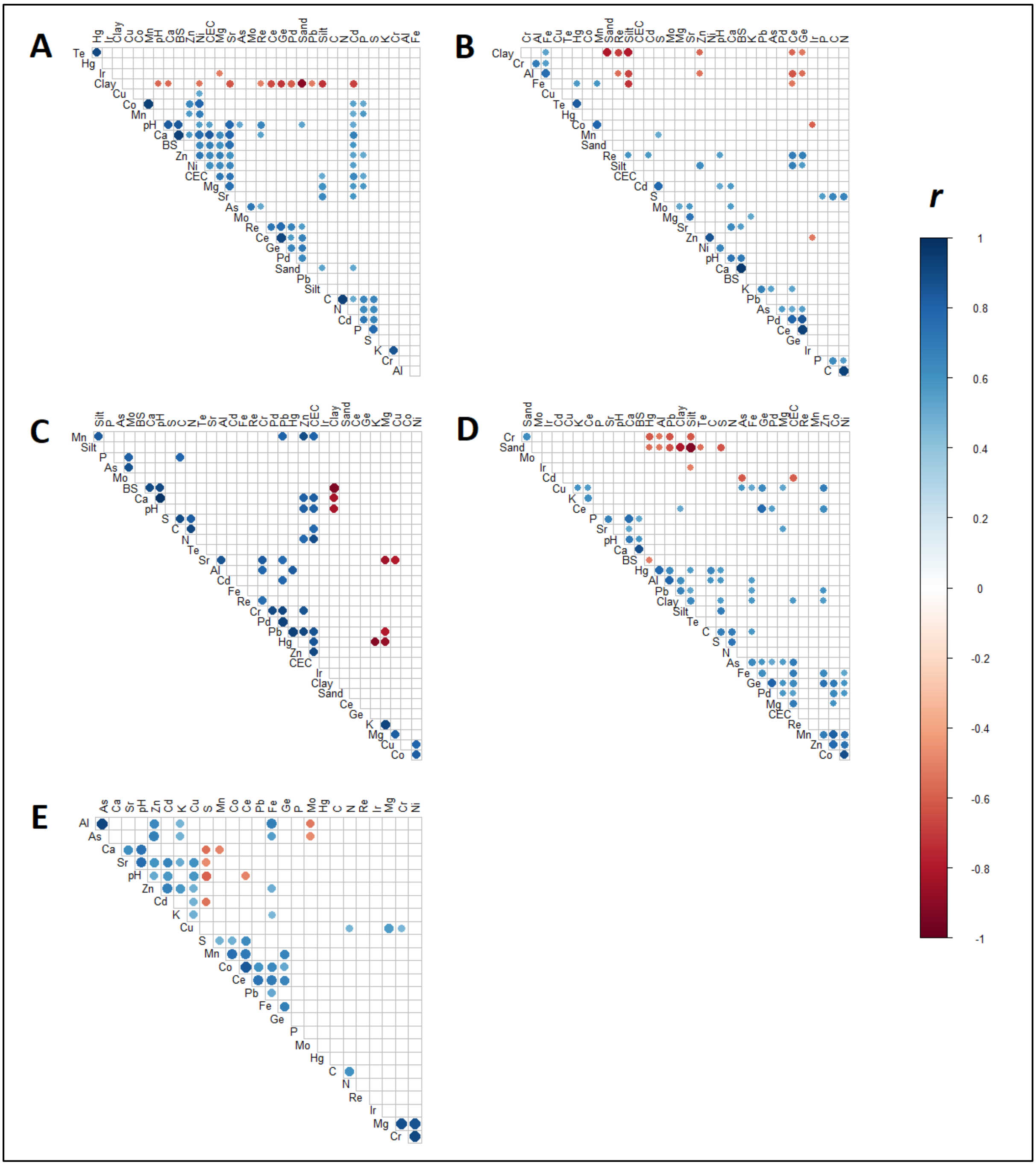

Figure 3.4. Correlograms showing intra-site correlations ($p < 0.01$) between variables, grouped by hierachical clustering

3.4 Conclusions

The soils we have investigated under *E. guineensis* and *H. brasiliensis* production were acidic, with low P concentrations. Soil N, P and S were associated with organic matter in *E. guineensis* production soils, which was low compared to published data on forest soils from this region. Production at *E. guineensis* sites is likely to be severely limited by deficiency of N, P, K, Mg and Mo in soils. There was evidence of agriculturally-derived TEs accumulating in both *H. brasiliensis* and *E. guineensis* soils: Mn, Zn, Ce and Pb were significantly higher in *H. brasiliensis* soils, likely due to the application of these TEs as ingredients or contaminants in agrichemicals at this site. These TEs may already limit production. In addition, *E. guineensis* soils contained elevated concentrations of Cu, Zn, As and Pb. Based on K_D values, most TEs were more mobile in soils under *E. guineensis* production compared to other arable production, likely due to the low pH of these tropical soils. Consequently, toxicity thresholds may be reached at lower concentrations than SGVs suggest. Soils under *H. brasiliensis* production were chemically distinct from soils under *E. guineensis* production and the chemistry at *E. guineensis* sites was consistent with that of soils with a short fertiliser history. While our sites varied in terms of landscape elements and prior or current management, the trends in the data were consistent. This indicates that nutrient deficiencies may be present in *E. guineensis* production soils throughout Indonesia. Correction of nutrient deficiencies in these *E. guineensis* plantations would necessitate significant fertiliser inputs to soils, which would be of considerable cost and negatively affect the economic revenue gained through *E. guineensis* production. These inputs would also lead to increased TE-loadings in soils, exacerbating issues of TE accumulation. As Cu, Zn, As and Pb were already elevated at both active and former *E. guineensis* production sites, likely as a result of fertiliser use, these TEs in particular are likely to accumulate past SGVs and eventually limit production with increased fertiliser inputs. Subsidised *E. guineensis* production may not be economically viable, yet still results in the degradation of significant areas of tropical ecosystems for production that is challenged by low soil fertility, and sometimes abandoned. Further work should determine whether *E. guineensis* production would be economically viable without subsidies. Such an analysis should include the costs and ecological impacts of applying sufficient fertiliser to enable economically-viable *E. guineensis* production.

3.5 Statements and declarations

Conflicts of interest: The authors have no competing interests to declare that are relevant to the content of this article.

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Data availability: The data that support the findings of this study will be made openly available upon publication at www.kiwiscience.com [full URL will be generated]. Data is available from the authors upon request preceding publication.

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Chapter 4: Chemical elements in plants and oils from current and former palm oil production systems

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Abstract

The production of oil palm (*Elaeis guineensis* Jacq.) in Southeast Asia is vital to the economies of Indonesia and Malaysia. Both fertilisers and pesticides used in *E. guineensis* production can contain elevated concentrations of Trace Elements (TEs) that may accumulate in soils and leaf tissues of plants. We hypothesised that leaves from *E. guineensis* may be deficient in essential elements, while containing elevated concentrations of non-essential TEs commonly found in agrichemicals. Samples of plant materials (leaves and fruitlets) were collected from active and former plantations in Sumatra, Indonesia, and analysed for essential and non-essential elements. Indonesian palm oil samples were sourced in New Zealand and their elemental concentrations determined. Leaf materials from both active and abandoned production sites were deficient in N, K, S and Mo, while leaf materials from abandoned sites were deficient in P. These deficiencies may have been a contributing factor to the abandonment of production at these sites. Concentrations of non-essential elements were below or comparable to average plant concentrations and no evidence of contamination was found in plant tissues. Copper however, had bioaccumulation coefficients in plant tissues from two sites that indicated either elevated plant uptake up from soils or surface contamination from direct application of Cu-containing agrichemicals. This may pose toxicity risks for *E. guineensis*-based food and fodder products if plants are grown in soils with excessive Cu concentrations. Palm oil contained low concentrations of TEs, which did not pose any toxicity risks. However, Na and Al were present in concentrations of 1198 and 159 mg kg^{-1} respectively, which were higher than have been previously reported. Tropical *E. guineensis* production could benefit from the determination of bioaccumulation factors for fertiliser contaminants in *E. guineensis* grown under varying conditions, to limit the transfer of contaminants to plants and products if increased fertiliser applications were used to correct nutrient deficiencies.

Keywords

Elaeis guineensis, fertilisers, trace elements, nutrients, contaminants

4.1 Introduction

The African oil palm (*Elaeis guineensis* Jacq.) is an economically important crop, producing the world's most consumed vegetable oil (Shahbandeh, 2022). Palm oil represents 10% of Indonesia's exports and 5% of Malaysia's, with these two countries being the world's major cultivators of *E. guineensis* (OEC, 2020). A 2016 assessment determined that 45% of harvested *E. guineensis* plantation land in Southeast Asia had been converted from tropical forests since 1989 (Vijay et al., 2016). Tropical forest soil is typically nutrient-poor and requires inputs of both capital and maintenance nutrients through fertilisers to support production (Foy, 1984; Hartemink, 2002). The essential elements of most importance for *E. guineensis* are N, P, K, Mg and B (Ng, 2002). Inadequate supply of these elements through fertilisers can lead to yield reductions in *E. guineensis* production, particularly on acidic and weathered tropical soils such as those from Indonesia and Malaysia (Ng, 2002; Woittiez et al., 2015). Plant deficiencies can be assessed through the leaf concentrations of essential elements.

Fertiliser-borne trace elements (TEs) can accumulate in soil to concentrations that exceed guideline values (Chaney, 1989). Pesticides and other agrichemicals used to support production are also potential sources of TEs (Aderungboye, 1977; MPOB, 2015; Ng, 2002). If soil-accumulated TEs are taken up by plants, this has potential for products to exceed food safety standards (Chaney, 1989; Rys, 2011). The transfer of TEs into plant products is a function of plant TE uptake, surface deposition and post-harvest contamination. The root absorption of TE ions in soil solution occurs via the symplastic and apoplastic pathways, followed by xylem transport to leaves and potential phloem transport to fruit tissues (Marschner, 2012). Spray residues containing TEs can sorb to plant leaves after direct pesticide applications, while deposition of soil containing TEs onto plant parts used for manufacturing can lead to plant products containing TEs (Kabata-Pendias & Mukherjee, 2007). Trace elements can also enter products due to contamination from machinery and equipment during manufacturing, processing and storage post-harvest (Corley & Tinker, 2015; Kabata-Pendias & Pendias, 2001). The uptake of ions from soil solution is governed by its bioavailability: high total concentrations of TEs in soil do not always correspond with high uptake (Hooda, 2010). Bioavailability is a function of the solubility and speciation of the TE in soil, the presence of competing ions as well as the receptor organism (Marschner, 2012). Leaf concentrations — particularly pinnae concentrations in *E. guineensis* — of essential elements are better indicators of plant nutrition than the total soil concentration (Von Uexküll & Fairhurst, 1991). The presence of TEs in vegetable oils as a result of plant uptake and transport is typically limited due to their partitioning away from the lipid phase in plants (Salisbury & Ross, 1992). This is supported by no identification of concentrations of TEs in processed palm oils that breached food safety standards (Thompson-Morrison et al., 2022a). Although some TEs may be present in crude palm oil, these are removed during refining processes (Gupta, 2017; Rossi et al., 2003; Szydłowska-Czerniak et al., 2013).

Concurrent research on *E. guineensis* plantation soils from Indonesia (Thompson-Morrison et al., 2022b) (Chapter 3) determined that the soils analysed were deficient in essential nutrients N, P, K, Mg and Mo, and elevated above background levels in TEs Cu, Zn, As and Pb. Furthermore, the soils analysed were acidic and TEs were more mobile compared to other arable systems. In the current work, we analysed *E. guineensis* plant materials – leaves, rachides, endosperms and mesocarps — from the same plantations as well as the *E. guineensis* product palm oil for their essential nutrient and TE composition.

We hypothesise that *E. guineensis* leaf concentrations of N, P, K, Mg and Mo will be deficient and below concentrations of 2.3%, 1400 mg kg^{-1}, 7500 mg kg^{-1}, 2000 mg kg^{-1} and 0.3 mg kg^{-1}, respectively. Furthermore, we hypothesise that concentrations of Cu, Zn, As and Pb may be elevated in plant tissues. We hypothesise that palm oil will contain inconsequential levels of all elements below tolerable upper limits for food safety. This research aims to determine whether plants are deficient in essential nutrients and elevated in TEs, and thus the benefits and risks of using *E. guineensis* products with respect to elements.

4.2 Methods

4.2.1 Site descriptions and sample collection

Plant material samples were collected from three *E. guineensis* plantations in Sumatra, Indonesia in May 2019. All plantations shared a common soil type of Acrisol (taxonomically related to Oxisols according to the USDA soil classification (Encyclopaedia Britannica, 2016)). The sampling locality (approximately 3.5952° N, 98.6722° E) is reported in Thompson-Morrison et al. (2022b). Materials collected included leaves, rachides and whole kernels, which were divided into mesocarp and endosperm prior to analysis. Plant material was cleaned with water and oven dried at 60°C until a constant weight was obtained. All samples were ground prior to analysis.

The study was conducted in accordance with the relevant institutional, national, and international guidelines. Permissions were obtained from landowners for the collection of plant material and plants were identified by the landowners or farmers present during sampling. Samples were sent to the University of Canterbury in New Zealand under approved permits from both New Zealand's Ministry for Primary Industries (permits #2019072466 for kernel material and #2018068845 for leaf material) and Indonesia's Agency for Agricultural Quarantine (phytosanitary certificate #2019.2.0702.0.K10.E.000785 for kernel material and #2019.2.0702.0.K10.E.000728 for leaf material). Kernels were crushed and ground prior to export to ensure non-viability. Once in New Zealand, samples were stored and analysed in Physical Containment Level 2 approved facilities with kernels stored at ~4°C.

Sites A and B bordered Gunung Leuser National Park and were abandoned after ca. 15 years of production. These sites were adjacent to each other and were separated northeast (Site A)/southwest (Site B). At the time of sampling, the sites were overgrown and had had no amendments applied for at least 3 years. Sites A and B had slopes of approximately 12° in places and samples were taken based on practicality and accessibility from areas in which *E. guineensis* trees remained.

Site C was also near to Gunung Leuser National Park, however, was an active smallholder *E. guineensis* plantation. This site had been under production for ca. 20 years. The slope of this site was <4°.

Samples of refined palm oil were sourced from an Indonesian food manufacturer whose food products are marketed in New Zealand.

4.2.2 Analytical methods

4.2.2.1 Pseudo-total element concentrations

We analysed pseudo-total element concentrations (hereafter referred to as elemental concentrations) of all samples using microwave digestion, following EPA Method 3051A (US EPA, 2007). A Ultrawave Single Reaction Chamber Microwave Digestion Unit was used. In each analysis, 0.20 g (±0.01 g) was weighed into a digestion tube and 5 mL 70% HNO_3 was added. Samples were pre-digested overnight and then digested in the Ultrawave at 230°C for 25 minutes (Milestone, n.d.). Two reagent blanks and two standard reference materials (INCT, 2010; NIST, 1995) were included with every 11 samples for quality assurance. Digestates were analysed by inductively coupled plasma-mass spectrometry along with standard reference materials (NIST, 2015). Recoveries of SRMs ranged from 79-134% of certified values. A full list of SRM recoveries is available in Appendix D. All concentrations reported are on a dry weight basis. The limit of quantification for analyses was 0.01 mg kg^{-1}.

4.2.2.2 Total carbon and nitrogen

Total C and N concentrations of all samples were analysed by C and N analyser: CN828 Elemental Analysis by Combustion. Blanks and SRMs (LECO, 2019) were included with samples for quality assurance.

4.2.3 Statistical analysis

R (version 4.1.0) was used for all statistical analysis (R Core Team, 2021). One-way parametric Analysis of Variance (ANOVA) was run to compare means of each sample type between site using the package multcomp (Hothorn et al., 2008). Data was (natural) log-transformed where assumptions of normality were not met and non-parametric permutational ANOVA was used when assumptions of homoscedacity were not met. In most cases there were no differences between the results of the parametric and non-parametric ANOVA and the parametric test was considered robust. When significant differences were identified, Tukey's Honest Significant Difference test was run to determine which sites these existed between.

4.2.4 Bioaccumulation coefficients

Bioaccumulation coefficients (BACs) give an indication of a plant's ability to accumulate heavy metals from soils (Koleli et al., 2015). They can be calculated as the plant / soil TE concentration quotient for each plant tissue using the equation

$$BAC = \frac{TE\ (plant)}{TE\ (soil)} \qquad \text{(Mishra \& Pandey, 2019).} \qquad \text{(Eq. 4.1)}$$

Bioaccumulation coefficients were calculated for pinnae, endosperm and mesocarp tissue from each site. These were calculated for Cu, Zn, As, Sr, Cd and Pb using soil concentrations from these sites detailed in Thompson-Morrison et al. (2022b). These TEs were chosen as they are common contaminants of agrichemicals used to support production (Taylor et al., 2016).

4.3 Results and discussion

4.3.1 Elements in *E. guineensis* leaves (pinnae and rachides)

4.3.1.1 Essential elements

Deficiencies of N, K, S and Mo were present in pinnae samples from all sites, whereas P deficiency was present in pinnae samples from abandoned Sites, A and B (Table 4.1). The leaf tissue samples tended to contain higher concentrations of both essential and non-essential elements compared to fruit (kernel and mesocarp) tissue. This is consistent with plant physiology, given leaves are a water-sink and elements taken up via the roots are transported here via xylem flow (Marschner, 2012).

Mean concentrations of N, S and Mo in pinnae samples were in the deficient range for *E. guineensis* production at all sites. In contrast, P was deficient in 100% of samples at the abandoned sites (A and B) and mean K was deficient at Sites B and C (Table 4.1). Nitrogen deficiencies were present in all pinnae samples from the three sites. The mean N concentrations at Site C (1.9%) were comparable to those of *E. guineensis* grown on Malaysian Oxisols of 1.8% (Corley & Tinker, 2015). Across all sites, the N concentrations (1.31.9%) were lower than those of *E. guineensis* grown in Nigeria (2.1%) and Indonesia (2.3-2.7%) (Corley and Tinker, 2015; Ng et al., 1968, as cited in Corley and Tinker, 2015; Woittiez et al., 2015). For the Indonesian *E. guineensis*, 2.3% N was considered deficient. Nitrogen deficiencies have also been reported in *E. guineensi*s from Cameroon (Rafflegeau et al., 2010).

Phosphorus in all pinnae samples from Sites A and B were in the deficiency range for *E. guineensis* (Table 4.1). These P concentrations were lower than those reported in Indonesia, Malaysian and Nigerian *E. guineensis* leaves of 1500-1700 mg kg^{-1} (Woittiez et al., 2015), 1210-1350 mg kg^{-1} (Corley & Tinker, 2015) and 1280 mg kg^{-1} (Ng et al., 1968, as cited in Corley and Tinker (2015)), respectively. The P concentration in leaves at Site C (1689 mg kg^{-1}) was within the adequate range for *E. guineensis* and comparable to the value of 1700 mg kg^{-1} from *E. guineensis* also grown in Indonesia reported by Woittiez et al. The range of leaf P concentrations across the sites were comparable to those of New Zealand native species grown in low-fertility systems in nutrient-poor, non-fertilised soils (Esperschuetz et al., 2017). This indicates the low nutrient status of the *E guineensis* trees we sampled and is consistent with inadequate fertilisation. Sites A and B may have had low native P concentrations followed by insufficient capital and maintenance nutrients applied during production. Unlike N, P concentrations do not typically drop precipitously following site abandonment as P may bind to soil colloids and be retained in soils (McLaren & Cameron, 1996). This indicates that P concentrations at Sites A and B were sub-optimal during production and may have been a contributing factor to low yields and subsequent site abandonment.

Rachis concentrations of K have been suggested as a more accurate indication of plant K status compared to pinnae concentrations as they provide a more accurate indication of fruit yield (Chew & Teoh, 1987). The K concentrations of the rachis samples range from 6204 mg kg^{-1} at Site B to 8969 mg kg^{-1} at Site A, indicating K deficiency (Table 4.1). Potassium deficiency is the most limiting nutrient factor affecting yield in *E. guineensis* production (Cui et al., 2020; Rankine & Fairhurst, 1999; Von Uexküll & Fairhurst, 1991). This deficiency has been attributed to inadequate K fertilisation of production soils that already have naturally low background levels (Rafflegeau et al., 2010; Woittiez et al., 2015). Cost has been a barrier to K fertilisation for Indonesian smallholders, as K fertilisers are not subsidised (Woittiez et al., 2015). Concentrations of K in *E. guineensis* leaves from Indonesia and Malaysia have also been reported in deficient ranges of 6000-6300 mg kg^{-1} (Woittiez et al., 2015) and 5700-7600 mg kg^{-1} (Corley & Tinker, 2015) respectively. Non-deficient levels of 8800 mg kg^{-1} were measured in Nigerian *E. guineensis* leaves (Corley and Tinker, 2015; Ng et al., 1968, as cited in Corley and Tinker, 2015).

Table 4.1. Mean essential element concentrations in *E. guineensis* leaf material (pinnae and rachides) at each site (standard error of the mean in parentheses) compared with available literature values for deficiency, adequate growth and toxicity, where available. All values in mg kg^{-1} unless otherwise specified. Sites that share the same letter(s) for a single variable are not significantly different from each other.

Site			**A (n=10)** [*]	**B (n=7)**	**C (pinnae n=2, rachides n=1)**	**Deficient** [+]	**Adequate** [+]	**Excessive/ Toxic** [+]
Pinnae	**Macro-elements**	**C (%)**	48(0.35)^{a}	47(0.20)^{a}	48(0.15)^{a}	—	—	—
		N (%)	1.4(0.090)^{a}	1.3(0.077)^{a}	1.9(0.010)^{b}	<2.3	2.4-2.8	>3.0
		P	1102(49)^{a}	1054(27)^{a}	1689(144)^{b}	<1400	1500-1800	>2500
		K	9266(982)^{a}	7109(857)^{a}	5350(249)^{a}	<7500	9000-12,000	>16,000
		S	1729(119)^{a}	1799(148)^{a}	1804(24)^{a}	<2000	2500-3500	>6000
		Mg	3139(376)^{a}	2883(354)^{a}	2476(316)^{a}	<2000	2500-4000	>7000
	Micro-elements	**B**	21(3.1)^{a}	24(1.8)^{a}	12(5.5)^{a}	<8.0	15-25	>40
		Mn	272(44)^{a}	910(197)^{b}	705(150)^{ab}	<35	50 [†]	400-1000 [‡]
		Fe	83(5.1)^{a}	80(5.3)^{a}	68(7.1)^{a}	—	100 [†]	—
		Cu	5.8(0.26)^{a}	5.9(0.73)^{a}	7.6(2.2)^{a}	<3.0	5.0-8.0	>15
		Zn	25(1.4)^{a}	24(1.6)^{a}	23(0.68)^{a}	<10	12-18	>80
		Mo	0.13(0.025)^{ab}	0.060(0.010)^{a}	0.24(0.040)^{b}	0.10-0.30 [‡]	0.50-0.80	10-50 [‡]
Rachides	**Macro-elements**	**C (%)**	47(0.14)^{a}	47(0.19)^{a}	48^{a}	—	—	—
		N (%)	0.20(0.024)^{a}	0.21(0.037)^{a}	0.61^{b}	—	—	—
		P	462(52)^{a}	687(184)^{a}	1316^{a}	—	—	—
		K	8969(1760)^{a}	6204(876)^{a}	8614^{a}	<13,000	13,100-16,000	>16,000
		S	837(96)^{a}	998(126)^{a}	947^{a}	—	—	—
		Mg	742(145)^{a}	898(43)^{a}	1117^{a}	—	—	—
	Micro-elements	**B**	28(4.2)^{a}	25(5.0)^{a}	22^{a}	—	—	—
		Mn	35(7.0)^{a}	132(26)^{b}	78^{ab}	—	—	—
		Fe	63(17)^{a}	61(12)^{a}	19^{a}	—	—	—
		Cu	2.4(0.17)^{a}	2.3(0.22)^{a}	2.3^{a}	—	—	—
		Zn	10(1.1)^{a}	13(2.1)^{a}	7.8^{a}	—	—	—
		Mo	0.20(0.048)^{a}	0.16(0.031)^{a}	0.020^{a}	—	—	—

[*] B n=9

[+] Values specific to *E. guineensis* growth for palms >6 years old (Von Uexküll & Fairhurst, 1991)

[†] Typical values for adequate plant growth as none specific to *E. guineensis* were identified (Marschner, 2012)

[‡] Typical values for adequate plant growth as none specific to *E. guineensis* were identified (Kabata-Pendias & Pendias, 2001)

Among the three sites, 90% of pinnae samples from Site A, 71% of samples from Site B and 100% of samples from Site C were deficient in S. The S concentrations were comparable to those of Nigerian *E. guineensis* (1740 mg kg^{-1}) (Corley and Tinker, 2015; Ng et al., 1968, as cited in Corley and Tinker, 2015). Boron was below the optimal range at Site C, and lower than reported concentrations for *E. guineensis* leaves also grown in North Sumatra (16.4-19.5 mg kg^{-1}), which were expressing B deficiency symptoms (Jacquemard et al., 2006). No studies reporting Mo concentration of *E. guineensis* leaves could be identified for comparison.

Magnesium, Fe, Cu and Zn in pinnae samples were within the adequate range for *E. guineensis* growth at all sites (Table 4.1). The Mg concentrations were comparable to those in leaves of *E. guineensis* grown in Indonesia and Nigeria of 2800-3000 mg kg^{-1} and 2330 mg kg^{-1} (Corley and Tinker, 2015; Ng et al., 1968, as cited in Corley and Tinker, 2015), respectively. Zinc concentrations were comparable to leaf concentrations from Malaysian *E. guineensis* plants of ca. 15-28 mg kg^{-1} (Aini Azura et al., 2012). The Fe and Cu pinnae concentrations were within typical ranges contained in plant leaves (Marschner, 2012). Manganese was present in high concentrations in pinnae samples, however, still within the range of concentrations likely to be present in plants (El-Jaoual & Cox, 1998). Toxicity thresholds for Mn are plant- and even cultivar-specific (Marschner, 2012), and no specific thresholds for *E. guineensis* have been identified. Considering differences between active and former production sites, the active site (C) was significantly higher in N and P than former sites (A and B) and significantly higher in Mo than Site B. This may be due to the Site C's ongoing cultivation at the time of sampling, and thus more recent fertilisation.

Our leaf samples were collected from plantations with soils deficient in N, P, K and Mo (Thompson-Morrison et al., 2022b). Thus, it is unsurprising that plant concentrations of these essential elements are sub-optimal and likely to be limiting production, as plants source the majority of their nutrient requirements from bioavailable elements in soils through diffusion and mass flow (Marschner, 2012). These deficiencies are attributed to the conversion of acidic, low-fertility forest soils to production without the full amount of necessary inputs of both capital and maintenance nutrients (Foy, 1984; Hartemink, 2002). As *E. guineensis* producers in Indonesia rely heavily on subsidised fertilisers, these deficiencies — particularly K for which no subsidised fertilisers are available — are commonplace, particularly in smallholder plantations (Cui et al., 2020; Woittiez et al., 2015).

4.3.1.3 Non-essential trace elements

Most non-essential elements were present in concentrations less than the average concentrations for terrestrial plants (Table 4.2). The low concentrations of Cr (1.7-4.7 mg kg^{-1} in pinnae, 1.1-10 mg kg^{-1} in rachides) and Ti (1.5-1.8 mg kg^{-1} in pinnae, 0.60-0.87 mg kg^{-1} in rachides) indicate that the non-essential elements present in *E. guineensis* leaves were not a result of contamination of leaf material with soil dust. While Ti, Cr, Ni and Hg were present above average terrestrial plant concentrations (Mason & Moore, 1982), they were below toxicity thresholds for leaf tissues (Kabata-Pendias & Pendias, 2001).

Mean concentrations of Na and Si were higher in rachides (170-192 mg kg^{-1} and 188-291 mg kg^{-1}, respectively) compared with pinnae (54-116 mg kg^{-1} and 1.3 mg kg^{-1}, respectively). Cadmium concentrations (0.020-0.027 mg kg^{-1} in pinnae, 0.010-0.014 mg kg^{-1} in rachides) were lower than those reported from Malaysian *E. guineensis* leaves (0.18-0.38 mg kg^{-1}) (Aini Azura et al., 2012). Silver and Au were present in leaf tissues in concentrations <0.1 mg kg^{-1} and pose no toxicity risks (Saleeb et al., 2019).

The pinnae concentrations of Te at Site C (0.29 mg kg^{-1}) were significantly higher than at Sites A and B, and higher than those reported in Angiosperm leaves of 0.017 mg kg^{-1} (Cowgill, 1988). Cerium was also significantly higher in pinnae from Site C compared to Sites A and B. There is a scarcity of data on Te and Ce in plants with which to compare our results. Pinnae from Site C contained significantly higher concentrations of Cs than Site A, while rachides from Site C were significantly higher in Cs than Sites A and B. Caesium concentrations in pinnae at Sites B and C (0.25 and 0.41 mg kg^{-1}, respectively) and rachides at Site C (0.74 mg kg^{-1}) were higher than average plant concentrations of 0.2 mg kg^{-1}

(Mason & Moore, 1982). The higher Te, Cs and Ce at Site C may be due to current production, as these TEs may be present in NPK fertilisers that were being used at the site at the time of sampling.

Table 4.2. Mean concentrations of non-essential trace elements in *E. guineensis* leaves (pinnae and rachides) at each site (standard error of the mean in parentheses) with available literature values for toxicity and tolerability in agronomic crops. All values in mg kg^{-1}. Sites that share the same letter(s) for a single variable are not significantly different from each other.

Site		**A (n=10)**	**B (n=7)**	**C (pinnae n=2, rachides n=1)**	**Terrestrial plants average [*]**	**Excessive/ toxic to plants [+]**
Pinnae	**Na**	116(15)^{a}	111(21)^{a}	54(20)^{a}	1200	—
	Al	46(5.4)^{a}	44(4.5)^{a}	30(9.2)^{a}	500	—
	Si	1.3(0.019)^{a}	1.3(0.014)^{a}	1.3(0.035)^{a}	200	—
	Ti	1.8(0.18)^{a}	1.6(0.20)^{a}	1.5(0.20)^{a}	1	50-200
	Cr	4.2(0.92)^{a}	4.7(1.1)^{a}	1.7(0.52)^{a}	0.23	5-30
	Co	0.071(0.010)^{a}	0.093(0.020)^{a}	0.040(<0.01)^{a}	0.5	15-50
	Ni	3.6(0.51)^{b}	5.8(1.0)^{b}	1.1(0.080)^{a}	3	10-100
	As	0.030(<0.01)^{a}	0.031(<0.01)^{a}	0.025(<0.01)^{a}	0.2	5-20
	Sr	16(2.7)^{a}	9.7(0.85)^{a}	17(0.90)^{a}	26	—
	Ag	<0.01^{a}	<0.01^{a}	<0.01^{a}	0.06	—
	Cd	0.027(<0.01)^{a}	0.023(<0.01)^{a}	0.020(<0.01)^{a}	0.6	5-30
	Te	<0.01^{a}	<0.01^{a}	0.29(0.29)^{b}	—	—
	Cs	0.18(0.030)^{a}	0.25(0.035)^{ab}	0.41(0.045)^{b}	0.2	—
	Ce	0.22(0.048)^{a}	0.39(0.061)^{a}	0.85(0.26)^{b}	—	—
	Au	<0.01^{a}	<0.01^{a}	<0.01^{a}	0.002	—
	Hg	0.045(<0.01)^{a}	0.044(<0.01)^{a}	0.052(0.015)^{a}	0.015	1-3
	Pb	0.64(0.060)^{a}	0.76(0.12)^{a}	0.62(0.15)^{a}	2.7	30-300
Rachides	**Na**	173(19)^{a}	170(14)^{a}	192^{a}	1200	—
	Al	18(2.7)^{a}	24(3.6)^{a}	13^{a}	500	—
	Si	273(28)^{a}	291(29)^{a}	188^{a}	200	—
	Ti	0.60(0.063)^{a}	0.87(0.11)^{a}	0.60^{a}	1	—
	Cr	10(3.6)^{a}	8.8(2.1)^{a}	1.1^{a}	0.23	—
	Co	0.134(0.040)^{a}	0.14(0.030)^{a}	0.020^{a}	0.5	—
	Ni	5.3(1.6)^{a}	5.5(1.2)^{a}	0.50^{a}	3	—
	As	<0.01^{a}	<0.01^{a}	0.010^{a}	0.2	—
	Sr	11(1.2)^{a}	6.9(0.86)^{b}	6.6^{ab}	26	—
	Ag	<0.01^{a}	<0.01^{a}	<0.01^{a}	0.06	—
	Cd	0.014(<0.01)^{a}	0.010(<0.01)^{a}	0.010^{a}	0.6	—
	Te	<0.01^{a}	<0.01^{a}	<0.01^{a}	—	—
	Cs	0.092(0.020)^{a}	0.18(0.020)^{b}	0.74^{c}	0.2	—
	Ce	0.060(0.010)^{a}	0.090(0.020)^{a}	0.070^{a}	—	—
	Au	<0.01^{a}	<0.01^{a}	<0.01^{a}	0.002	—
	Hg	<0.01^{a}	<0.01^{a}	<0.01^{a}	0.015	—
	Pb	0.27(0.051)^{a}	0.35(0.080)^{a}	0.080^{a}	2.7	—

[*] Mason and Moore (1982)

[+] Kabata-Pendias and Pendias (2001)

4.3.2 Elements in *E. guineensis* endosperms and mesocarps

4.3.2.1 Essential elements

The essential elements in the endosperm tissues (Table 4.3) were comparable to those in Malaysian and Nigerian *E. guineensis* endosperms, except for Mg and Fe, which were both lower in our samples (Aini Azura et al., 2012; Akpanabiatu et al., 2001; Kok et al., 2011). Endosperm tissues contained the highest P and Cu concentrations of all plant materials analysed. Mesocarp tissues tended to contain the lowest concentrations of most elements, particularly essential plant elements, of all plant materials analysed.

Table 4.3. Mean concentrations of essential elements in analysed *E. guineensis* endosperms and mesocarps from each site (standard error of the mean in parentheses). All values in mg kg^{-1} unless otherwise specified. Sites that share the same letter(s) for a single variable are not significantly different from each other.

	Site		**A (n=8)**	**B (n=7)**	**C (n=3)**
Endosperm	**Macro-elements**	**C (%)**	61(0.37)^{a}	62(0.41)^{a}	60(1.0)^{a}
		N (%)	1.4(0.10)^{a}	1.3(0.12)^{a}	1.3(0.19)^{a}
		P	3618(131)^{a}	3827(189)^{a}	3125(16)^{a}
		K	4870(109)^{a}	5045(326)^{a}	4932(414)^{a}
		S	1665(79)^{a}	1554(89)^{a}	1456(114)^{a}
		Mg	1958(76)^{a}	1937(115)^{a}	1574(105)^{a}
	Micro-elements	**B**	67(7.8)^{b}	37(3.8)^{a}	52(6.4)^{ab}
		Mn	136(18)^{a}	285(69)^{a}	157(20)^{a}
		Fe	32(1.3)^{a}	37(2.7)^{a}	37(5.5)^{a}
		Cu	17(0.73)^{a}	16(0.8)^{a}	17(1.7)^{a}
		Zn	24(1.4)^{a}	27(2.3)^{a}	26(2.6)^{a}
		Mo	0.24(0.033)^{ab}	0.34(0.060)^{b}	0.10(0.019)^{a}
Mesocarp	**Macro-elements**	**C (%)**	66(1.7)^{a}	70(1.0)^{a}	64(4.4)^{a}
		N (%)	0.29(0.020)^{a}	0.31(0.044)^{a}	0.42(0.035)^{a}
		P	513(53)^{a}	484(25)^{a}	849(85)^{b}
		K	4487(605)^{ab}	3044(192)^{a}	6876(1894)^{b}
		S	981(92)^{a}	815(29)^{a}	994(74)^{a}
		Mg	1166(180)^{a}	803(84)^{a}	1516(206)^{a}
	Micro-elements	**B**	51(4.4)^{a}	45(4.0)^{a}	57(11)^{a}
		Mn	5.6(0.70)^{a}	10(2.4)^{a}	14(5.5)^{a}
		Fe	35(2.9)^{b}	25(1.8)^{a}	33(6.3)^{ab}
		Cu	15(1.4)^{a}	14(1.0)^{a}	14(0.80)^{a}
		Zn	4.0(0.47)^{a}	3.7(0.44)^{a}	8.2(1.1)^{b}
		Mo	0.056(0.011)^{a}	0.063(0.016)^{a}	0.033(<0.01)^{a}

Phosphorus and K in endosperm samples ranged from 3125-3827 mg kg^{-1} and 4870-5045 mg kg^{-1}, respectively, within concentration ranges reported in Nigerian endosperms: 2600-4700 and 2770-6600 mg kg^{-1}, respectively (Kok et al., 2011). These concentrations were lower than those in Malaysian endosperms: 6520-6950 and 6930-7510 mg kg^{-1}, respectively (Akpanabiatu et al., 2001). The concentrations of Mn in endosperm samples (136-285 mg kg^{-1}) overlapped with the range reported in Malaysian endosperms (82-145 mg kg^{-1}) (Kok et al., 2011) but were lower than Mn concentrations reported in Nigerian samples of 410-610 mg kg^{-1} (Akpanabiatu et al., 2001). Our Cu and Zn concentrations in endosperms tissue (16-17 and 24-27 mg kg^{-1}, respectively) were similar to the ranges present in Malaysian endosperms of 16-18 and 25-36 mg kg^{-1}, respectively (Kok et al., 2011) and lower than those of Nigerian endosperm tissue (Akpanabiatu et al., 2001). Magnesium and Fe concentrations in our analysed samples were lower than those from Malaysia (2050-3060 mg kg^{-1} Mg and 43-52 mg kg^{-1} Fe) (Akpanabiatu et al., 2001) and Nigeria (2250-5540 mg kg^{-1} Mg and 110-220 mg kg^{-1} Fe) (Kok et al., 2011). No data could be identified for comparison on concentrations of C, N, S, B or Mo in *E. guineensis* endosperm tissue. There were only two significant differences in concentrations of essential elements in endosperm tissues between sites: Site A was significantly higher in B than site B, while Site B was significantly higher in Mo than Site C.

The low concentrations in mesocarp tissues (Table 4.3) are typical of *E. guineensis*: as fruit ripens, essential elements in the mesocarp decrease due to dilution by growth or translocation into the endosperm (Corley & Tinker, 2015). Our samples were ripe when collected, and the concentration ranges of N, P, K and Mg — 0.29-0.42 %, 484-849 mg kg^{-1}, 3044-6876 mg kg^{-1}, 803-1516 mg kg^{-1}, respectively — are comparable to those of ripe mesocarp: 0.33-0.41%, 440-530 mg kg^{-1}, 2900-3700 mg kg^{-1}, 1300-1500 mg kg^{-1}, respectively (Mathews et al., 2004). Of the essential elements analysed, N, P, Mg, S Mn, Cu, Zn, and Mo were significantly higher in endosperms compared to mesocarps.

4.3.2.2 Non-essential trace elements

Excluding Na, Al, Si, Cr, Ni and Sr, all non-essential elements in *E. guineensis* endosperms and mesocarps were <1.0 mg kg^{-1} (Table 4.4). Non-essential elements posed no toxicity risks for manufactured products. As palm oil is expressed or extracted from the mesocarp, the low concentrations in this plant material indicates that TE concentrations in palm oil are likely to be low and of no concern regarding food safety standards, unless it is contaminated during the extraction process. As with leaf concentrations, Cs was significantly higher in mesocarps from Site C. Other than this, there were no significant differences in mesocarps between sites. Low elemental concentrations are typical of endosperm tissue, as to reach this location of the plant elements must be carried by phloem transport and cross the placenta (Singh et al., 2020). As with leaves, Ni was significantly lower in mesocarp tissue from Site C site compared with Sites A and B. Chromium was also significantly higher in mesocarps from Site A compared to Site B.

Mean Na in the endosperm samples ranged from 112–223 mg kg^{-1}, within the range of 80-240 mg kg^{-1} from Malaysian kernels (Kok et al., 2011) and below the range of 610-1200 mg kg^{-1} from Nigerian kernels (Akpanabiatu et al., 2001). Cadmium in our endosperm samples (0.02-0.03 mg kg^{-1}) was lower than in samples from Malaysia (0.09-0.31 mg kg^{-1}) (Aini Azura et al., 2012), while Pb (0.02-0.03 mg kg^{-1}) was comparable to the below-detection concentrations of ≤0.05 mg kg^{-1} reported in Nigerian kernels (Akpanabiatu et al., 2001).

Table 4.4. Mean concentrations of non-essential elements in analysed *E. guineensis* endosperms and mesocarps from each site (standard error of the mean in parentheses). All values in mg kg^{-1}. Sites that share the same letter(s) for a single variable are not significantly different from each other.

Site		**A (n=8)**	**B (n=7)**	**C (n=3)**
Endosperm	**Na**	223(33)^{a}	128(15)^{b}	112(24)^{ab}
	Al	31(5.2)^{a}	17(2.9)^{a}	27(4.9)^{a}
	Si	352(38)^{a}	303(38)^{a}	226(22)^{a}
	Ti	0.33(0.016)^{a}	0.39(0.051)^{a}	0.30(<0.01)^{a}
	Cr	0.29(0.018)^{b}	0.19(0.027)^{a}	0.24(0.064)^{ab}
	Co	0.013(<0.01)^{a}	0.014(<0.01)^{a}	0.010(<0.01)^{a}
	Ni	1.6(0.17)^{b}	2.2(0.35)^{b}	0.60(0.13)^{a}
	As	<0.01^{a}	<0.01^{a}	<0.01^{a}
	Sr	5.0(0.59)^{a}	4.6(0.32)^{a}	3.9(0.44)^{a}
	Ag	<0.01^{a}	<0.01^{a}	<0.01^{a}
	Cd	0.018(<0.01)^{a}	0.027(<0.01)^{a}	0.017(<0.01)^{a}
	Te	<0.01^{a}	<0.01^{a}	<0.01^{a}
	Cs	0.068(0.015)^{a}	0.094(0.023)^{a}	0.13(0.022)^{a}
	Ce	0.16(0.028)^{a}	0.080(0.014)^{a}	0.14(0.023)^{a}
	Au	<0.01^{a}	<0.01^{a}	<0.01^{a}
	Hg	<0.01^{a}	<0.01^{a}	<0.01^{a}
	Pb	0.031(<0.01)^{a}	0.021(<0.01)^{a}	0.027(<0.01)^{a}
Mesocarp	**Na**	213(29)^{a}	164(9.4)^{a}	180(27)^{a}
	Al	35(6.3)^{a}	28(3.1)^{a}	31(8.6)^{a}
	Si	328(22)^{a}	320(12)^{a}	313(38)^{a}
	Ti	0.50(0.17)^{a}	0.40(0.044)^{a}	0.30(0.12)^{a}
	Cr	3.8(0.28)^{a}	2.6(0.39)^{a}	4.0(1.0)^{a}
	Co	0.070(0.0050)^{a}	0.066(0.010)^{a}	0.06(0.012)^{a}
	Ni	2.1(0.15)^{a}	1.6(0.18)^{a}	2(0.36)^{a}
	As	<0.01^{a}	<0.01^{a}	<0.01^{a}
	Sr	4.5(0.64)^{a}	4.4(0.47)^{a}	6.3(1.3)^{a}
	Ag	<0.01^{a}	<0.01^{a}	0.010(<0.01)^{a}
	Cd	0.014(0.003)^{a}	0.011(0.003)^{a}	0.010(<0.01)^{a}
	Te	<0.01^{a}	<0.01^{a}	<0.01^{a}
	Cs	0.24(0.074)^{a}	0.28(0.071)^{a}	0.73(0.082)^{b}
	Ce	0.13(0.014)^{a}	0.11(0.014)^{a}	0.13(0.040)^{a}
	Au	<0.01^{a}	<0.01^{a}	<0.01^{a}
	Hg	<0.01^{a}	<0.01^{a}	<0.01^{a}
	Pb	0.053(0.014)^{a}	0.047(0.011)^{a}	0.060(0.015)^{a}

4.3.3 Bioaccumulation coefficients

Most BACs for TEs in *E. guineensis* at the sites sampled were <0.5 (Table 4.5). A BAC of ≥1 indicates that a plant may take up higher than typical concentrations of a TE from soil (Koleli et al., 2015). These results are consistent with low BACs of most non-essential TEs measured in other studies (Aladesanmi et al., 2019; Reiser et al., 2014) except for Sr, for which no BACs could be identified from literature.

Table 4.5. Bioaccumulation coefficients of TEs in plant tissues at each if the *E. guineensis* plantation sites sampled. Coefficients for As in endosperm and mesocarp tissues are presented as < their calculated value as As concentrations in these fruit tissues was <0.01.

	Site A			**Site B**			**Site C**		
	Pinnae	**Endosperm**	**Mesocarp**	**Pinnae**	**Endosperm**	**Mesocarp**	**Pinnae**	**Endosperm**	**Mesocarp**
Cu	0.31	0.89	0.79	0.16	0.42	0.37	1.4	3.0	2.5
Zn	0.27	0.26	0.044	0.26	0.30	0.041	0.31	0.35	0.11
As	0.0079	<0.0026	<0.0026	0.0072	<0.0026	<0.0023	0.0027	<0.0011	<0.0011
Sr	0.67	0.21	0.19	0.69	0.33	0.31	0.35	0.81	0.13
Cd	0.19	0.13	0.10	0.26	0.31	0.13	0.24	0.20	0.12
Pb	0.040	0.0019	0.0033	0.045	0.0012	0.0028	0.022	0.0010	0.0021

The BAC of Cu in all plant tissues at Site C was >1, indicating *E. guineensi*s here contained high concentrations of Cu relative to the soils at this site. This may have been due to direct application of Cu-containing spray residues onto plant tissues, as this site was still under active management, however, this is unlikely as endosperm and mesocarp tissue had higher BACs relative to pinnae. Leaf tissues typically receive foliar spray applications while internal fruit tissues do not, therefore if this were the reason for the high BACs at this site, it is likely that endosperm and mesocarp BACs would be lower than that of pinnae tissues. While it is possible that Cu may be translocated from the leaf to the fruit following direct contamination of leaves with Cu spray, the higher BACs of fruit tissue relative to leaf tissue indicate that Cu was likely entering plants via root uptake from soil solution and was highly phloem-mobile. This is supported by the BACs for Cu in fruit tissues at Site A (0.89 and 0.79 for endosperm and mesocarp, respectively), which were higher than the pinnae BAC at this site. The high BAC of Cu in *E. guineensis* fruit tissues may result in high Cu concentrations in plant products, including oil and PKE, if production soils contain excessive Cu concentrations. This may present risks to human and animal health if food and fodder safety standards, respectively, for Cu are exceeded.

The BAC of Sr in pinnae at Sites A and B (0.67 and 0.69, respectively) indicated that Sr was likely entering plant tissues *E. guineensis* at these sites via soils. The BAC of Sr at Site C (0.81) also indicated that Sr maybe phloem mobile and easily transported to fruit tissues. There is little published information on Sr BACs or transport in plants with which to compare these results.

As *E. guineensis* is cultivated in plantations with differing management styles, including smallholder, corporate- and government-owned (Chalil, 2008), the determination of BACs for fertiliser contaminants in *E. guineensis* grown under a range of conditions with differing agrichemical input rates may be of benefit to this production systems. This research may develop strategies to limit the transfer of contaminants to plants and products if increased fertiliser applications were used to correct nutrient deficiencies.

4.3.4 Elements in palm oil

Unlike TEs, plant oils are not transported by xylem or phloem from leaves to fruit tissue but are synthesised from sugars *in situ* (Salisbury & Ross, 1992). Most TEs occur as hydrophilic metal complexes in plants, partitioning away from oils in plant tissues (Ouerdane et al., 2006). As such, high TE concentrations in vegetable oils are not common. Elemental concentrations in refined palm oil were lower than food safety standards and posed no risk of elemental toxicity. However, this study did not analyse pesticide residues that may be present in oils. No elements in the oil we analysed exceeded tolerable upper limits or maximum dietary reference intakes (Table 4.6). This is consistent with other studies addressing the elemental composition of plant-derived oils, in which TEs were found either below detection limits or in concentrations ≤1.0 mg kg^{-1} (Almeida & Carpenter, 2015; Perlein et al., 2021).

The following elements were present in our analysed palm oil at concentrations comparable to or less than concentrations in palm oils from previous studies: Mg, P, K, Ca, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg and Pb (Table 4.6). Concentrations of Na and Al in our palm oil samples were higher than previously reported. The mean concentration of Na in our samples was 1198 mg kg^{-1}. This is higher than reported levels in Nigerian palm oils of 30 mg kg^{-1} (Obi et al., 2001) and 115-533 mg kg^{-1} (Nnorom et al., 2014), however, would not result in consumption beyond the tolerable upper limit of 2300 mg Na day^{-1}. The mean concentration of Al in our palm oil samples (159 mg kg^{-1}) was also higher than those previously reported in Nigerian palm oil by Obi et al (31 mg kg^{-1}) and Asemave et al. (2012) (1.9 mg kg^{-1}), however, below concentrations that would surpass its tolerable weekly intake of 1 mg kg^{-1} bw wk^{-1}, assuming an average body weight of 70 kg (Otten et al., 2006) and an average daily palm oil intake of 37 g (Zhao et al., 2021). For people weighing <41 kg, exceedance of the tolerable weekly intake may occur if they are consuming ≤37 g of palm oil per day, the average vegetable oil consumption in China – a major importer of palm oil. The high Al concentrations in palm oil are supported by the mean Al concentrations in mesocarp samples (28-35 mg kg^{-1}). Unlike most other TEs, Al may form hydrophobic organic complexes in plants that are phloem mobile, thereby partitioning into palm oil in the mesocarp (Benning et al., 2012).

Table 4.6. Mean concentrations of elements in refined palm oil (standard error of the mean in parentheses) with tolerable upper limits, where available, and concentration ranges reported in previous studies.

Element	**Mean (mg kg^{-1}) (n=10)**	**Maximum consumed quantity (mg day^{-1}) ***	**Tolerable upper limit (mg day^{-1}) ⁺**	**Total ranges reported in other studies (mg kg^{-1})**
Na	1198(155)	44	2300	30-533 ᵡˠ
Mg	3.1(0.41)	0.12	350	0.02-192 ꜖ᵡˠ≭
Al	159(8.5)	0.59 (mg kg^{-1} bw wk^{-1}) †	1 (mg kg^{-1} bw wk^{-1}) †	1.9-31 ᵡ⊦
P	3.5(0.077)	0.13	4000	8-47 ǁ
S	60(3.9)	2.2	—	—
K	155(11)	5.8	4500-4700 ‡	0.39-165 ˠ≭
Ca	51(2.8)	1.9	2500	0.34-867 ꜖ᵡˠ≭
Cr	0.080(0.019)	<0.01	0.02-0.04 ‡	0.02-2.3 ˠ⊦ᴨ
Mn	0.25(0.049)	<0.01	11	0.24-12 ᵡˠ⊥
Fe	2.6(1.3)	0.096	45	0.12-232 ꜖ǁˠ≭⊦⊥
Co	0.019(<0.01)	<0.01	—	0.00-0.06 ⊥
Ni	0.031(0.010)	<0.01	1	0.00-0.81 ˠᴨ⊥
Cu	0.048(0.018)	<0.01	10	0.00-2.1 ꜖ǁᵡˠ⊦⊥
Zn	<0.01	<0.01	40	0.05-15 ˠ≭⊥
As	0.014(<0.01)	<0.01	—	≤0.03 ᴨꟻǀ
Sr	0.22(<0.01)	<0.01	—	—
Zr	1.4(0.096)	0.052	—	—
Mo	0.021(<0.01)	<0.01	2	—
Ag	<0.01	<0.01	—	—
Cd	0.015(<0.01)	<0.0001 (mg kg^{-1} bw wk^{-1}) †	0.007 (mg kg^{-1} bw wk^{-1}) †	0.02-0.09 ˠ⊦ᴨ
Te	0.010(<0.01)	<0.01	—	—
Au	<0.01	<0.01	—	—
Hg	<0.01	<0.0001 (mg kg^{-1} bw wk^{-1}) †	0.0016 (mg kg^{-1} bw wk^{-1}) †	0.00-0.06 ᴨ
Pb	0.075(0.012)	<0.0001 (mg kg^{-1} bw wk^{-1}) †	0.025 (mg kg^{-1} bw wk^{-1}) †	≤0.07 ꜖ǁˠ⊦ᴨ

* Calculated amount of element that is likely to be consumed per day in palm oil, based upon an average of 37.1 g day^{-1} vegetable oil consumed (Zhao et al., 2021), assuming palm oil is the primary vegetable oil in the diet. This is the average daily consumption of vegetable oil in China, one of the world's largest palm oil importers.

⁺ Values for males and females 19-70 years of age (Otten et al., 2006). Refers to the amount that can be consumed daily by most individuals with no adverse health effects.

† Values for Al, Cd, Hg and Pb are in mg kg^{-1} bw week^{-1} due to these elements having a specified tolerable weekly intake rather than a tolerable upper limit (Aguilar et al., 2008; FAO/WHO, 2000, 2004). The calculated maximum consumed quantity assumes an average body weight of 70 kg. Persons under 41 kg may be at risk of consuming above the maximum weekly intake of Al if consuming ≤37.1 g of palm oil per day.

‡ Recommended dietary allowance as no tolerable upper limit could be identified. Refers to the daily intake that is sufficient for meeting nutrient requirements in 97-98% of the general population (Otten et al., 2006). Applies to non-pregnant or lactating females and males aged 9 – >70 years.

ᵡ Obi et al. (2001)

ˠ Nnorom et al. (2014)

꜖ Szydłowska-Czerniak et al. (2013)

≭ Njoku et al. (2010)

⊦ Asemave et al. (2012)

ǁ Rossi et al. (2003)

ᴨ Adepoju-Bello et al. (2012)

⊥ Aigberua et al. (2017)

ꟻ Chen et al. (2001)

ǀ Chen et al. (2003)

4.4 Conclusions

The *E. guineensis* plant materials from North Sumatra, Indonesia, were deficient in essential elements including N, P, K and Mo, likely reducing palm oil yields from these sites. This is consistent with our hypotheses that *E. guineensis* leaves will show nutrient deficiencies, however, Mg was not deficient in leaf tissue as we had hypothesised. Leaf tissue from all sites was deficient in K, indicating that K may have been a yield-limiting factor potentially leading to the abandonment of production at two of the sites sampled. There is clear evidence that element deficiencies contribute to site abandonment. Leaf tissues showed no sign of elevated Cu, Zn, As and Pb, which falsified out hypothesis that leaves may contain elevated levels of TEs. Copper, however, had BACs >1, indicating either elevated plant uptake or surface contamination from Cu-containing agrichemicals. This may result in *E. guineensis*-based products that exceed food and fodder safety standards If plants are grown on soils with excessive Cu concentrations. Palm oil generally contained low concentrations of TEs that were not likely to pose any toxicity risks. This was consistent with our hypothesis and with plant physiology considering trace elements often occur in plants as metal complexes that partition away from the oil phase. Aluminium and Na were present in palm oil at higher concentrations than had been previously reported in other studies, however, were not in breach of food safety standards. Future work could assess the bioaccumulation coefficients of TEs in *E. guineensis* under a range of conditions to determine whether plant tissues or oils may exceed toxicity thresholds or food safety standards with large fertiliser applications. This could be achieved in a dose-response pot trial. This would inform reasonable limits for fertiliser applications and contaminants to limit the transfer of TEs to plants and products in palm oil production systems.

4.5 Statements and declarations

Conflicts of interest: The authors have no competing interests to declare that are relevant to the content of this article.

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Data availability: The data that support the findings of this study will be made openly available upon publication at www.kiwiscience.com [full URL will be generated]. Data is available from the authors upon request preceding publication.

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Chapter 5: Elemental composition of palm kernel expeller used as supplementary stock fodder

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Abstract

Palm Kernel Expeller (PKE) is a biowaste by-product of palm oil processing in Southeast Asia that is exported as stock fodder. Global production of PKE totals 11,000,000 t yr^{-1}, of which New Zealand imports 1,850,000 t yr^{-1}, worth >USD$325,000,000, most of which supports New Zealand's dairy herd of 5,500,000 cows. We aimed to determine the concentrations of the chemical elements in PKE imported into New Zealand and compare this to pasture, as well as assessing chemical concentrations against Maximum Tolerable Levels (MTLs) in stockfeed for animal health and ruminant requirements. Palm kernel expeller was analysed for a suite of essential and trace elements using a HNO_3 digestion and analysis by inductively coupled plasma-mass spectrometry. Palm kernel expeller contained statistically significantly higher concentrations of B, Mg, P, Cr, Mn, Fe, Ni, Cu and Zn than pasture. Magnesium, P and Fe exceeded MTLs in PKE, whereas Al, S, K and Cu were within 90% of their MTL. The N, P and K contained in PKE represent the equivalent of 14%, 20% and 28%, respectively, of dairy fertiliser use in New Zealand. As PKE contained 3.3 mg Cd kg P $^{-1}$, there may be potential for PKE to offset fertiliser use in dairy systems, with a low Cd source of P. There were statistically significant differences in elemental concentrations between different batches of PKE indicating that this product is not uniform. Further research of the effects of PKE on animal health is recommended. The excess elements contained in PKE may present risks or benefits to dairy farming systems and determining these would be beneficial in protecting both animal health and environmental sustainability.

Keywords

Trace elements, dairy cattle, *Elaeis guineensis*, Indonesia, New Zealand, copper, iron

5.1 Introduction

The African oil palm *(Elaeis guineensis* Jacq.) produces edible palm oil and palm kernel oil, which are expressed or solvent extracted from the mesocarp and endosperm respectively, and palm kernel expeller (PKE), a biowaste product comprising the ground endosperm after oil has been extracted. Palm kernel expeller is sold as a supplementary stockfeed globally. New Zealand is the world's largest importer of PKE, buying 1,900,000 t, or approximately 17% of global supply, in 2021 [1, 2] at a cost of

UDS$327-$453,000,000 (based on the January-April 2020 spot price range of PKE in New Zealand) [3]. This PKE is shipped from Southeast Asia to New Zealand, primarily for use on dairy farms [4]. The reuse of a biowaste such as PKE may be beneficial in terms of creating circular bio-economies that minimise waste and reuse nutrients, thereby adding value to by-products. However, the use of PKE is contentious as the production of palm oil and PKE have been linked to deforestation of virgin tropical forests and habitat destruction for endangered animals [5]. The plant and animal nutrients that PKE contains may substantially offset the requirements for fertilisers and animal supplements respectively. Conversely, the importation of non biologically-essential elements or nutrients in excess of agricultural requirements may exacerbate soil contamination and potentially endanger animal or human health [6].

The effects of any excess elements contained in PKE on livestock will depend on various factors including the proportion of animals' diets made up of PKE, as well as trace element (TE) interactions that may help or hinder their metabolism [7]. Excess dietary elements may either be absorbed by the animal or excreted through dung, urine or milk [6, 8]. Excess elements may endanger food safety or accumulate in soil. Dairy cattle in New Zealand are routinely monitored for element concentrations in liver and serum [6]. These concentrations may indicate whether these elements are absorbed and metabolised by animals, while elemental concentrations in faeces may indicate what excess elements are likely to be excreted.

While palm and palm kernel oils generally contain low levels of TEs [9], in PKE, there is a precedent for TE concentrations that may approach or exceed Maximum Tolerable Levels (MTLs) for cattle feed. Iron, Cu and As in PKE may reach 6130, 29 and 3.1 mg kg^{-1}, respectively [10-12]. Maximum tolerable levels for these TEs are 500, 40 and 4 mg kg^{-1}, respectively [13, 14]. Elevated concentrations of Cu, As and Pb have been measured in the soils where *E. guineensis* is produced likely due to the use of agrichemicals that contain TEs as impurities (including phosphate fertilisers) and active ingredients (including Cu-based fungicides) [15]. Post-harvest processing and transport, as well as contamination with other (waste) products may also contribute substantial concentrations of chemical elements [16, 17]. Previous studies measuring chemical elements in PKE have focussed either on a limited number of elements or have deficiencies including a lack of specified PKE sources and chemical methods used for analysis. A wider study that analyses PKE from multiple sources and across multiple years for a wide range of elements is needed to elucidate the chemical profile of PKE and any risks or opportunities it may present for agricultural systems.

We hypothesised that PKE may contain elevated concentrations of Fe, Cu and As. We aimed to compare the elemental profile of PKE to that of New Zealand pasture and to MTLs for cattle feed and cattle requirements. Additionally, we sought to analyse preliminary data on the Cu concentrations of dairy cattle livers to gain insight into the potential effects of PKE on liver Cu status of dairy herds. We used this data as a pilot study to determine the sample size necessary for a comprehensive study on the effects of PKE on dairy cattle liver Cu concentrations in a New Zealand context. Based on this, we have offered directions for future research.

5.2 Materials and methods

5.2.1 Palm kernel expeller collection

Grab samples from five shipments (hereafter referred to as batches) of PKE imported into New Zealand were collected for analysis. Five or six sub-samples were analysed from each batch. Batch #1 and #2 were collected from separate Canterbury farms in May 2021. Batches #3–#5 were collected from a port facility in Timaru, New Zealand, in March 2019 (#3, #4) and March 2020 (#5). Batch #4 had been screened to ≤4 mm while Batch 5 had not been screened and was in chunks >4 mm. Samples were oven dried at 60°C for 48 hours.

5.2.2 Pasture sampling

Pasture samples — 79 in total — were collected from 26 farms across the South Island of New Zealand during Spring (September—November), 2009. Locations of sample collection are detailed in Appendix E. A pasture sward was cut from each sampling location 2-3 cm above the ground's surface. Samples were rinsed with deionised water and dried until a constant weight was achieved at 60°C. Pasture samples were then ground using a Cyclotech grinder.

5.2.3 Pseudo-total element concentrations

Pseudo-total element concentrations (hereafter referred to as total concentrations) for PKE were determined using microwave digestion. Samples (0.20 ±0.01 g) were weighed into digestion tubes and 5 mL of ~69% HNO_3 added. Samples were pre-digested overnight and following this digested using EPA Method 3051A [18] at 230°C for 25 mins. A Srl Milestone ultraWAVE Single Reaction Chamber Microwave Digestion Unit was used. With every 11 samples, two reagent blanks and two standard reference materials (SRMs) [19-22] were included for quality assurance. Inductively coupled plasma-mass spectrometry (Agilent Technologies 8900) was used to determine elemental concentrations in the digestates. Recoveries of certified values in SRMs ranged from 79–156% of the published values. A list of elemental recoveries is available in Appendix E.

Pasture samples (0.5 g) were digested in 5 mL ~69% HNO_3 and diluted to 25 mL with ultrapure water (Sartorius Arium® Pro UV), before filtering through Whatman 52 filter paper. Elemental concentrations were determined using Inductively Coupled Plasma Optical Emission Spectroscopy (Varian 720 ES). Blanks and SRMs NIST [19] were also analysed for quality assurance. Recoveries were 92–105% of the published values.

5.2.4 Total carbon and nitrogen

A CN828 Elemental Analysis by Combustion C and N analyser was used to determine total C and N of PKE samples. Blanks and SRMs [23, 24] were included in analyses for quality assurance.

5.2.5 pH

Palm kernel expeller pH was measured following the method of Blakemore, Searle [25]. Five (±0.05) g of sample was weighed into a falcon tube and 25 mL of deionised water added. This 1:5 ratio was used in place of the more common 1:2.5 ratio due to PKE's high organic matter content. Samples were then stirred vigorously and left to stand overnight. The pH of each sample was measured with a Hach HQ440d multi pH probe.

5.2.6 Cattle liver data

Secondary data on elemental concentrations in dairy cattle livers was collected from a New Zealand veterinary practice for a preliminary investigation into potential effects of PKE on dairy cattle liver TE status. During routine screening of dairy herds' nutritional status, liver biopsies were collected from dairy cows by veterinary staff and TE concentrations including copper were analysed by an IANZ accredited veterinary pathology laboratory. This biopsy collection and analysis was completed as part of routine dairy herd management and was not undertaken on behalf of the authors. No ethical approval was required for this as the purpose of the biopsies was veterinary in nature. Data from 11 farms was collated by veterinary staff and passed on to the authors with the permission of farmers under the condition of farm anonymity. Between five and seven cows were sampled from each farm using convenience sampling. Liver data from a total of 58 cows was used for this analysis. Data was also provided on whether the farms fed PKE and if so at what rates, other feeds utilised and TE supplements that cows were receiving. Reported feeding rates of PKE were between 0.5–2.0 kg PKE cow^{-1} and cows were fed varying combinations of grass, grain, baleage and fodder beet. Every cow analysed from farms feeding PKE had been fed PKE, and it is unlikely that any of the cows analysed consumed any PKE from the same batches as were analysed by the authors in this study; however, this cannot be confirmed.

5.2.7 Calculation of nutrient inputs to soils through palm kernel expeller

Considering the elemental concentrations in PKE, the quantities of N, P and K imported into New Zealand through PKE were calculated. These calculations used New Zealand's 2021 import figure of 1,850,000 t PKE [26] and the average concentration of N, P and K in our five PKE batches. These imported quantities were compared to New Zealand's annual fertiliser N, P and K use in dairy systems, based upon quantities of fertiliser sold in New Zealand in 2019 (the most recent figure available) [27] and the proportion of N, P and K fertilisers in New Zealand utilised by dairy farms [28].

5.2.8 Statistical analysis

Statistical analyses were conducted using R (version 4.1.0) [29]. A one-way analysis of variance (ANOVA) was used to determine whether any statistically significant differences in mean elemental concentrations existed between batches of PKE. Where assumptions of normality were not met, data was log transformed. Where assumptions of homoscedacity were not met, a parametric permutational ANOVA was used to determine the robustness of the parametric ANOVA. The results of the parametric and non-parametric ANOVAs did not differ. Tukey's Honest Significant Difference test was implemented in multcomp package [30] to determine which batches were significantly different from each other when ANOVA identified statistically significant differences.

For each element mean concentrations in PKE and pasture were compared using repeated measures ANOVA. The packages lme4 [31] and lmerTest [32] were used for this analysis.

Repeated measures ANOVA was used to compare the average Cu contents in the livers of dairy cattle between the farms with and without PKE in their feed. This was implemented via the lme4 and lmerTest packages. A post-hoc power analysis was run on this data to determine the statistical power with the current sample size. A power curve was then produced for the number of farms ranging from 10 to 1400, with five cows per farm in order to determine the minimum number of farms required to detect the observed effect at 5% significance level with at least 80% probability. It was then repeated for ten cows per farm. The power curve estimation was based on 10^3 simulations for each setting.

5.3 Results and discussion

5.3.1 Elements in palm kernel expeller

5.3.1.1 Essential plant or animal nutrients

5.3.1.1.1 Macronutrients

Nitrogen is commonly used as an indicator for protein (not measured here) in food studies [33]. Mean N concentrations in PKE ranged from 2.0-2.2%. This is lower than concentrations measured in previous studies (Table 5.1). Nitrogen was statistically significantly higher in PKE (2.0-2.2%) than in *E. guineensis* endosperms (1.3–1.4%) [9]. Palm oils contain only 0.8% of the N contained in a whole fruit bunch (the fruit bunch cut from the tree containing whole *E. guineensis* fruitlets as well as the surrounding plant material) [34] meaning that the N concentrations in PKE are likely to reflect ca. 99% of N present in endosperms. It is unlikely that higher N concentrations in PKE are a result of contamination and therefore it is possible that the PKE we analysed was manufactured from plant material with higher N concentrations than have been reported by Thompson-Morrison et al. [9], Corley and Tinker [35], Woittiez, Slingerland [36], Rafflegeau, Michel-Dounias [37], Law, Zaharah [38] and Ng et al., as cited in Corley and Tinker [35], all of whom reported N deficiency in *E. guineensis* plant material. Alternatively, the higher N in PKE may result from the post-harvest mixing with other high N material, as reported in fish meal and soybean meal [17, 39, 40].

Mean P in batches of PKE ranged from 5229-7265 mg kg^{-1} (Table 5.1). This concentration range is comparable to the total range previously reported in Malaysian PKE of 4800-7900 mg kg^{-1} [10, 11, 41]. The mean concentrations of K in the analysed PKE ranged from 5834–18,927 mg kg^{-1}. A previous study found Malaysian PKE contained 7600-9300 mg kg^{-1} K [11], while PKE used in the United Kingdom contained 6900 mg kg^{-1} K [13]. Mean Mg in PKE ranged from 2723–6209 mg kg^{-1} (Table 5.1). All batches exceeded concentrations of Mg in Malaysian PKE from Yeong [10] of 2700 mg kg^{-1}, and Batches #1 and #2 exceeded the range of 1600–3300 mg kg^{-1} in Malaysian PKE reported by Alimon [11]. Sulphur in PKE ranged from 1916-2745 mg kg^{-1}, which was comparable to previously reported concentrations (Table 5.1).

Table 5.1. Mean essential elemental composition of the five PKE batches analysed (standard error of the mean in parentheses), with mean pasture concentrations, MTLs for cattle feed and cattle requirements for comparison. Elements that are within 50% of the MTL in one or more batches in PKE are marked with *, elements that exceed their MTL in one or more batches of PKE are marked with **. Batches that share the same letter(s) for a single variable are not statistically significantly different from each other. All values are mg kg^{-1} dry weight unless otherwise specified.

PKE Batch	1 (n=5)	2 (n=5) ⁺	3 (n=6) ǂ	4 (n=6) ǂ	5 (n=6) ǂ	Concentrations reported from other sources	Pasture concentration	Cattle feed MTL ^	Cattle requirements ‡
C (%)	47(0.45)^{a}	49(0.81)^{b}	49(0.36)^{b}	48(0.31)^{ab}	47(0.26)^{ab}	—	43 ᵡ	—	—
N (%)	2.2(0.09)^{a}	2.1(0.11)^{a}	2.0(0.12)^{a}	2.2(0.03)^{a}	2.1(0.02)^{a}	2.2 ҂ 2.9 ⊥	2.0ᵡ	—	—
P **	5251(107)^{a}	5229(90)^{a}	5923(100)^{ab}	7265(160)^{c}	6325(330)^{b}	6500 ˫ǁ 7900 ɤ 4800-7100 ǁ	3404(148)	6000	3100
K *	17,034(950)^{c}	18,927(496)^{c}	6065(45)^{ab}	6757(187)^{b}	5834(243)^{a}	8000 ˫ǁ 7600-9300 ǁ	22,428(1265)	20,000	10000
S *	2038(44)^{ab}	1916(37)^{a}	2148(27)^{ab}	2745(92)^{c}	2325(136)^{b}	1900-2300 ǁ	2538(105)	3000 (high-concentrate) 5000 (high-forage)	2000
Mg **	5948(277)^{b}	6209(146)^{b}	2826(24)^{a}	3083(128)^{a}	2723(107)^{a}	3000 ˫ǁ 2700 ɤ 1600-3300 ǁ	998(48)	6000	1400
B	18(3.5)^{a}	20(2)^{a}	24(1.4)^{a}	21(1.9)^{a}	20(2.6)^{a}	—	6.4(0.41)	150	—
Na	280(71)^{b}	63(35)^{a}	145(17)^{ab}	131(18)^{a}	126(14)^{a}	200 ˫	2226(282)	—	1200
Cr	4.8(0.2)^{a}	5.5(3.1)^{a}	2.5(0.1)^{a}	4.0(0.58)^{a}	2.3(0.49)^{a}	—	0.90(0.13)	100	—
Mn	317(7.6)^{a}	298(9.3)^{a}	271(0.92)^{a}	468(40)^{b}	302(31)^{a}	225 ɤ 132-340 ǁ	119(10)	2000	25
Fe **	2406(174)^{c}	786(29)^{b}	624(49)^{ab}	624(28)^{ab}	447(107)^{a}	4.1 ɤ 835-6130 ǁ	433(97)	500	40
Co	0.68(0.07)^{c}	0.1(0)^{ab}	0.07(0.02)^{ab}	0.13(0.02)^{b}	0.03(0.02)^{a}	—	0.93(0.16)	25	0.06
Cu *	28(0.43)^{b}	29(0.61)^{b}	22(0.32)^{a}	36(1.1)^{c}	27(1.8)^{b}	22 ˫ǁ 29 ɤ 21-29 ǁ	6.9(0.28)	40 ˥	9–11
Zn	54(0.65)^{bc}	47(1.2)^{ab}	44(0.53)^{a}	59(1.4)^{c}	49(3.1)^{ab}	77 ɤ 41-50 ǁ	27(1.5)	500	25
Mo	0.42(0.04)^{a}	0.42(0.02)^{a}	0.33(0.02)^{a}	0.35(0.03)^{a}	0.43(0.03)^{a}	0.70-0.79 ǁ	0.43(0.061)	5	—

⁺ Na n=2. ǂ C, N n=5. ^ [13]. Although PKE can be used as a stockfeed for animals including sheep, goats and poultry, MTLs specific to cows are used where available as the prevailing use for PKE as a stockfeed is in New Zealand dairy farms [1, 4]. ‡ [42]. ᵡ [43]. ҂ [44]. ⊥ [45]. ˫ [41]. ǁ [11]. ɤ [10]. ˥ Based on Mo 1–2 mg kg^{-1} and S 1500–2500 mg kg^{-1}, Cu may become toxic at lower doses if Mo and S are below these values.

5.3.1.1.2 Micronutrients

The mean Fe concentrations in PKE (447–2406 mg kg^{-1}) were of comparable ranges in Malaysian PKE of 835–6130 mg kg^{-1} [11]. The Fe present in PKE is likely a result of processing or shipping that occurs post-harvest, and not a result of plant uptake. This is corroborated by the lower Fe concentrations reported in *E. guineensis* plant tissues by Thompson-Morrison et al. [9], and the recognition that production and storage processes are potential sources of contamination — particularly heavy metals — for *E. guineensis* products [16]. Metal contamination of PKE has been a known issue, evidenced by regulations for screening and magnet-scanning of PKE imported into New Zealand [46].

The mean Cu in PKE ranged from 22-36 mg kg^{-1}. This concentration range is higher than has been previously reported in PKE (Table 5.1). Copper may enter PKE as a result of Cu-fungicide use in *E. guineensis* plantations where it is applied directly to soil and plants [47]. It can then be absorbed by plants and transported to fruit tissues [48].

The concentrations of Mn (271–468 mg kg^{-1}) and Zn (44–59 mg kg^{-1}) in the PKE we analysed were higher than ranges previously reported from Malaysian PKE (Table 5.1). Concentrations of Mo in PKE were less than those previously reported, which is consistent with Mo deficiencies in the soils and leaves of *E. guineensis* [9, 15].

5.3.1.2 Non-essential elements

Concentrations of Ag, Cd, Sb, Te, Au and Hg in PKE were ≤0.03 mg kg^{-1} and unlikely to pose toxicity risks (Table 5.2). Concentrations of TEs including Ti, As, Sr and Pb were higher in PKE (11–179, 0.20–0.52, 11–26 and 0.27–1.2 mg kg^{-1}, respectively) compared to *E. guineensis* endosperms (0.30–0.39, <0.01, 3.9–5.0 and 0.021–0.031 mg kg^{-1}, respectively) — the plant material PKE is produced from [9]. These TEs may have entered PKE through pathways other than plant uptake, including contamination during processing and shipping.

All batches contained higher concentrations of Al than Malaysian PKE (sourced in New Zealand in a previous study), which measured 178 mg kg^{-1} [49]. The range of As present in our analysed PKE — 0.20–0.52 mg kg^{-1} — was within the range of As in Malaysian PKE of 0.18–3.05 mg kg^{-1} [12]. Some of the As levels reported by Hammid, Kuntom [12] were within 75% of the MTL for As in cattle feed (4 mg kg^{-1}), and thus this element has been identified as an element of concern in PKE [50]. The As present in the PKE we have analysed indicates that concentrations in PKE imported into New Zealand are low and not of concern, which falsifies our hypothesis that PKE would contain elevated As concentrations. Given the large variation in sample TE concentrations, it is possible that some batches will contain unacceptable As concentrations.

Table 5.2. Mean non-essential elemental composition of the five PKE batches analysed (standard error of the mean in parentheses), with mean pasture concentrations and MTLs for cattle feed for comparison. Elements that exceed their MTL in one or more batches of PKE are marked with **. Batches that share the same letter(s) for a single variable are not statistically significantly different from each other. All values are mg kg^{-1} dry weight unless otherwise specified.

Batch	**1 (n=5)**	**2 (n=5)**	**3 (n=6) +**	**4 (n=6) +**	**5 (n=6) ‡**	**Concentrations reported from other sources**	**Pasture concentration**	**Cattle feed MTL**
Al **	2767(97)^{c}	867(211)^{b}	420(88)^{ab}	226(34)^{a}	257(82)^{a}	178 ^	469(127)	1000 ‡
Ti	179(13)^{e}	25(0.95)^{d}	19(1.5)^{c}	15(0.38)^{b}	11(0.51)^{a}	—	—	—
Ni	2.3(0.05)^{ab}	1.8(0.2)^{a}	1.4(0.05)^{a}	4.2(0.77)^{b}	1.6(0.16)^{a}	—	0.77(0.070)	100 ‡
As	0.52(0.05)^{b}	0.31(0.04)^{ab}	0.29(0.02)^{a}	0.20(0.04)^{a}	0.24(0.07)^{a}	0.18-3.1 χ	0.41(0.030)	30 ‡ 4 ⊥
Sr	26(0.94)^{b}	12(0.3)^{a}	11(0.41)^{a}	21(2.7)^{b}	11(0.5)^{a}	—	—	2000 ‡
Zr	0.49(0.06)^{c}	0.025(0.02)^{a}	0.13(0.02)^{b}	0.17(0.01)^{b}	0.12(0.02)^{ab}	—	—	—
Ag	<0.01^{a}	<0.01^{a}	<0.01^{a}	<0.01^{a}	<0.01^{a}	—	—	—
Cd	0.024(0.0024)^{bc}	<0.01^{a}	0.023(0.0021)^{bc}	0.027(0.0021)^{c}	0.017(0.0021)^{ab}	—	0.14(0.014)	10 ‡ 1 ⊥
Sb	0.018(0.0012)^{a}	0.021(0.0028)^{a}	0.021(0.0030)^{a}	0.022(0.0039)^{a}	0.014(0.0021)^{a}	—	—	—
Te	0.014(0.0012)^{c}	<0.01^{b}	<0.01^{ab}	<0.01^{a}	<0.01^{a}	—	—	—
Cs	1.1(0.037)^{d}	0.80(0.032)^{c}	0.43(0.021)^{b}	0.22(0.060)^{a}	0.35(0.022)^{ab}	—	—	—
Ce	3.7(0.28)^{b}	0.24(0.024)^{a}	0.22(0.060)^{a}	0.15(0.022)^{a}	0.10(0)^{a}	—	—	—
Au	<0.01^{b}	<0.01^{b}	<0.01^{a}	<0.01^{a}	<0.01^{a}	—	—	—
Hg	<0.01^{a}	<0.01^{a}	<0.01^{a}	<0.01^{a}	<0.01^{a}	—	—	2 ‡ 0.1 ⊥
Pb	1.2(0.084)^{b}	0.35(0.024)^{a}	0.44(0.10)^{a}	0.30(0.058)^{a}	0.27(0.067)^{a}	—	0.37(0.054)	100 ‡ 10 ⊥

+ C, N n=5. ‡ Ag, C, N n=5. ^ [49]. ‡ [13]. χ [12]. ⊥ [14].

5.3.2 Implications for stock fodder

Compared to pasture, PKE contained higher concentrations of essential elements P, Cr, Mg, B, Mn, Fe, Cu and Zn. Concentrations Mg, Al, P and Fe in PKE exceeded the MTL for cattle feed in one or more batches, while S, K and Cu were within 90% of MTLs in one or more batches (Table 5.1). Except for Al, these elements are essential for livestock and thus their high concentrations in PKE may represent a potential benefit if animals are deficient in these elements or grazed on deficient soils, or conversely a risk if their concentrations are high enough to induce toxicity in animals. All other elements analysed were either below MTLs for cattle feed, or values for MTLs or cattle requirements could not be identified. If elements were within 50% of their MTL they were noted and discussed, as this is a benchmark commonly used in environmental monitoring [51]. Effects of TEs contained in PKE on animal health and nutrition will be dependent on the diet-proportionality of PKE, which is determined on-farm.

Phosphorus exceeded the MTL for cattle feed of 6000 mg kg^{-1} in Batches #4 and #5 at 7265 and 6325 mg kg^{-1} respectively and was within 87% the MTL in Batches #1–#3 (5229–5923 mg kg^{-1}). Exceedance of this MTL has also been previously reported in Malaysian PKE (Table 5.1). Effects of excess P in the diet depend on its bioavailability and solubility, and the concentration of other elements including Ca. In cows, the majority of dietary excess P is excreted once nutrient requirements have been met and no adverse health outcomes or effects on milk production have been observed as a result of excessive P in the diet [8]. Phosphorus is excreted via milk as well as urine and faeces [52], so excess P through PKE may affect milk composition. The factors governing P excretion via milk are not fully understood and may vary between cows [52]. Excess P can adversely interact with Mg, and Ca: P ratios <1:1 in feed are likely to affect animal health and performance regardless of whether P concentrations are below the MTL [53]. A Ca: P ratio of between 1:1 and 6:1 is tolerated in cattle, and if this is well-managed, P concentrations in feedstock above the MTL can be tolerated [53]. Both Ca and P have been flagged by the National Research Council as being of concern for toxicity in livestock. The excretion of excess P may present a substantial input of nutrients to grazed soils in addition to calculated fertiliser inputs, increasing the risk of eutrophication of surface water bodies through runoff of topsoil and nutrients. On-farm nutrient balance calculations that consider inputs through PKE may be beneficial in determining and mitigating this risk.

Batches #1 and #2 contained 17,034 and 18,927 mg kg^{-1} of K, respectively, which were within 85% of the MTL of 20,000 mg kg^{-1}. All PKE contained lower concentrations of K than pasture. Potassium has been flagged by the National Research Council [13] as being of potential concern for toxicity and excess K, while usually excreted in urine [54], can interfere with absorption of Ca and Mg [13].

Magnesium exceeded the MTL for cattle feed of 6000 mg kg^{-1} in Batch #2 (6209 mg kg^{-1}) and was within 99% the MTL in Batch #1 (5928 mg kg^{-1}). All Batches exceeded cattle requirement concentrations for Mg of 1400 mg kg^{-1}. The excess Mg in PKE may be beneficial in the prevention of milk fever, as Mg supplementation is often used for this purpose [55]. This benefit may be realised during pre- and post-calving when Mg supplements are traditionally used. While excess dietary Mg is excreted in urine in dairy cows, Mg-rich diets may lead to an increase in phosphatic calculi (kidney stones) in animals if the diet is also high in P [13]. As PKE contains higher concentrations of both Mg and P than pasture, monitoring dairy herds for signs of kidney stones may be beneficial when feeding high rates of PKE.

Concentrations of S approached the MTL for high-concentrate diets (3000 mg kg^{-1}) in all batches of PKE, which ranged from 1916-2745 mg kg^{-1}. Cattle requirements (2000 mg kg^{-1}) were exceeded in every batch except Batch #2. For cattle fed a high-forage diet, the MTL for S is 5000 mg kg^{-1} and thus any risk associated with S in PKE will depend on the individual feeding patterns of farms [13]. Despite higher MTLs, adverse effects on growth performance may be possible when cattle are fed diets with >2000 mg kg^{-1} S [53]. Our results exceeded the previously reported range of S in Malaysian PKE, 1900-2300 mg kg^{-1} [11].

Iron exceeded the MTL of 500 mg kg^{-1} in Batches #1–4 (624–2406 mg kg^{-1}) and was within 89% of the MTL in Batch #5 (447 mg kg^{-1}). Chronic Fe toxicity in mammals causes damage to intestinal tract and liver and delays in blood clot formation [56]. Excessive Fe intake in bovines can reduce milk production and decrease body weight with <30 mg kg^{-1} day^{-1} estimated as the minimum harmful dose [57]. Cattle deaths due to suspected Fe toxicity have occurred when fed feedstuffs containing 1992 mg kg^{-1} Fe [58]. The effects of excessive Fe intake will of course depend on the solubility of and bioavailability of Fe in the diet [42].

Mean Cu in PKE was within 55-90% of the MTL (40 mg kg^{-1}) in all five batches. Copper may become toxic at lower levels than MTLs suggest if Mo and S are not present in the diet at concentrations of 1-2 and 1500-2500 mg kg^{-1}, respectively [13]. It has been suggested that excess TEs in PKE such as Cu may present a potential benefit to livestock in countries where soils are deficient in essential nutrients, such as New Zealand [50]. The plausibility of this depends heavily on TE interactions and possible antagonism with respect to elements found in PKE. Effects of excess dietary Cu will be affected by the S, Mn, Fe, Zn and Mo (all Cu antagonists) concentrations in animal feed [11, 13, 42]. For example, intake of dry matter containing 500-800 mg kg^{-1} Fe has been shown to cause depressed Cu concentrations in serum and livers of dairy cattle due to interference with Cu metabolism [59, 60]. Thus, benefits that may be provided through the addition of Cu to animals' diets through PKE may be negated by the antagonistic effects of Fe also present in PKE. Investigating this would be beneficial, as in New Zealand Cu supplements for livestock are often reduced when feeding PKE [41]. Alimon [11] noted that animals may develop symptoms of Cu-toxicity when their diets contain >50% PKE. Mortality rates in sheep fed PKE were 100% when their diet was not supplemented with Zn [61]. This was attributed to Cu-toxicity. Appropriate management of elements in animals' diets is essential if using PKE as a stockfeed and future studies should address the digestibility/bioavailability of elements in PKE, and interactions that occur between these including K and Mg, Mg and P, and Fe and Cu.

Aluminium was the only non-essential element in PKE that exceeded MTLs. The mean Al concentration of Batch #1 (2767 mg kg^{-1}) exceeded the MTL of 1000 mg kg^{-1}, while Batch #2 was within 86% of the MTL at 867 mg kg^{-1}. No cases of Al toxicity in cattle as a result of Al in feedstock could be identified, although Al is identified as a potentially toxic element by the National Research Council [13]. A risk assessment of the excess Al and other elements contained in PKE and their potential effects on animal health is recommended.

Batches contained statistically significantly different concentrations of both essential and non-essential elements, and thus the non-uniformity of PKE should be factored into decisions surrounding its use where animal mineral nutrition is concerned, as consistent levels of elements in each batch cannot be relied upon. Statistically significant differences existed between the pH of different batches of PKE (5.4–5.5). This may be a result of variation in production conditions, including fertiliser and pesticide applications in *E. guineensis* plantations [62]. Palm kernel expeller contained statistically significantly higher concentrations ($p < 0.05$) of B, Mg, P, Cr, Mn, Fe, Ni, Cu and Zn compared to pasture and thus may be a dietary source of these elements. Concentrations of Na, K, Co and Cd were statistically significantly lower ($p < 0.05$) in PKE than in pasture, while concentrations of Al, S, As, Mo and Pb were comparable between PKE and pasture (Table 5.1). Palm kernel expeller represents a substantial source of TEs for farm systems utilising this by-product.

In addition to effects on animal health and performance, excess dietary elements may affect the composition of meat and milk products, as well as elemental concentrations of grazed soils [8, 53]. This may have implications for the use of mineral supplements for livestock. On-farm mass-balance calculations that take into account element inputs through PKE would be beneficial in understanding the tangible and long-lasting impacts the use of PKE may have on importing agricultural systems. However, the variation in concentrations of essential nutrients (Mg, P, S, K, Cu and Zn) across PKE batches will make farm nutrient balance calculations complex.

5.3.3 Pilot study using dairy cattle liver data

The potential effects of PKE on animal health concerning TEs may be elucidated by preliminary analysis of data on the TE status of dairy cattle livers. Our analysis failed to find a statistically significant difference ($p = 0.804$) between the liver Cu concentrations of cows fed PKE and those not fed PKE using data from 11 farms in Canterbury. A weighted mean estimate for cows fed PKE was 2023 µmol kg^{-1} while cows not fed PKE had a mean estimated liver Cu concentration of 2116 µmol kg^{-1} (Figure 5.1). Both fall within the adequate range of 95-3000 µmol kg^{-1}. The variation in cows both between farms and within farms is large, and some individual cows have liver Cu concentrations above the adequate range (Figure 5.1). The lack of statistical difference in the liver chemistry between the PKE-fed cows and the non PKE-fed cows may be due, in part, to the limited sample size. Our analysis did not have sufficient statistical power to draw generalisable conclusions. A more extensive study is required to determine categorically whether PKE affects liver chemistry. A comprehensive study assessing the potential effects of PKE on liver Cu concentrations in dairy cattle, would require approximately 1310 farms with five cows sampled each to achieve statistical power — the probability that an existing effect will be detected — of 0.8 at 5% statistical significance level. (Figure 5.2). This would represent data from approximately 12% of New Zealand’s dairy herd. Testing a larger number of cows from each farm would slightly decrease the number of farms needed: if testing ten cows from each farm, approximately 1260 farms would be needed (Figure 5.2). Despite the large number of farms required, this data is routinely collected from dairy herds and thus such a study may be feasible with a dedicated effort. The primary challenge would be coordination between researchers and veterinarians and the increased administration on the part of vet practices around gaining farmers’ consent for thew study as well as anonymising and sending data. Our preliminary analysis shows no clear evidence for PKE having profound effects on animal Cu status. However, future work is clearly needed to confirm this.

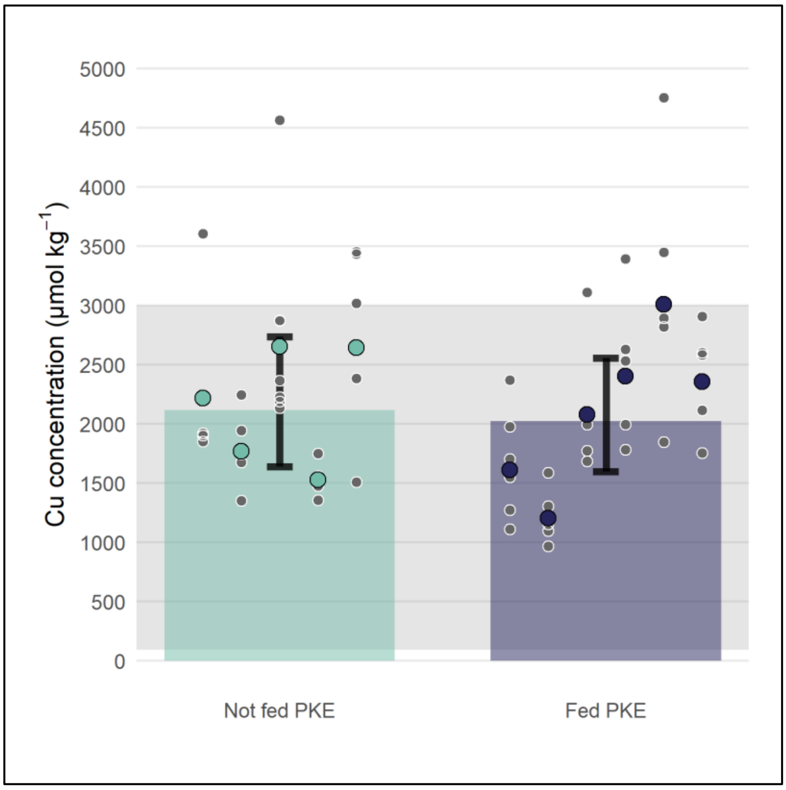

Figure 5.1. Bar graph shows weighted mean estimates for dairy herd liver Cu concentrations by feed regime. Error bars indicate 95% confidence around the mean. Shaded area represents adequate concentration range. Points stratified in each vertical line overlaid on the bar graph represent data from one of 11 farms in Canterbury. The coloured circle indicates the farm mean while the grey circles represent Cu concentrations from livers of individual cows from that farm.

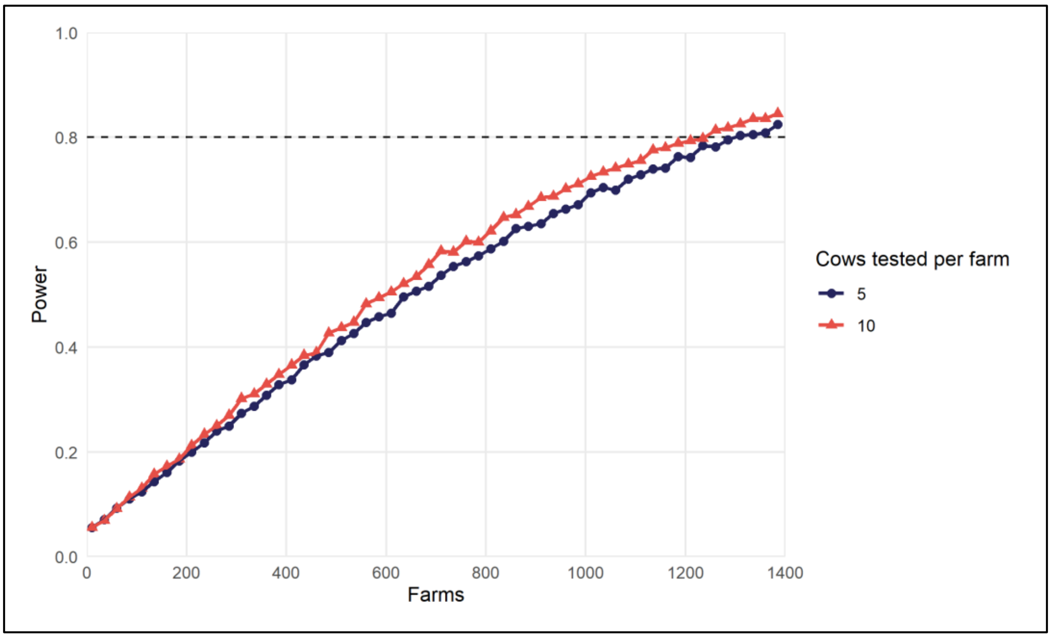

Figure 5.2. Power curve showing statistical power against number of farms needed to test effects of PKE on liver Cu concentrations, with either five or ten cows tested per farm. Commonly recognised sufficient statistical power of 80% is indicated by the dashed line.

5.3.4 Elements in palm kernel expeller as soil nutrient inputs

Given dairy systems in New Zealand use 63% of total N fertiliser, 36% of total P fertiliser and 56% of total K fertiliser [28], N, P and K inputs to dairy systems through PKE represent the equivalent of 14%, 20% and 24% of total nutrient inputs through fertilisers, respectively (Table 5.3). In comparison, the equivalent amount of N, P and K fertilisers in New Zealand based on June 2022 prices [63] and exchange rates would cost approximately USD$78,000,000 for N (as urea), USD$39M for P (as superphosphate) and USD$22,000,000 for K (as standard KCl), a total of USD$138,000,000 (Table 5.3). This represents approximately one third of New Zealand's total expenditure on PKE [3]. Thus, there may be potential to offset the use of fertiliser nutrients on dairy systems through PKE. However, the economic and environmental cost of shipping PKE between continents should be factored into any calculations of this offset.

Table 5.3. Use of N, P and K fertiliser in New Zealand dairy systems with the equivalent proportion of nutrient inputs through PKE and the equivalent cost of these nutrients as fertilisers

Element	New Zealand dairy farm fertiliser use (t yr^{-1})	Mean New Zealand imports through PKE (t yr^{-1})	Mean fertiliser equivalent imports through PKE to New Zealand dairy farms (t yr^{-1})	Fertiliser equivalent cost (USD$)
N	284,760	39,258	14%	77,879,500
P	55,440	11,097	20%	38,500,729
K	73,360	20,208	28%	21,756,711

Using an average concentration of Cd in PKE, we calculated that the Cd:P ratio in PKE is 3.3 mg Cd kg P^{-1}. Phosphate fertilisers in New Zealand contain on average 184 mg Cd kg P^{-1} and are voluntarily capped at 280 mg Cd kg P^{-1} [64]. This is due to New Zealand's history of high Cd-phosphate fertiliser use, leading to issues of Cd accumulation in productive soils [65]. This accumulation presents risks for food chain transfer and land use flexibility. Palm kernel expeller represents a low Cd source of P for farms and may have potential for the offsetting of high-Cd phosphate fertiliser use. Offtake of P by animals is assumed negligible, as most excess dietary P is excreted [8], however, the bioavailability of excreted nutrients including P is not clear. As some elements may be excreted in milk [6, 8], the potential for release into milk for the range of excess dietary elements contained in PKE should be assessed. The bioavailability of excreted elements through urine and faeces warrants investigation to determine the feasibility of any fertiliser offsets through PKE.

The practicality of offsetting fertiliser use would also depend on several factors including cost, feeding methods and TE concentrations in PKE. Importantly, imports of Cu and Zn to farm systems should be considered. Based on our PKE analyses, Cu and Zn have average concentrations in PKE of 28 and 50 mg kg^{-1}, respectively. In total, an average of 53 t Cu and 93 t Zn are brought into New Zealand dairy systems through PKE each year. Zinc is fed to cows to prevent facial eczema, particularly in warm, humid farming environments, and this has led to Zn accumulation in soils in some areas of New Zealand [66]. It is unclear how much further inputs of Zn, such as those through PKE, may exacerbate this issue. As mentioned in Section 5.3.2, there are potential benefits to animal nutrition from the Cu contained in PKE, however, a full investigation into the interaction of Cu with other TEs and the resulting bioavailability of Cu to animals is needed to understand this. Furthermore, as nutrients contained in PKE are applied to farm soils through animal excretions, their distribution to pasture should be considered. Unlike fertiliser applications that are evenly applied to pasture, animal urine and faeces are concentrated and non-uniform and this may exacerbate the challenges of ‘urine patches’ [67], whereby concentrated nutrient excretions lead to increased nutrient losses.

Potentially, other biowastes sourced in New Zealand may provide similar benefits to PKE for agricultural systems in terms of beneficial nutrient inputs. Each year, a total of 2,200,000 t of biowastes, including horticultural and viticultural wastes, are produced in New Zealand, of which the majority end up in landfill [68]. Further work might assess the suitability of some of these locally-sourced biowastes for potential use as stock fodder. Reducing the transport requirements of stock fodder may further contribute to the creation of circular bio-economies, recovering resources from and thus adding value to waste products that can be used to sustainably feed New Zealand’s agricultural sector.

5.4 Conclusions

Potentially, the chemical elements in PKE may offset nutrient deficiencies in agricultural systems. However, concentrations of Mg, Al, P and Fe exceeded MTLs for cattle feed and concentrations of S, K and Cu in PKE were within 90% of the MTL for cattle feed in one or more batches analysed. Concentrations of B, Mg, P, Cr, Mn, Fe, Ni, Cu and Zn were higher in PKE compared to pasture. Nutrient management on farms utilising PKE may be complicated by the statistically significant differences in essential and non-essential elements between batches of PKE. Our preliminary data analysis did not show a profound effect on liver Cu concentrations explained by the feeding of PKE to dairy cattle. The power analysis indicated that data from around 1300 farms would be needed to achieve adequate statistical power to produce meaningful and extrapolatable results if investigating potential effects of PKE on dairy cattle liver Cu concentrations. The concentrations of N, P and K represent a substantial source of nutrients into New Zealand dairy farms that may have potential for offsetting fertiliser use. In particular, PKE is low in Cd with a P: Cd ratio of 3.3 mg Cd kg P^{-1}. Thus, PKE may be useful as a low-Cd source of P. There is potential for future work to assess the feasibility of PKE to offset fertiliser use, including investigating the bioavailability of excreted nutrients, the potential excretions of elements through milk and any potential effects of excess elements such as Cu and Zn to dairy soils. Additionally, a risk assessment concerning elements that breach MTLs in PKE would be beneficial to ensure the use of this by-product as a supplementary stockfeed is sustainable.

5.5 Statements and declarations

Supplementary Materials: The following supporting information can be downloaded at: www.mdpi.com/xxx/s1, Table S1: Sampling locations for pasture; Table S2: SRM recoveries of elements.

Author Contributions: H. T.-M. Writing — original draft, formal analysis, investigation. E. M. Writing — review and editing, formal analysis, visualisation. S. G. Writing — review and editing, supervision. B. R. Writing — review and editing, supervision.

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Chapter 6: Development and use of a mass-balance model to calculate the likely effects of agrichemicals on trace element accumulation in soils supporting palm oil production

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Abstract

Trace elements (TEs) can reduce both the quantity and quality of agricultural produce when essential trace elements are deficient or when any trace element accumulates beyond threshold concentrations in soil. Therefore, TEs in agricultural systems should be managed to ensure that soil concentrations are kept within guideline levels and do not affect production. Oil palm (*Elaeis guineensis* Jacq.) agriculture in Indonesia occurs on weathered, acidic soils that require substantial fertiliser inputs to maintain production. To understand whether TE accumulation in soils resulting from fertiliser and pesticide use may affect production in the future, we have developed a mass-balance model to calculate likely concentrations of TEs in soils after given time periods under production conditions. Our model was developed to simulate processes that occur in the soil-plant system and involve the movement of TEs. It was developed to be rationally convenient, working at environmentally relevant concentrations, and able to be applied at the hectare scale to large agricultural systems. The model uses the mass-balance equation

$$\sum_{d} M_{Y,d} = \sum_{d} M_{0,d} + \sum_{y} M^{+}_{y,\bullet} - \sum_{y} \left(M_{y}^{(P)} + M_{y}^{(L)} + M_{y}^{(R)} \right)$$

and uses the critical parameters of rainfall, evaporation, erosion, soil adsorption coefficient, initial plant and soil contaminant concentrations, as well as the soil and crop specific parameters. We validated our model using a dataset from 64-year fertiliser trials, which had known inputs and application rates of F, Cd and U. We applied the model to a smallholder *E. guineensis* plantation in Indonesia, modelling accumulation of F, Cu, Cd and U under recommended production conditions, to determine which TEs might limit production and the likely timeframes for this. Our results indicate that topsoil concentrations of F and Cu were likely to reach phytotoxic concentrations in soils (719 and 289 mg kg^{-1}, respectively), within fifty years under these conditions. The future feasibility of intensive *E. guineensis* production on weathered, low fertility soils such as those in Indonesia should be assessed to avoid long-term negative impacts on soil quality.

Keywords

Trace elements; mass-balance; model; rhizosphere processes; *Elaeis guineensis*

6.1 Introduction

Most agricultural production requires applications of agrichemicals and amendments to promote crop growth and aid soil fertility. These include fertilisers, pesticides and organic amendments, such as composts. Many agrichemicals contain trace elements (TEs) as either active ingredients (e.g., Cu-fungicides) or contaminants. Phosphatic fertilisers may contaminate soil with TEs due to the impurities, especially F, Cd, and U that phosphate rock naturally contains (CSIRO, 2009; Loganathan et al., 2003; McGrath & Tunney, 2010; Taylor et al., 2016). Similarly, organic amendments such as sewage sludge and composts may contain elevated levels of these and other TEs (European Commission, 2004), while pesticides may contain TEs as active ingredients, particularly Cu, Zn and Mn (Gimeno-García et al., 1996; Rocha et al., 2014). Additionally, water used for irrigation may contain contaminants such as As and constitute a source of TEs to production soils (Bhatti et al., 2013).

Agrichemical application rates and type vary by system: e.g., in dairy farms, the predominant input is N at recommended rates (for New Zealand pasture) of 25-50 kg ha^{-1} yr^{-1}, while in horticultural systems the dominant agrichemicals are pesticides containing TEs such as Cu at rates of ≤80 kg ha^{-}1 yr^{-1} (Bivi et al., 2016; Fungicide Resistance Action Committee, 2021; Schneider et al., 2019). Factors influencing application rates of agrichemicals and associated TEs include the intensity of the system as well as the crops grown (Liu et al., 2021; López-Alonso et al., 2012). In intensive oil palm (*Elaeis guineensis* Jacq.) systems, recommended rates of N, P and K are 218–286 kg ha^{-1}, 143–215 kg ha^{-1} and 358–429 kg ha^{-1}, respectively (Woittiez et al., 2015). Varying rates of Cd are applied through P fertilisers due to varying native concentrations of Cd in phosphate rock: applications of 215 kg P could contain from 9.8 to 98 g Cd ha^{-1} (Oosterhuis et al., 2000).

When TE-containing amendments are applied to soils and crops over prolonged periods, these TEs have potential to accumulate in soils of productive systems (Luo et al., 2009; Molina et al., 2009). Consequently, soil fertility may be affected if these TEs reach threshold levels (Obbard, 2001), and plant uptake of TEs may reach critical levels for plant growth and food safety (Asati et al., 2016; Tóth et al., 2016). Repeated applications of TE-containing agrichemicals to productive land may limit both the sustainability of productive systems and future land use (Mills et al., 2005). In France, repeated application of $CuSO_4$ fungicides has resulted in the accumulation of Cu in soils that now limit agricultural production (Besnard et al., 1999). As well as degrading soil fertility and quality, TE accumulation in soils may endanger human health through food-chain transfer (Chaney, 1989). The export of offal from animals in New Zealand is restricted due to the risk of Cd accumulation in these animal products as a result of Cd-contamination of grazed pasture (Rys, 2011). Cadmium is highly plant-available in acidic soils and can pose food-chain transfer risks in concentrations <1 mg kg^{-1} (Elbana & Selim, 2010; Rys, 2011). Other TEs that may accumulate in soils as a result of agrichemical use include F, Zn, As, Pb and U. Accumulation of F in soils may be detrimental as it can contribute to soil degradation via dealumination processes, whereby excess F in soil results in accelerated weathering of aluminosilicates and release of Al^{3+} into soil solution (Taylor et al., 2012). This produces unfavourable conditions for plant growth due to Al^{3+} toxicity (Taylor & Kim, 2009). In response to these risks, soil guideline values (SGVs) have been developed to set limits on TE concentrations in soils. Such SGVs have been set to protect a variety of values, including human health, soil quality, land use flexibility and international trade (Cavanagh & Munir, 2016). Soil guideline values may be land use- and soil type-specific and differ for each TE: International SGVs for Cu and U in agricultural soils are 63 and 23 mg kg^{-1}, respectively (Canadian Council of Ministers of the Environment, 2007), while in acidic soils, Cd has a SGV of 1 mg kg^{-1} (New Zealand Water & Wastes Association, 2003). Exceedance of agricultural SGVs may reduce yields and result in economic losses (Rys, 2011).

Land converted to *E. guineensis* plantations from tropical forest in Southeast Asia often has low fertility and requires high fertiliser inputs, particularly P and K, to support production (Corley & Tinker, 2015; Woittiez et al., 2015). High applications of P fertilisers are associated with an increased soil Cd burden (Roberts, 2014). Pesticides, including Cu-based fungicides, are recommended to control plant pests common in *E. guineensis* production including *Ganoderma* spp. (Bivi et al., 2016). In contrast to traditional *E. guineensis* growing systems in parts of Africa (Corley & Tinker, 2015), *E. guineensis* production in Southeast Asia have undergone significant expansion since 1989 (Vijay et al., 2016) and most active plantations can be considered new systems that are naïve to fertiliser use. Soils from Indonesian *E. guineensis* growing regions are deficient in N, P and K, likely due to insufficient fertilisation (Thompson-Morrison et al., 2022a; Woittiez et al., 2015). *Elaeis guineensis* agriculture is subsidised in Indonesia, particularly with fertiliser subsidies (Kissinger, 2016; Woittiez et al., 2018). As a result, fertiliser applications are often limited to subsidised products (Woittiez et al., 2018). This has led to low levels of key nutrients that are not available in subsidised forms, such as K, in Indonesian soils (Thompson-Morrison et al., 2022a; Woittiez et al., 2015). Other forms of subsidisation include incentives for land use change to *E. guineensis* agriculture, often from native forests (Kissinger, 2016). Soils analysed by Thompson-Morrison et al. (2022a) from active and abandoned *E. guineensis* plantations in Indonesia displayed characteristics typical of soils with limited fertiliser histories, including P associated with mineral soil phases and concentrations of most TEs at or below background levels. However, accumulation of some TEs including Cu (Section 3.3.2) was noted. This accumulation was attributed to agrichemical use as correlations existed in these soils between P (used as a proxy for agricultural production) and TEs commonly associated with agrichemicals. There is potential that increased fertilisation in *E. guineensis* systems that raised soil nutrient levels to enable viable production would also result in the accumulation of TEs in these soils at concentrations that would eventually become phytotoxic (Thompson-Morrison et al., 2022a). Soils from older *E. guineensis* systems in Africa have been reported to have high TE concentrations (Olafisoye et al., 2020) and it is perceivable that the same may eventually occur in Southeast Asia.

Current agricultural systems are leading to TE accumulation and in some cases, this is unsustainable: the soil will either become infertile or crops will come to contain unacceptable TE concentrations. The likely timeframes to reach this endpoint can inform sustainable application rates (Mills et al., 2005). Mass balance models can calculate future soil TE concentrations under specified conditions. Mass balances entail the matching of inputs into a system with outputs from that system and are able to simulate relevant processes in order to calculate likely future scenarios (Wania & Mackay, 1999).

Several mass balance models specific to the determination of agriculturally-derived soil TEs have been developed in New Zealand (Roberts & Longhurst, 2005, as cited in Cavanagh, 2014), Australia (De Vries & McLaughlin, 2013), Canada (Sheppard et al., 2009), the European Union (ERM, 2000, as cited in Cavanagh, 2014), France (Michaud et al., 2019) and the United States (Chang et al., 2004) (Table 6.1). Of the models identified here, two are inaccessible (those described by Roberts and Longhurst (2005) and ERM (2000), as cited in Cavanagh, 2014) and as such, knowledge of their details is limited to secondary sources. The New Zealand model to assess Cd accumulation is available for public use however, the model itself remains unpublished. Some of the parameters used in this model are country-specific in that they depend on the selection of a New Zealand soil type that specifies the parameter. All of the models excluding the one described by De Vries and McLaughlin (2013) are analytical in that they are able to be calculated by hand with given inputs.

For the purposes of modelling TE fluxes in *E. guineensis* agroecosystems, we wanted a model that incorporates processes that occur in the soil plant system, specifically, the dynamic redistribution of TEs in soil resulting from plant uptake and the downward percolation of water. This necessitated a numerical rather than an analytical model. We wanted to be able to model a range of TEs including F, Cu, Cd and U to determine which TEs might limit production and under what timeframes. We also needed to be able to tailor model parameters to tropical soils and therefore the model had to be generally applicable rather than country-specific. We sought to model the primary processes that occur in agricultural systems and to determine the fate and movement of TEs in the soil-plant system. None of the models detailed in Table 6.1 were sufficient for our purposes or had been validated. We therefore aimed to develop a whole-system model that met these requirements and was easily parameterized and able to be applied to environmentally relevant concentrations of TEs in agricultural systems. The model should be able to be applied regardless of location or soil type, at the hectare scale for large-scale agricultural land. We validated our model using data from long-term agricultural trials with known sources and rates of TE inputs to soils over time. Finally, we used the model to calculate the TE concentrations likely to occur under *E. guineensis* production in Southeast Asian soils. We aimed to understand likely timeframes and limiting TEs of any accumulation.

6.2 Methods

6.2.1 Model development

Our modelling approach entails the simulation of the fluxes of TEs in the soil-plant system using lumped parameters. The primary processes considered here are inputs of water and contaminant to soil; downward movement of water and contaminants through soil; incorporation of applied contaminant (e.g. by ploughing) and downward movement of water and contaminants through soil; leaching of water and contaminant to groundwater; uptake of water and contaminant by plant roots; evapotranspiration, which drives the uptake of water from soil by plant roots; offtake of biomass containing contaminant from the site; return and reincorporation of biomass containing contaminant to soil; and soil and contaminant removal through erosion (Figure 6.1). The model takes a total of 17 input parameters specified by the user, divided into general, soil, plant and contaminant categories (Table 6.2). These processes are treated as one-dimensional, i.e. they do not incorporate lateral movement of water or TEs. The hectare scale equivalent is used in the model and therefore, it may be applied to any area.

Table 6.1. Comparison of parameters considered in mass balance models for the determination of soil fertiliser-derived Cd

Author and country	TEs applied to	Model type	Inputs considered	Outputs considered	Soil depth modelled to	Parameters considered
Roberts and Longhurst (2005), as cited in Cavanagh (2014), New Zealand (CadBal)	Cd	Analytical	Atmospheric deposition, fertiliser, biosolids	Harvested crop offtake, leaching (based on soil type), erosion (based on land use)	7.5–15 cm (land use dependent)	Soil bulk density (soil group dependent), fertiliser application rate, Cd fertiliser content, crop yield, calculated crop concentration, literature-based Cd leaching estimates
De Vries and McLaughlin (2013), Australia	Cd	Numeric	Atmospheric deposition, fertiliser, biosolids, irrigation	Harvested crop offtake, leaching (using K_D)	10–30 cm (land use dependent)	K_D, soil bulk density (using organic matter and clay content), crop yield, precipitation, evapotranspiration, runoff, pH *($CaCl_2$)*
Sheppard et al. (2009), Canada	Cu, Zn, As, Se, Cd, Pb	Analytical	Atmospheric deposition, fertiliser, biosolids, animal feed	Harvested crop offtake, leaching (using K_D), volatilisation (Se only), bioturbation	30 cm	K_D, soil bulk density (specified), crop yield
ERM (2000), as cited in Cavanagh (2014), European Union	Cd (possibly others)	Analytical	Atmospheric deposition, fertiliser, biosolids, animal feed	Harvested crop offtake (estimated), leaching (using K_D)	20–30 cm (plough depth)	K_D, soil bulk density (specified), soil background concentration, precipitation, infiltration, plough depth, crop TE uptake coefficient
Michaud et al. (2019), France	Cr, Ni, Cu, Zn, Cd, Hg, Pb	[Not reported]	Organic waste products, atmospheric deposition	Leaching, harvested crop offtake (measured)	<45 cm	Bulk density
Chang et al. (2004), United States	As, Cd	Analytical	Fertiliser and irrigation	Leaching, harvested crop offtake	30 cm	Volumetric water content of soil, hydraulic conductivity of soil, diffusion coefficient of TE in soil solution, root density of soil, root permeability coefficient

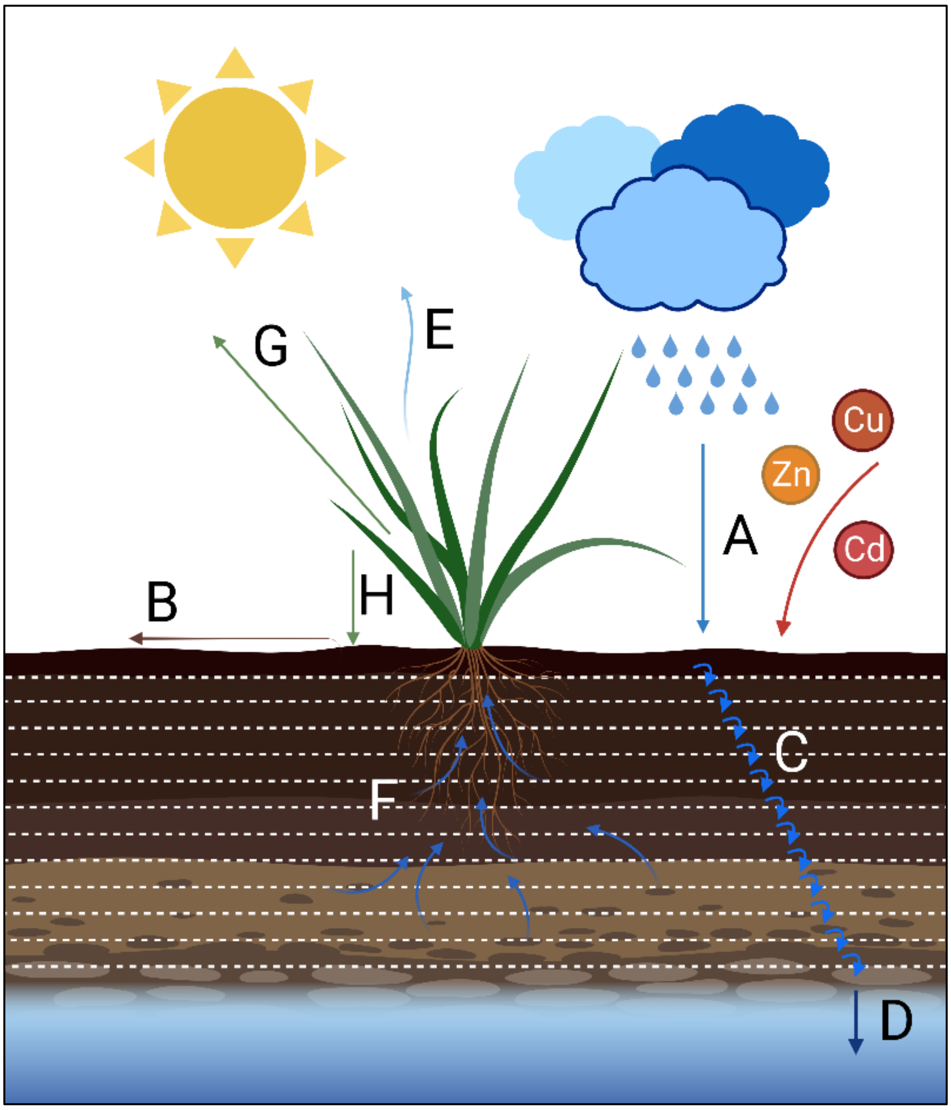


Figure 6.1. Key TE interactions in soil-plant systems. (A) Rainfall (and irrigation) water infiltrates the soil, while TEs are added to the soil surface through various mechanisms (fertiliser and amendment application, atmospheric deposition, presence in irrigation water). (B) Soil erosion may remove TEs from the topsoil as the top layer of soil itself is removed (although this represents a redistribution of TEs within the environment rather than a true loss). (C) Water percolates downwards through the soil profile, with the amount of water held in each 1 cm layer of soil (represented by dotted lines) determined by the soil's porosity and associated water-holding capacity. As water is the dominant TE-transport mechanism in soils, this percolation also mobilises and transports TEs downwards. The amount of contaminant mobilised here is a function of the soil adsorption coefficient (K_D). (D) Water and contaminant continue to move downwards beyond the soil profile and may be leached to groundwater. (E) Evapotranspiration removes water from the system and drives (F) plant uptake of water and TEs by plant roots (considered here to have a uniform density to a specified depth). (G) Removal of plant biomass removes taken up TEs, while the return of plant biomass to the soil (H) and subsequent re-incorporation of this returns TEs to the soil (Image created with BioRender.com).

Table 6.2. Model input parameters with relevant units and associated information. Notation refers to how the parameter is identified in equations and text throughout this chapter, while Code ID refers to the variable name in the written R code of the model (Appendix F).

Notation	Code ID	Parameter	Unit	Notes
General parameters				
y	iTime	Simulation time	yr	
D	iSimulationDepth	Simulation depth	cm	Must be ≥ root depth
T	iRainfall	Rainfall and irrigation	mm yr^{-1}	Rainfall infiltrates soil and is taken up by plants down to root depth
V	iET	Evapotranspiration	mm yr^{-1}	This is taken up evenly by plants down to the root depth (assumes uniform root distribution)
Soil parameters				
C_0	iInitialSoilCon	Initial contaminant concentration	mg kg^{-1}	Must be >0
E	iSoilLoss	Erosive loss	t ha^{-1} yr^{-1} (equiv)	Mass of soil lost to erosion and runoff
K	iKdSoil	K$_D$		Soil adsorption coefficient specific to contaminant
γ	iSoilDensity	Bulk density	t m$^{3\ -1}$	
W	iSliceWaterMass	Porosity	%	Equates to the mass of water (t ha^{-1}) that a 1 cm layer of soil can hold
Plant parameters				
[NA]	iDryBiomassProduced	Dry biomass produced	t ha^{-1}	Mass of removeable crop produced at the site
B	iPercentRemoved	Biomass removed from site	%	Harvested crop as a proportion of total removeable crop produced
D_R	iRootDepth	Root depth	cm	Must be ≤ simulation depth. Roots are considered to have a uniform density.
i	iInitialCropCon	Initial contaminant concentration	mg kg^{-1}	Must be >0
N	iK	Decay constant		$0 \leq K < 0.5$
Contaminant parameters				
M	iConApplied	Mass applied	g ha^{-1} yr^{-1}	Mass of contaminant applied in amendments, irrigation water etc
[NA]	iAtmosDeposition	Atmospheric deposition	g ha^{-1} yr^{-1}	Added to mass applied to give total contaminant added
D_I	iIncorporationDepth	Incorporation depth	cm	The depth to which the contaminant is incorporated into the soil

We wanted the model to be simple, rationally convenient and able to work with biologically relevant concentrations. The model can be envisioned as a tipping bucket, which means that it has to be implemented via numerical iterations rather than analytically. The model monitors the dynamics of soil properties over time by explicitly modelling a number of processes, including those described in Figure 6.1 over time at different depths. Below we describe the modelling process step by step. The full model code can be found in Appendix F.

6.2.2 Model processes

6.2.2.1 Initialisation

In what follows, we divide the soil into D 1 cm layers. Let $M_{y,d}$ describe the TE mass (g) in year $y = 1,\ldots,Y$ in layer $d = 1,\ldots,D$ with $d = 1$ corresponding to the topmost layer. The model is then initialised as follows.

Given soil bulk density γ, we evaluate soil mass at depth d as

$$S_d = 100\gamma \qquad \text{for } d = 1,,\ldots,D, \tag{Eq. 6.1}$$

and the initial TE mass at depth d as

$$M_{0,d} = S_d C_0, \tag{Eq. 6.2}$$

where C_0 is the initial TE concentration in soil. The first mass-balance variable, the mass of contaminant initially in the soil is then

$$M_{0,\bullet} = \sum_{d=1}^{D} M_{0,d}. \tag{Eq. 6.3}$$

6.2.2.2 Incorporation

Let $M_{y,\bullet}^{+}$ be the total contaminant mass (g) added to the soil in year y down to incorporation depth D_I. The addition to each layer of the soil can then be evaluated as

$$M_{y,d}^{+} = \begin{cases} M_{y,\bullet}^{+}/I & \text{for } d = 1,\ldots,D_I \\ 0 & \text{for } d > D_I, \end{cases} \tag{Eq. 6.4}$$

and the resulting TE mass in each layer can thus be updated as

$$M_{y,d} = M_{y-1,d} + M_{y,d}^{+}. \tag{Eq. 6.5}$$

In the current model we assume the amount of added contaminant to be constant over the years, i.e., $M_{y,\bullet}^{+} =$ constant. Maintenance phosphate applications, for example, are typically consistent over time (Mokany et al., 2010). Similarly, spraying applications are prescribed on an annual basis (MPOB, 2015).

6.2.2.3 Soil loss

The first mechanism considered in the model after the initial application of TEs is the loss of TEs from soil through runoff. This applies only to the topmost soil layer ($d = 1$). Given the annual erosive loss mass E (assumed constant in time), the fraction of TE mass lost due to runoff in the top layer is evaluated as

$$E^{(f)} = \min\left(\frac{E}{S_1}, 1\right). \tag{Eq. 6.6}$$

The TE mass in the top layer is then updated as

$$M_{y,1} = M_{y,1}(1 - E^{(f)}). \tag{Eq. 6.7}$$

The annual run-off soil loss is thus

$$M_y^{(R)} = M_{y,1}E^{(f)}. \tag{Eq. 6.8}$$

6.2.2.4 Drainage and leaching

The percolation of water and TEs downwards through the soil profile is simulated using the drainage and leaching function. This function uses the inputs of rainfall and irrigation (T), evapotranspiration (V) and porosity (W). All of these are assumed to be constant in the current model, as it uses average annual rates. The model does not include extreme climatic events and will be less accurate in climates where there is large annual variation in rainfall. For historical applications of the model, past climatic data that offers higher accuracy than average annual rates may be useful. Drainage and leaching are calculated on a yearly timestep. The soil profile is divided into 1 cm slices and where the water content in a single slice recahes above saturation, soil solution is transported to the underlying layer. Water leached from the bottom layer is assumed to be drainage. The mass of water moving through the soil profile (T^m) is described by the simple subtraction of water removed through evapotranspiration from the total water inputs as

$$T^{(m)} = 10 * \max(T - V, 0). \tag{Eq. 6.9}$$

The residual precipitation is multiplied by 10 to translate mm in cm. The model then determines the number of iterations (or loops) L to run T^m through each layer of soil as

$$L = \lfloor T^{(m)}/W \rfloor, \tag{Eq. 6.10}$$

where $\lfloor\rfloor$ denote downward truncation to ensure that L is an integer.

During each loop, $l = 1, \ldots, L$, at each layer of soil down to the specified simulation depth d, the TE mass in soil is mobilised by the downwards movement of water and is thus removed from that soil layer d and added to the next one $d + 1$:

$$M_{y,d} = M_{y,d} - M_{y,d}^{(s)} \qquad \text{for } d = 1, \ldots, D \tag{Eq. 6.11}$$

and

$$M_{y,d+1} = M_{y,d+1} + M_{y,d}^{(s)} \qquad \text{for } d = 1, \ldots, D, \tag{Eq. 6.12}$$

where $M_{y,d}^{(s)}$ is the solution mass evaluated as

$$M_{y,d}^{(s)} = \frac{M_{y,d}/S_d}{K} W. \tag{Eq. 6.13}$$

The annual leached mass $M_y^{(L)}$ is then the sum of all the $M_{y,D}^{(s)}$ over all the loops. Note, that water and contaminant continue to move downwards beyond the soil profile (the outflow boundary condition).

6.2.2.5 Plant uptake

Like the drainage and leaching function, plant uptake is calculated on a yearly timestep. Given the initial crop concentration i, the initial concentration of TE in soil C_0, and the coefficient of adsorption K, the root adsorption factor can be calculated as

$$\varnothing = \frac{i}{C_0/K}. \qquad \text{(Eq. 6.14)}$$

This parameter is dimensionless and represents the plant TE concentration / soil TE concentration quotient (Robinson et al., 2009). This is a simplified measure of the proportion of TE in soil solution taken up by plant roots, which is dependent on various complex factors.

For most TEs, plant uptake will be constant over time. The uptake of some TEs including Cu and Zn however, is plant-regulated and uptake rates will change over soil solution concentration ranges (Wairich et al., 2022). The associated change in $\varnothing$ can be modelled with a decay constant N as

$$\varnothing\,(H) = \frac{\varnothing_1 H_1}{H_1 + N(H - H_1)} \quad \text{(Robinson et al., 2003)} \qquad \text{(Eq. 6.15)}$$

where $\varnothing\,(H)$ is the root absorption factor at soil solution concentration H (mg L^{-1}) and $\varnothing_1$is the measured root absorption factor at soil solution concentration H_1. N represents the decay constant ($0 \le N < 0.5$).

Assuming, that roots grow to depth D_R and are evenly distributed through layers $d = 1, \ldots, D_R$, the contaminant mass removed from a layer of soil through plant uptake can then be evaluated as

$$M_{y,d}^{-} = \begin{cases} \dfrac{M_{y,d}^{(s)} \varnothing\,(H) B}{D_R W} & \text{for } d = 1, \ldots, D_R \\ 0 & \text{for } d > D_R \end{cases} \qquad \text{(Eq. 6.16)}$$

where B is the proportion of biomass removed (assumed constant over time) and W is the mass of water (t ha^{-1}) that a 1 cm of soil can hold, i.e., essentially the pore space of the soil. The values for the contaminant mass in the soil can be updated as

$$M_{y,d} = M_{y,d} - M_{y,d}^{-}. \qquad \text{(Eq. 6.17)}$$

The plant uptake of the contaminant solution in year y can then be evaluated as

$$P_y^{(s)} = \sum_d M_{y,d}^{-}. \qquad \text{(Eq. 6.18)}$$

and after the proportion B is removed, the rest gets redistributed back into the soil prompting a further update so that

$$M_{y,d} = M_{y,d} - (1 - B) P_y^{(s)} / D_I \qquad \text{for } d = 1, \ldots, D_I, \qquad \text{(Eq. 6.19)}$$

and $M_{y,d}$ remains otherwise unchanged. The annual TE loss due to plant removal is thus

$$M_y^{(P)} = P_y(s) B. \qquad \text{(Eq. 6.20)}$$

The values for the contaminant mass in the soil can be updated as

$$M_{y,d} = M_{y,d} - M_{y,d}^{-}. \qquad \text{(Eq. 6.21)}$$

6.2.2.6 Mass-balance equation

The mass balance model requires that the final TE mass in soil equals the initial TE mass in soil plus the TE mass added over the years minus the overall mass lost through plant uptake, leaching and run-off:

$$\sum_{d} M_{Y,d} = \sum_{d} M_{0,d} + \sum_{y} M_{y,\bullet}^{+} - \sum_{y} (\, M_{y}^{(P)} + M_{y}^{(L)} + M_{y}^{(R)}). \qquad \text{(Eq. 6.22)}$$

This equation functions as a check for the model, to ensure it has run correctly and that no error has been introduced through the model processes.

6.2.3 Other relevant model assumptions and limitations

The model has several key assumptions to facilitate parameterisation. Volatilisation was assumed to be negligible and not included in the model processes. Exceptions to this assumption would include modelling Hg, Se and As under anaerobic conditions. The model assumes linear K_D. We did not use a Freundlich equation for this parameter as a linear relationship was sufficient for the purposes of this model, for use within the contaminant ranges for which it was developed: i.e. those that are environmentally relevant and representative of sites with low to moderate levels of contamination such as agricultural soils, as opposed to heavily contaminated sites such as mine spoils (Clothier et al., 1988). The model does not account for lateral movement of water and TEs within the soil or use macropore flow elements. Consideration of macropore flow is more relevant for modelling of groundwater and receiving waters and therefore we considered it unnecessary for the purposes of modelling soil TE accumulation. The model does not account for a change in variables over the course of the simulation, e.g. a change in soil bulk density occurring over the simulation time. All input variables are assumed constant throughout the course of the simulation.

6.3 Model validation

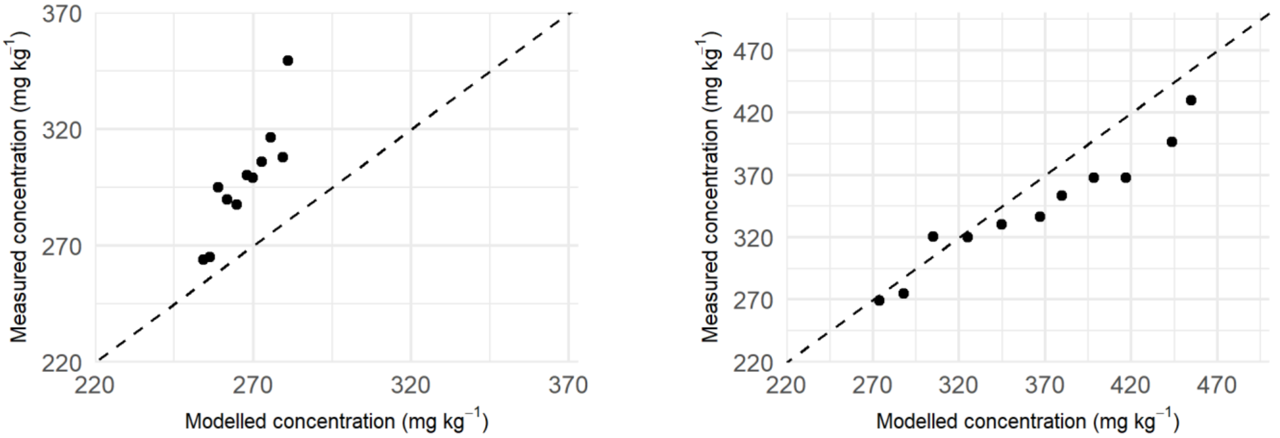
To validate the model, we used a data set from an agricultural site with a known history of contaminant inputs. The Winchmore long-term fertiliser trials have been running in Canterbury, New Zealand, since 1952, and have measured the concentrations of TEs including F, Cd and U in soil under different fertiliser regimes over time (Rickard & Moss, 2012). The Winchmore trials have a known fertiliser history, including known contaminant concentrations of the fertilisers applied. Parameters including annual irrigation, pasture production and climate parameters are well documented for these trials (Gray & Moss, 2021; Rickard & Moss, 2012). This provides an ideal data set for model validation, as both inputs and accumulation of TEs have been measured over a period of ca. 70 years. Sites such as Winchmore are rare and to our knowledge, no such sites with known histories exist in tropical regions where *E. guineensis* is grown. Therefore, we used the Winchmore data set to validate the model before applying it to tropical *E. guineensis* systems.

At the Winchmore trial site, two trials were run with single superphosphate applied to pastoral soil plots at rates of 188 and 376 kg ha^{-1} yr^{-1} (equiv) (McDowell et al., 2021). These trial plots are hereafter referred to as the 188 and 376 trial plots, respectively. The F, Cd and U concentrations of soils under each trial plot were measured at various time points since the commencement of the experiments. Cadmium had been measured in soils for 64 years, F for 63 years and U for 51 years. The

Cd and U concentrations of applied fertilisers were also measured at several time points (McDowell et al., 2021). This data provided known contaminant input model parameters and measures against which to compare our model outputs. Model inputs were determined for each parameter detailed in Table 6.2. Where data specific to the Winchmore site was not available, literature values were sourced from studies with conditions as close as possible to the conditions at Winchmore. A full list of model inputs used can be viewed in Appendix F.

To determine our model's accuracy at determining TE concentrations in soil after a specified number of years, scenarios were modelled with a simulation time equal to every year of the Winchmore trials that a specific TE was measured in. This was not the same between TEs: F was measured and hence modelled 11 times over 63 years, while Cd was measured 16 times over 64 years and U was measured and modelled 11 times over 51 years. The soil concentrations produced by the model were compared with the measured soil concentrations from the fertiliser trial (Figure 6.2). In each plot presented, each point corresponds to a point in time when the TE concentration was measured in the soil from the trial plot (16 for Cd, 11 each for F and U). Over the simulation time of 63 years, on average the model produced likely concentrations of F in soils of 90% (range 80-97%) and 106% (range 95-113%) of measured concentrations in the 188 and 376 trial plots, respectively. Likely Cd soil concentrations over 64 years simulation time on average were 92% (range 84-108%) and 78% (range 66-87%) of the measured concentration for the 188 and 376 trial plots, respectively. Over 51 years, likely U soil concentrations in the 188 and 376 trial plots were calculated by the model, on average, to be 109% (range 100-123%) and 123% (range 106-140%) of measured concentrations, respectively. The measurement of both F and U concentrations in soils encounters analytical challenges (Boschetti et al., 2017; del Carmen Lamas, 2005) and the variability and fluctuating concentrations of measured U in particular may be due to this. There is little published on the K_D of F and the value used in the modelling was determined by the authors' best judgement from literature values (Nakamura et al., 2017).

Root mean square error (RMSE) and mean error (ME) were calculated for each modelled scenario using log-transformed values so as to make errors comparable between scenarios (Table 6.3). The model tended to underestimate concentrations of Cd in both trial plots and F in the 188 trial plot, indicated by negative ME values. No error measurements for other models identified in Table 6.1 could be found for comparison, however the CadBal model from New Zealand was found to be accurate to within 90% of measured concentrations after 41 years (Gray & Cavanagh, 2022)

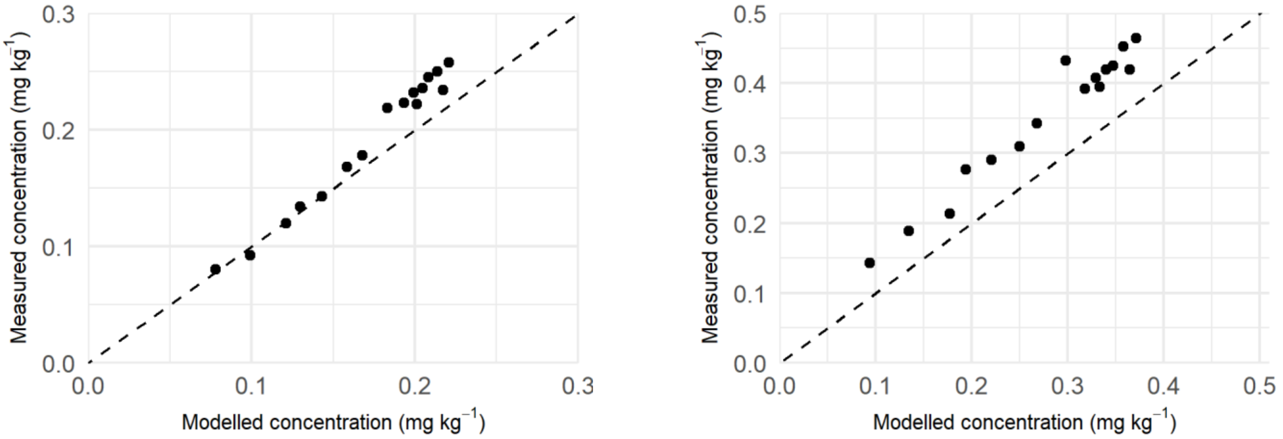


F 188 trial plot

$y = 2.3 x - 317 \quad r^2 = 0.80$

F 376 trial plot

$y = 0.75 x + 68 \quad r^2 = 0.94$

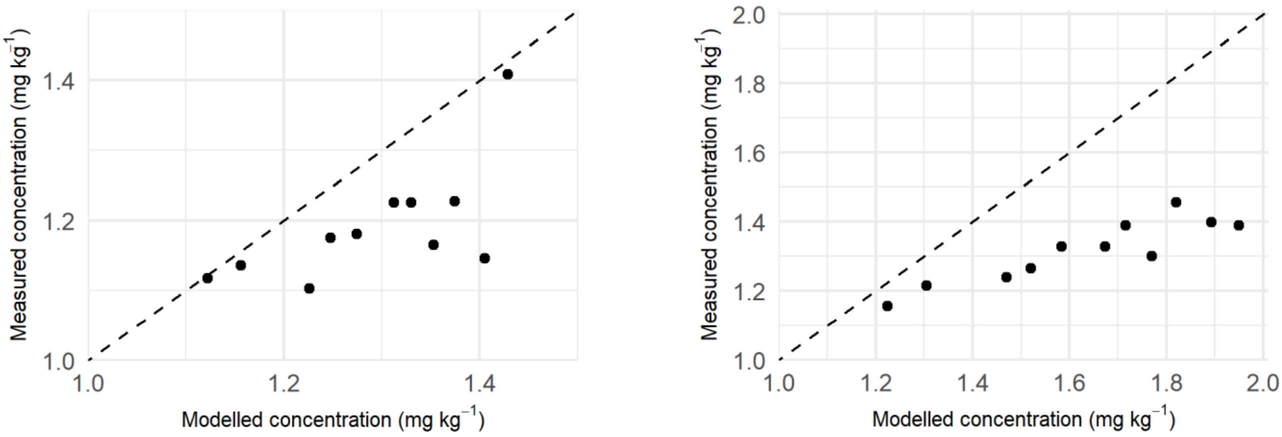


Cd 188 trial plot

$y = 1.3 x - 0.034 \ r^2 = 0.98$

Cd 376 trial plot

$y = 1.1 x + 0.039 \ r^2 = 0.96$



U 188 trial plot

$y = 0.56 x + 0.47 \ r^2 \ 0.44$

U 376 trial plot

$y = 0.35 x + 0.74 \ r^2 \ 0.82$

Figure 6.2. Scatter plots showing modelled and measured concentrations of F, Cd and U in soils at application rates of 188 and 376 kg single superphosphate ha^{-1} yr^{-1} using Winchmore trial data. Dashed lines represent a 1:1 relationship between measured and modelled concentrations.

Table 6.3. RMSE and ME using log-transformed values for each modelled scenario from the Winchmore trials

Element	Trial plot	RMSE	ME
F	188	0.12	-0.11
	376	0.07	0.06
Cd	188	0.11	-0.08
	376	0.26	-0.25
U	188	0.10	0.08
	376	0.22	0.21

6.4 Modelled trace elements in *Elaeis guineensis* production soils

We applied the model to an active *E. guineensis* plantation in Sumatra, Indonesia to determine whether TE accumulation was likely to occur as a result of agrichemicals used in production. Should accumulation be likely to occur, we also sought to understand the timeframes before SGVs were exceeded. The plantation used is described in Thompson-Morrison et al. (2022a) (Site C). This plantation was a smallholder plantation with flat topography soils classified as Acrisols (FAO, 2015) or Oxisols (USDA, 1999). Soils had a silt loam texture class, average pH of 5.1, and C and N concentrations of 2.1% and 0.035%, respectively (Thompson-Morrison et al., 2022a). Previous analysis of the TE concentrations of soil and plant materials from this plantation are detailed in Thompson-Morrison et al. (2022a) and Thompson-Morrison et al. (2022b), respectively. The soil and plant material at this site were deficient in N, P, K, Mg and Mo. Four TEs were modelled: Cu, due to its application in Cu fungicide and F, Cd and U due to their presence in phosphate fertilisers used in *E. guineensis* production.

6.4.1 Model input parameters

Two timeframes of eight and 50 years were chosen for the modelled scenario (Table 6.4). These represented both a mid- and long-term analysis. The eight-year timeframe was chosen to determine whether any degradation to soil as a result of TE accumulation will have occurred by the year that the United Nations Sustainable Development Goals (SDGs) are to be met (i.e. 2030). Soil quality is paramount to several of the SDGs, including SDG2: zero hunger, SDG6: clean water and sanitation, SDG12: responsible production and consumption, SDG13: climate action and SDG15: life on land (European Environment Agency, 2019). The 50-year timeframe was chosen to determine the sustainability of current recommended inputs into the next half century, as *E. guineensis* production is expected to continue and expand within this timeframe (Murphy et al., 2021). We modelled scenarios for F, Cd and U due to their presence in phosphate fertilisers, and Cu due to its application in Cu-fungicides that are used in *E. guineensis* plantations. Site-specific and contaminant-specific model input parameters were used to run each scenario (Tables 6.4 and 6.5, respectively).

Where possible, site-specific data was used to model the TE fluxes in the *E. guineensis* plantation. Where this data was not available, literature values from areas with conditions as close as possible to the site were used. Initial soil and plant concentrations of Cu and Cd were taken from Thompson-Morrison et al. (2022a) (Table 6.5). Initial plant concentrations of Cu and Cd were calculated as site-specific weighted mean kernel and mesocarp concentrations, as these plant materials are removed from the site during harvest. Literature values were used for F (Kumar, 2014; Mason & Moore, 1982) and U (Evans & Eriksson, 1983; Taylor et al., 2016) where there was no site-specific information available. Literature K_D values were used for Cd (USEPA, 1999), F (Nakamura et al., 2017) and U (USEPA, 1999) while Cu was site-specific (Thompson-Morrison et al., 2022a). A decay constant of 0 was used for non-essential TEs F, Cd and U while 0.1 was used for Cu as the uptake of this element is plant-regulated (Wairich et al., 2022).

The mass of contaminant added through fertilisers was calculated from TE contaminant rates in NPK (15-15-15) fertiliser (Oosterhuis et al., 2000; Taylor et al., 2016). NPK fertiliser was used as this was identified as the most commonly used fertiliser in a survey of smallholder plantations in Sumatra by Woittiez et al. (2018). Application rates were based on recommended per-palm P application rates to support *E. guineensis* production from Woittiez et al. (2015) using a recommended tree density of 143 trees ha^{-1} from FAO (1990). The maximum rate recommended by Woittiez et al. (2018) was used as the soils at this site were deficient in P. In the modelled scenario, this application was applied as maintenance fertiliser every year. Applications of initial capital fertiliser were not modelled due to the high application rates of maintenance fertiliser and the increased likelihood of nutrient runoff via overland flow in tropical environments with high rainfall. The modelled application rate of Cu was based on the mean fungicide application rate in Indonesian horticulture reported by Adiyoga and de Putter (2015) with the addition of 64 g ha^{-1} yr^{-1} added as NPK contaminants (Taylor et al., 2016).

Table 6.4. Site-specific model input parameters for the modelling of TE inputs to an *E. guineensis* plantation.

General parameters	
Simulation time (yr)	8; 50
Simulation depth (cm)	50
Rainfall and irrigation (mm yr^{-1})	2950 [a]
Evapotranspiration (mm yr^{-1})	1538 [b]
Soil parameters	
Initial contaminant concentration (mg kg^{-1})	[contaminant specific]
Erosive loss (t ha^{-1} yr^{-1})	5 [c]
K_D	[contaminant specific]
Bulk density (t m$^{3\ -1}$)	1.36
Porosity (%)	49 [d]
Plant parameters	
Dry biomass produced (t ha^{-1} yr^{-1})	10 [e]
Biomass removed from site (%)	100 [e]
Root depth (cm)	30 [f]
Initial contaminant concentration (mg kg^{-1})	[contaminant specific]
Decay constant	[contaminant specific]
Contaminant parameters	
Mass applied (g ha^{-1} yr^{-1})	[contaminant specific]
Atmospheric deposition (g ha^{-1} yr^{-1})	[contaminant specific]
Incorporation depth (cm)	1

[a] Rochayati et al. (2010)

[b] Takizawa et al. (2009)

[c] Aflizar et al. (2018). 48% of area in a measured watershed had erosion up to 5 t ha yr. Erosion up to >200 t ha^{-1} yr^{-1} was reported in this study. However, at rates this high, the issue becomes soil depletion rather than TE accumulation.

[d] Wasis (2012)

[e] Ng et al. (1999); Woittiez et al. (2018) specify 20 t ha^{-1} fresh fruit bunch, assumed 50% water

[f] Intara et al. (2018); Paramananthan (2015)

Table 6.5. Contaminant-specific parameters for the modelling of TE inputs to an *E. guineensis* plantation.

Contaminant	Initial soil concentration (mg kg^{-1})	Initial plant concentration (mg/kg)	K_D	Decay constant	Mass of contaminant added (g ha^{-1} yr^{-1})
F	321	0.5	365	0	32,890
Cu	5.6	15	5643	0.1	14,432
Cd	0.085	0.012	227	0	54
U	4	0.014	10,000	0	51

6.4.2 Model outputs

For each scenario modelled, a mass-balance output was produced (Table 6.6). We also produced four figures from the modelling results, corresponding with contaminant concentration in the final soil profile, the plant concentration of contaminant over the simulation time, the mass of contaminant leached over the simulation time and the contaminant concentration in soil over the simulation time (Figure 6.3).

Table 6.6. Mass-balances of all modelled scenarios. All masses are the total in 1 ha of soil down to the simulation depth of 50 cm.

Element	Simulation time (yr)	Initial mass in soil (g)	Mass added to soil (g)	Final mass in soil (g)	Final mass leached (g)	Final mass taken up by plants (g)	Final mass lost with runoff (g)
F	8	2,182,800	263,120	2,316,646	99,385	42	29,847
	50	2,182,800	1,644,500	2,991,878	621,154	329	213,938
Cu	8	38,080	115,456	134,826	112	1583	17,015
	50	38,080	721,600	418,896	700	11,866	328,218
Cd	8	578	432	932	42	1.5	34
	50	578	2700	2741	265	25	247
U	8	27,200	408	27,365	45	1.1	197
	50	27,200	2550	27,809	282	7.2	1652

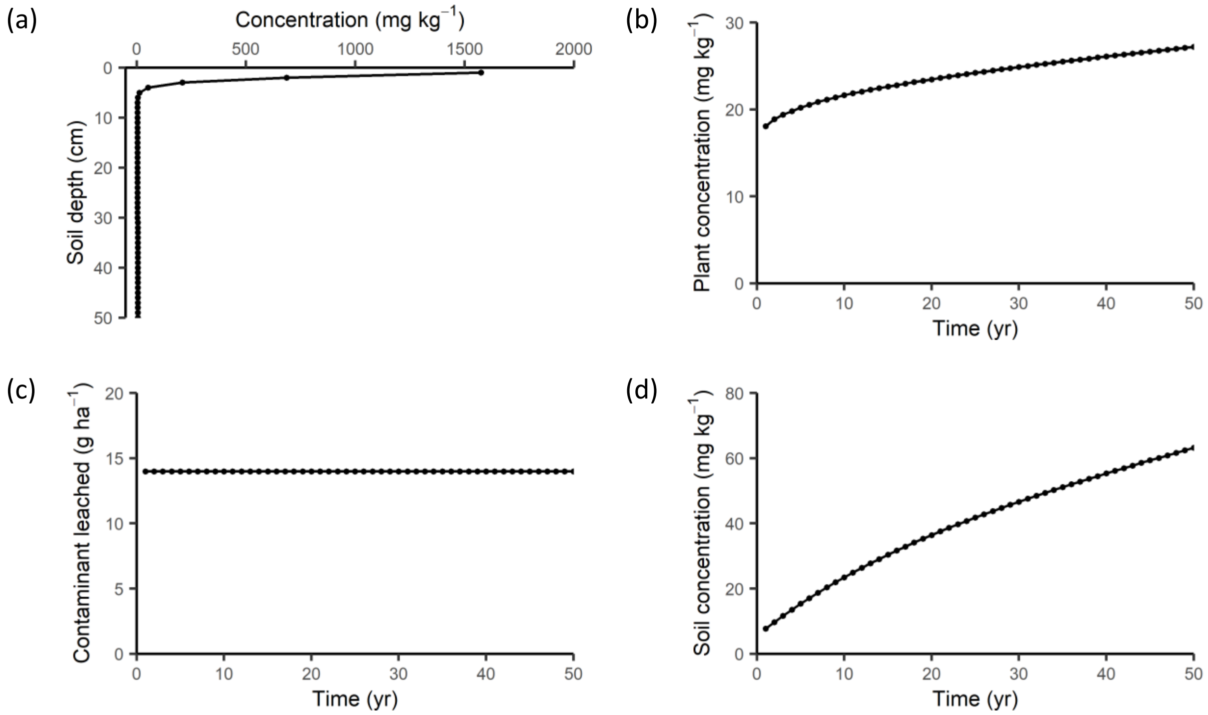


Figure 6.3. Output plots for Cu modelled in an *E. guineensis* plantation after 50 years, showing (a) Cu concentrations in the soil profile; (b) Cu concentrations in plants over time; (c) Cu concentrations leached over time; and (d) total soil concentrations over time.

6.6.4.3 Implications for *Elaeis guineensis* production

The modelling results indicate that accumulation of F, Cu and Cd above initial soil concentrations can be expected over time in the *E. guineensis* plantation because of NPK fertiliser and Cu-based fungicide applications (Figure 6.4). The amount of U added in fertiliser applications was low enough to effect no substantial change to background soil concentrations. The modelled concentrations of Cu surpassed SGVs within eight years while Cd and U did not surpass SGVs after 50 years. No SGV was used for F: only one relevant SGV could be identified for F, of 210 mg kg^{-1} (Cavanagh & Munir, 2016). This SGV was developed in New Zealand where F concentrations in soils are lower than the world average used in the modelled scenario, and therefore this SGV was not suitable as a guideline for F concentrations in Indonesian soil. Fluorine topsoil concentrations in the modelled scenarios rose sharply within eight years and more than doubled after 50 years, indicating that F accumulation in soils is likely to occur to potentially phytotoxic concentrations — estimated at 30-300 mg kg^{-1} for most plant species by Cronin et al. (2000) — in soils under *E. guineensis* cultivation. All TEs tended to accumulate in the topsoil and move down the soil profile over time. Fluorine accumulated in the topsoil (0-10 cm depth) to average concentrations of 419 and 719 after eight and 50 years, while Cu accumulated to average concentrations down to 10 cm depth of 77 and 289 mg kg^{-1} after eight and 50 years, respectively. At these concentrations, SGVs for Cu are surpassed and phytotoxicity of both TEs is likely to impede production. Accumulation of F in soils may also be detrimental as it can contribute to soil degradation under dealumination processes (Taylor & Kim, 2009; Taylor et al., 2012). The use of phosphate fertilisers and Cu fungicides in *E. guineensis* plantations presents risks to the sustainability of this system due to the presence of TE contaminants. We expect that concentrations of F and Cu will become sufficiently high within eight years that production will become unviable due to degraded soil and phytotoxic TE concentrations. This is not consistent with meeting SDGs that rely on soil quality. As TE accumulation in soils has been reported in *E. guineensis* plantations in Nigeria (Olafisoye et al., 2016; Olafisoye et al., 2020) and Malaysia (Aini Azura et al., 2012), we see this as an emerging issue for the industry. Management practices that minimize TE inputs to soil may be researched and adopted in *E. guineensis* cultivation to minimize this risk.

Our modelling results indicate that if sufficient nutrients were applied to *E. guineensis* soils, accumulation of F and Cu would be greater than it currently is. This current *E. guineensis* production system can be described as low intensity, with low levels of nutrients in the soil-plant system. This equates to a system with low land use efficiency (LUE). A lower LUE results in higher labour costs due to harvesting over a large area. A low LUE also results in larger areas of land being cleared to meet production. If the LUE of this system were increased through increasing the nutrient concentrations in the soil-plant system, TE accumulation in soils would likely be exacerbated. Therefore, fertiliser subsidies that incentivise *E. guineensis* agriculture in Indonesia and lead to low nutrient levels in soils (Woittiez et al., 2018) may also have the effect of reducing soil contamination – as nutrient levels are low, so too are TE levels in soil. This however, comes at the expense of clearing native forests as more land is required for production (Vijay et al., 2016). Trace element accumulation in soils under *E. guineensis* production in Indonesia is an issue that should be recognized and addressed, as the current state of production is not sustainable in terms of its impacts on soil quality.

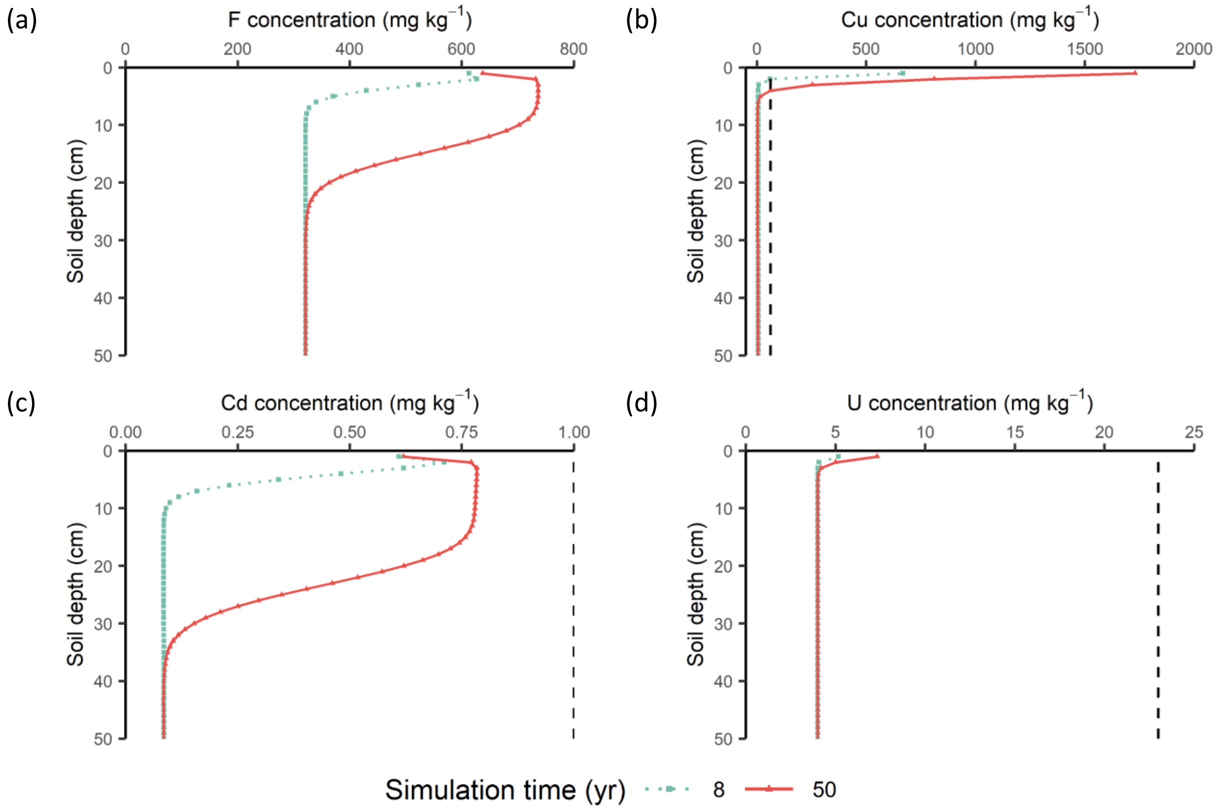


Figure 6.4. Final soil profiles for (a) F, (b) Cu, (c) Cd and (d) U concentrations after 8 and 50 years of inputs associated with *E. guineensis* production in a Sumatran plantation. Dashed vertical lines represent SGVs. The lower concentration in the top-most layer of profiles for F and Cd relative to the layer immediately below is attributed to the downward movement of the majority of TE in solution in the top-most layer, due to the K_D of these TEs.

6.5 Conclusions

To calculate likely timeframes and limiting elements associated with TE accumulation in *E. guineensis* plantation soils due to agrichemical applications, we have developed a mass-balance model to calculate the likely concentrations of TEs including F, Cu, Cd and U in production soils into the future. The model is rationally convenient, working with environmentally relevant concentrations, and is both parsimonious and mechanistic. This model is suitable for use with agricultural systems that receive some form of TE input and is applied at the hectare scale. This model has been validated with the Winchmore dataset for F, Cd and U over timeframes of up to 64 years. The results of our long-term modelling of *E. guineensis* plantation soils in Indonesia with recommended fertiliser inputs indicate that production will become unviable at this site due to phytotoxic concentrations of F and Cu within fifty years. In the modelled scenario, topsoil concentrations reached up to 719 and 289 mg kg^{-1} of F and Cu, respectively, within fifty years. While current nutrient levels in some production soils are low and likely to impede production, our findings indicate that if these deficiencies were corrected, TE accumulation would come to limit production. This ultimately brings the viability of this production system on weathered, acidic soils such as those in Indonesia, into question.

This model could be validated further using different datasets. It may also be possible to incorporate year-specific TE application rates that are not constant, as in the current model, to allow for varying application rates over time. As the current model is deterministic, introducing uncertainty factors may improve the usefulness of predictions. This may be introduced into the model using Monte Carlo simulations. As the focus of the current work was to use the model as a tool for the assessment of likely TE accumulation in *E. guineensis* soils into the future, this work will be carried out in future, separate publications.

6.6 Statements and declarations

Conflicts of interest: The authors have no competing interests to declare that are relevant to the content of this article.

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Chapter 7: General discussion

7.1 Elemental fluxes in *Elaeis guineensis* systems

This thesis has shown that African oil palm (*Elaeis guineensis* Jacq.) production and *E. guineensis* product saleability may be limited by several key elements in terms of deficiencies (N, Mo), excesses concentrations (Al, Fe, Cu, Zn, As, Pb) or both, in different parts of the system (P, K, Mg, S) (Table 7.1).

Table 7.1. Estimated population means of key elements in soils *and E. guineensis* materials determined using repeated measures analysis of variance (ANOVA) using the R package lme4 (Bates et al., 2015). Data was log-transformed for this analysis. All values in mg kg^{-1}, except for N. Elevated concentrations relative to standards or background values are signified by (+) while deficient concentrations are signified by (-). Plant deficiencies and excesses were determined by pinnae concentrations. Rachid concentrations are not included here as they are not directly relevant to the following discussion.

	Soil		Pinnae		Mesocarp	Endosperm	PKE		Oil	
N (%)	0.13	(-)	1.5	(-)	0.33	1.3	2.1		—	
P	201	(-)	1235	(-)	577	3529	5942	(+)	3.5	
K	1877	(-)	7212	(-)	4217	4907	9463	(+)	155	
S	171	(-)	1733	(-)	900	1571	2212	(+)	60	
Mg	2043	(-)	2785		1039	1823	3865	(+)	3.1	
Al	27,997		41		29	23	509	(+)	159	(+)
Fe	23,789		79		30	34	778	(+)	2.6	
Cu	9.5	(+)	5.8		14	16	28	(+)	0.05	
Zn	55	(+)	24		4.8	25	50		<0.01	
As	4.6	(+)	0.030		<0.01	<0.01	0.30		0.014	
Mo	0.80	(-)	0.13	(-)	0.055	0.22	0.39		0.021	
Pb	13	(+)	0.66		0.051	0.027	0.46		0.075	

7.1.1 Plant macronutrients

Macronutrients, particularly N, P, K, S, and Mg were deficient in soils and *E. guineensis* leaves in the sites tested. Deficiency here is used in the context that plant growth and production would benefit from the addition of these nutrients (Marschner, 2012). While the four sites tested are not representative of the *E. guineensis* Industry as whole, these findings have also been reported by (Corley & Tinker, 2015; Cui et al., 2020; Woittiez et al., 2015). This indicates that nutrient deficiency maybe widespread among *E. guineensis* plantations. In contrast to the *E. guineensis* soil, nearby forest soils were not deficient in most essential macronutrients (Wasis, 2012).

The macronutrients N, P, K, S and Mg were higher in palm kernel expeller (PKE) than in any plant materials analysed. One explanation for this is that the source material for the PKE analysed was higher in nutrients than the raw plant materials analysed. As the plant samples were sourced from one region in Sumatra, it is plausible that the PKE analysed was sourced from a different *E. guineensis* growing region with higher-nutrient plant materials. Another possibility is that post-harvest mixing occurred with other high-nutrient material (Section 5.3.1.1.1). It is possible that post-harvest contamination of PKE may have resulted in higher concentrations of P, K, S and Mg (Figure 2.11), however, this unlikely with regard to N because the amount of contaminant required to significantly increase N concentrations would be a significant (i.e. several %) proportion of the mass of the PKE. The N concentrations in PKE was likely a result of high N material included as source material for PKE, whether this be high N *E. guineensis* endosperm material or other material added post-harvest.

7.1.2 Aluminium and iron

Aluminium was present in high concentrations in both palm oil and PKE while having typical soil and plant concentrations (Table 7.1). Unlike most other TEs, Al may form phloem-mobile organic complexes in plants and partition into fruit tissues (Benning et al., 2012). **T**he provenance of Al in *E. guineensis* products cannot be confirmed, however, possible reasons include plant physiology, if Al in the plant materials was concentrating in fruit tissues and partitioning into the lipid phase of the plant. Aluminium speciation in environmental samples has many drivers and its toxicity often depends on the bioavailability of species present (Rengel, 2004). As the concentrations in the products analysed were in the range of an order of magnitude higher than the concentrations in plants, some of the Al present in oils and PKE may have been due to contamination during the processing and manufacturing stages of production. This is also likely true for Fe, which had typical concentrations in soils and plants (Mason & Moore, 1982) however, significantly larger concentrations in PKE. It is likely that Fe was entering the system post-harvest, in machinery containing iron or steel parts used in manufacturing, or during shipping if the PKE was stored and transported in the steel hull of a ship.

7.1.3 Other trace elements

While Zn, As and Pb were elevated in production soils, likely due to fertiliser use, they did not carry through the system: concentrations in plant materials and plant products were not elevated (Table 7.1). Copper, however, was elevated in production soils as well as PKE. Copper had high bioaccumulation coefficients (BACs) (>1) for all plant materials at one site and high BACs in fruit tissues at another site (0.89 and 0.79 for endosperm and mesocarp, respectively). This indicated that Cu is phloem-mobile in *E. guineensis*. Thus, the Cu concentrations in PKE likely originated in *E. guineensis* plantations where Cu entered the system, possibly through Cu-fungicide applications, and carried through to end-products. Copper-fungicide may have been applied indirectly to soils through spray-drift before being taken up by *E. guineensis* roots via root-surface exchange sites (Salisbury & Ross, 1992). From there, Cu may be transported via xylem and phloem transport to the fruit tissues (Marschner, 2012). The low Cu concentrations in palm oil indicated that like most TEs, Cu partitioned away from the oil phase of the plant and remained in the dry-matter portion of the endosperm. It is also possible that some Cu may have entered PKE post-processing: PKE has been demonstrated to have a high Cu-adsorption capacity from aqueous solutions (Ho & Ofomaja, 2006). If PKE was exposed to aqueous materials with substantial Cu contents, such as water used in processing with a high Cu content, some Cu may have been adsorbed onto PKE from this source. Despite the high Cu content of PKE, results from the pilot study (Section 5.3.3) showed no statistically significant difference between the Cu concentrations in the livers of dairy cattle fed and not fed PKE. The statistical power of this analysis was however, low, and the power analysis indicated that to achieve sufficient statistical power to draw conclusions about whether the Cu content of PKE was affecting the liver status of New Zealand dairy herds, an analysis would require data on cows from approximately 1300 farms.

7.2 Implications for environmental quality

Soils under *E. guineensis* plantations were largely deficient in essential plant macronutrients (Chapter 3) as well as Mo. These deficiencies were also indicated by sub-optimal concentrations of these nutrients in plant tissue (Chapter 4). It is likely that on the sites tested, nutrient deficiencies resulted in significantly reduced production, which in turn, reduced land use efficiency and raised the overall costs of production, since greater resources of land, agrichemicals and human capital would have been required. This was likely a contributing factor to the abandonment of three of the sites sampled.

This thesis posed the hypothesis that soils under *E. guineensis* production would contain elevated concentrations of TEs due to agrichemical applications. The data presented in Chapter 3 was consistent with this hypothesis as soils from three sites contained elevated concentrations of TEs including Cu, Zn, As and Pb, relative to background levels. The concentrations of these TEs present, however, did not surpass soil guideline values (SGVs). The soils from the sites sampled contained lower organic C relative to nearby forest soils (Wasis, 2012). The *E. guineensis* production soils analysed were deficient in key nutrients including N, P, K, Mg and Mo. The hypothesis that production soils would contain higher N concentrations than background levels was therefore falsified. The findings of the thesis indicate that plant nutrition requirements were not met in the plantations sampled and that the soil deficiencies present translated to plant deficiencies. Plants from these sites had concentrations of N, P, K and Mo, which were low enough as to affect yields and likely contributed to the abandonment of three of the four plantations sampled. There was no evidence of contamination in plant materials from any of the sampled sites or in refined oils, however, Na and Al were present in palm oil at higher concentrations than had been previously reported in other studies (Asemave et al., 2012; Nnorom et al., 2014; Obi et al., 2001).

7.2.1 Edaphic factors reducing fertility

The soils analysed in this thesis, which are typical of Sumatran *E. guineensis* growing regions, were acidic and highly weathered. The pH ranged from 4.6-5.4 and the soils exhibited a high P-fixation capacity, consistent with reported high P-retention in tropical Oxisols (Spain et al., 2018). The results indicated that N, P and S were associated with the organic phase of the soil and therefore highly unavailable to plants. Two of the sites sampled had high C:N ratios (60 and 95), which indicated that N in these soils is likely to undergo rapid immobilisation and become unavailable for plant uptake.

7.2.2 Climate-induced nutrient loss

As Sumatra receives approximately 3000 mm of annual rainfall, the risk of topsoil loss and nutrient runoff is substantial. Erosion rates in Sumatra have been measured at up to >200 t ha^{-1} yr^{-1} in steep areas (Aflizar et al., 2018). Three of the sites sampled contained steep areas, with slopes up to 12°C. With high erosion rates, the issues for soil management become loss of soil and nutrients and redistribution of elements in the environment, rather than accumulation of any TEs.

7.2.3 Insufficient fertiliser application

Elaeis guineensis is an intensive crop and substantial nutrient inputs are needed through fertilisers. Some fertilisers are subsidised and consequently, these are the most commonly used. The type of fertilisers subsidised — particularly NPK (15-15-15) — are not the most appropriate agronomic choice for *E. guineensis* production and alone, do not meet the nutritional needs of the crop (Woittiez et al., 2015). Soils in Sumatra typically have low background concentrations of K (Rafflegeau et al., 2010), a key nutrient for *E. guineensis*, and require inputs of 358–429 kg ha^{-1} (Woittiez et al., 2015). These requirements often go unmet, due to the lack of subsidised K fertiliser available.

The most common rate and application of fertiliser in Sumatran smallholder plantations is not a sufficient rate of P for production: 692 kg ha^{-1} yr^{-1} of NPK (15-15-15) equates to 104 kg P ha^{-1} yr^{-1} with recommended inputs being between 143–215 kg P ha^{-1} yr^{-1} (Table 1.1) (Woittiez et al., 2015). This is a likely reason that TE concentrations were generally similar to background levels in the soils measured. While F was not able to be measured in this thesis due to analytical limitations of the facilities available, the modelling undertaken in Chapter 6 indicates that should sufficient rates of fertiliser be applied, concentrations of F in soils would increase substantially within eight years. This would negatively affect production and soil quality, due both to phytotoxicity due to excessive F levels, combined with potential dealumination, which exacerbates the effects of soil acidification and Al^{3+} toxicity. Increases in the rates of fertilisers applied in *E. guineensis* plantations have also been shown to lead to statistically significantly higher leaching losses of N and base actions than from forest and rubber production (Clough et al., 2016). This can lead to the eutrophication of nearby surface water and the contamination of groundwater with nitrates, which further negatively affects environmental quality.

7.2.4 Implications for the sustainability of *Elaeis guineensis* production

These factors (Sections 7.2.1-3) combined may explain why typical oil yields (4 t ha^{-1}) from *E. guineensis* are far from optimal (8 t ha^{-1}) (Corley & Tinker, 2015; Mohd Din et al., 2014; Woittiez et al., 2019). This sub-optimal land use efficiency necessitates larger areas for production. Furthermore, the *E. guineensis* industry in Indonesia has indicators of an uncompetitive industry, with subsidies and high barriers to entry, which is likely to exacerbate the pattern of inefficient land use (Chalil, 2008) (Section 2.2). As the land converted for production in Indonesia primarily comes from virgin forest that is home to critically endangered species (Vijay et al., 2016), this results in a trade-off between economic and environmental values. This brings into question the economics of palm oil production. Subsidies and foreign direct investment play key roles in Indonesia's *E. guineensis* industry, and subsidies in particular can lead to market distortions that promote inefficient production and resource allocation (Kostadinov, 2013; Wuyts, 2020). Subsidies have also been recognised to promote intensive use of fertilisers in developing countries where environments may not be suitable to production (Kostadinov, 2013). The data from the thesis indicates that this is happening in Indonesia with regard to *E. guineensis* production, where economic policies are incentivising land use conversion to a production system that is not suited or sustainable given the limitations of the natural environment. Furthermore, multiple assessments have indicated that the economic value of leaving land as primary forest exceeds the value gained from conversion to *E. guineensis* in Sumatra (Clough et al., 2016; van Beukering et al., 2003). A further criticism of subsidised production is that it encourages wasteful consumption (Wuyts, 2020). The exponential increase of vegetable oil production (Figure 2.1), and in particular palm oil production, constitutes a potential example of this. It is clear, from the analysis done in the thesis and the supporting data that an assessment of the economic viability of *E. guineensis* agriculture without subsidies and foreign direct investment should be done. These economic incentives currently support production that, from a physiochemical perspective, is unsustainable.

The results in this study were from four *E. guineensis* production sites in a single region of Sumatra. While this region may not typify other *E. guineensis* production, the conditions of these sites in terms of conversion from rainforest and fertiliser inputs are typical for many areas of *E. guineensis* production. A widespread survey of soils and plants would reveal the extent to which *E. guineensis* production is constrained by nutrient deficiencies.

7.3 Correction of nutrient deficiencies

Correction of soil nutrient deficiencies would require significant capital inputs, particularly of P, K and Mg, which would increase soil concentrations of P-associated contaminants, especially F, Cd, and U. Contaminants are used in this context to describe the presence of TEs at concentrations above background – this is distinguished from pollution, which entails an exposure pathway that results in harm to the environmental, animals or humans (Chapman, 2007). The cost of such capital P would be in the order of USD$1860 per hectare, based on USD$595 per t of fertiliser (Berkahtani, 2022) and an application rate of 500 kg P ha^{-1}. Recommended capital fertiliser inputs for *E. guineensis* are not detailed, however, it is estimated, based on maintenance rates of 143–215 kg P ha^{-1} that these would be approximately 500 kg ha^{-1}. At current (unsubsidised) prices, this per hectare cost is substantial compared to the approximate land value of USD$3200 ha^{-1} (*Land for sale in Langkat, North Sumatra*, 2022). Maintenance P would be similarly expensive at ca. $670 ha^{-1} yr^{-1} (based on a mid-rate of recommended inputs, 180 kg P ha^{-1}). This combined with the necessary urea and potash amendments would likely render *E. guineensis* production uneconomic: current returns from palm oil production are in the order of USD$1550 ha^{-1} yr^{-1}, which has reduced during January–June 2022 from approximately USD$4200 ha^{-1} yr^{-1} (Musim Mas, 2022).

7.4 Phosphorus use efficiency in *Elaeis guineensis* plantations

As P is a non-renewable, limited and costly resource, the efficient use of P in agricultural systems is desirable from both an economic and a resource management perspective. It is possible to calculate P use efficiency (PUE) in *E. guineensis* plantations using the concentrations of P present in the removed biomass (i.e. endosperm and mesocarp) and the known P inputs to soils. The abandoned *E. guineensis* plantation sites (A and B) contained statistically significantly less P in mesocarp and pinnae tissue relative to the active site (C) (Tables 4.1 and 4.3), which is assumed due to the three years since these abandoned sites were last fertilised. Therefore, the P concentrations in plants from these sites were not assumed to be representative of *E. guineensis* plants under managed production, and PUE was calculated for the active site, using this site as a case study. Phosphorus use efficiency was calculated using the equation

$$PUE\% = \frac{B}{F}\,100 \qquad \text{(Selles et al., 2011)} \qquad (\text{Eq. } 7.1)$$

where *B* represents the amount of P removed in biomass and *F* is the amount of P added in fertilisers, all in kg ha^{-1}. A weighted mean P concentration for mesocarp and endocarp tissues for the *E. guineensis* variety tenera, which was grown at Site C, was used to calculate *B*, using an endosperm: mesocarp ratio of 23:77 (FAO, 2002). This mean P concentration of 1372 mg kg^{-1} was multiplied by average Sumatran smallholder fruit yields of 15.4 t ha^{-1}. This fresh fruit yield was calculated as average fresh fruit bunch yields of 20 t ha^{-1} (Woittiez et al., 2018) minus the proportion that constitutes empty fruit bunch yields, 23% (Baharuddin et al., 2010). The results of this calculation determined that the amount of P removed in biomass from the site in fruit tissues totalled 21 kg ha^{-1}. Input rates of P for Sumatran smallholder plantations were calculated from literature values sourced in farmer surveys: whereby an NPK (15-15-15) application rate of 692 kg ha^{-1} (Woittiez et al., 2018) equates to 104 kg P ha^{-1} applied. As leaf materials are generally returned as soil amendments in *E. guineensis* plantations, it was not considered necessary to include these in the equation and used a simplified 'balance method' to calculate PUE (Selles et al., 2011; Syers et al., 2008). Using Eq. 7.1, the calculated PUE at Site C was 20%. This is low compared to PUEs from other land uses — for example, crop-pasture systems across multiple countries in Asia, Europe, North America and the Pacific reported an average PUE of 72% (Biswas Chowdhury & Zhang, 2021).

This low PUE is consistent with the high P retention typical of oxidic tropical soils (Spain et al., 2018) and the association of P with the organic phase in these soils (Section 3.3.4). The majority of P applied to these soils is immobilised, and more P would be applied to Site C than would be harvested from it in plant biomass. This represents a substantial cost for these systems. Furthermore, the P that is harvested from these soils is then exported offshore in products including palm oils and PKE.

7.5 Elemental contaminants entering food systems through *Elaeis guineensis* products

Concentrations of most non-essential trace elements and micronutrients in the *E. guineensis* products analysed (PKE and palm oil) were low compared to food and fodder safety standards for humans and animals (Chapters 3 and 4). There were however, excessive concentrations of the macronutrients P, K, Mg, S in PKE, as well as Al and Cu in PKE (Table 7.1). Palm oil contained concentrations of Na (1198 mg kg^{-1}) and Al (159 mg kg^{-1}), which were higher than any previous sources had reported. The other elements analysed in palm oil were present in low concentrations that posed no risk to food safety standards, as the thesis had hypothesised. This thesis did not assess organic pesticides residues or contaminants other than TEs that may contaminate plant products and future work may address this.

The Al present in palm oil may pose a toxicity risk for children below 41 kg in body weight if they are consuming the average daily intake of palm oil (37 g day^{-1}) (Zhao et al., 2021). This would cause the maximum weekly intake of 1 mg kg^{-1} bw wk^{-1} to be exceeded (Otten et al., 2006). A wider survey of the concentrations of Al in palm oil from other products and origins would reveal whether any Al-toxicity risk is associated with the consumption of palm oil.

Aluminium is also present in PKE in concentrations that exceeded maximum tolerable levels (MTLs) for cattle feed (Table 5.2). Aluminium was the only non-essential element in PKE to exceed MTLs. Other elements present in high concentrations were essential animal nutrients: Mg, Al, P and Fe exceeded MTLs while K, S and Cu approached MTLs. These elements may be beneficial or deleterious to animal health depending on factors including diet proportions of PKE and antagonism or synergism occurring between elements. Copper is commonly supplemented in New Zealand agricultural systems while Mg is used for the prevention of milk fever in dairy cows. Potentially, their concentrations in PKE may provide benefits to these importing agricultural systems. The full implications of this are discussed in Section 5.3.2.

The high P concentration in PKE is in contrast to the low P status of production soils and plant materials in Indonesia. The low PUE of the production soils analysed is further emphasised given that the majority of P that is harvested from these production soils in plant biomass is exported offshore and provides benefit to foreign agricultural systems, such as New Zealand dairy systems through PKE. This may provide more value to Indonesian *E. guineensis* systems if it was returned to production soils as an organic amendment. Furthermore, the P:Cd ratio in the exported PKE is lower than commonly used fertilisers in New Zealand and Indonesia (Abraham et al., 2016; Taylor et al., 2016). The use of PKE to offset purchased nutrients on New Zealand dairy farms may be worth exploring, however, this would require further analysis on the fate of key nutrients in PKE in terms of animal absorption or excretion. Previous work has shown that feeding PKE to lactating cows affects milk composition (Van Wyngaard & Meeske, 2017). As elements such as P can be excreted through milk (Goselink et al., 2015), and as milk has been previously identified as one of the top ten contributors to Cd in female diets in New Zealand (Abraham et al., 2016), the potential for elemental contaminants in PKE to enter the human food chain through milk warrants investigation. Further work is also required on the effects of TEs including Cu, Fe and Al on animal health. As PKE contains potentially harmful TEs and there is potential that locally sourced biowastes could offset its use on New Zealand dairy farms (University of Canterbury, 2021), these questions should be addressed to ascertain whether the continued import and use of this product is appropriate, given the low LUE of production soils and the implications this has for continued clearance and conversion of tropical rainforests.

7.6 Challenges associated with investigating *Elaeis guineensis* production systems

The palm oil production industry is a major contributor to the Indonesian economy, with plantations spread throughout the country (UNDP, 2019). Indonesia has strong protectionist laws and vested domestic interests (Merkle, 2018) and this has resulted in difficulties associated with conducting scientific investigations into this industry. Law-changes have been introduced ("National System of Science and Technology," 2019) that restrict the ability of foreigners to conduct research in Indonesia and to export scientific samples (Rochmyaningsih, 2018, 2019). This has resulted in challenging conditions for scientists based outside of Indonesia attempting to conduct research on palm oil production. Indonesian scientists have indicated that they worry that investigation into *E. guineensis* production could have a negative impact on their careers if they produce data with negative implications for *E. guineensis* production (Anon, personal communication, 20th June 2021). This creates a difficult environment in which to conduct scientific research into this industry.

7.7 Conclusions and outlook

Tropical soils that are converted from virgin forests contain insufficient nutrients for *E. guineensis* production, particularly N, P, K, Mg and Mo. Correction of these nutrient deficiencies may be prohibitively expensive without subsidies. Inputs of sufficient fertilisers may improve yields but are likely to cause *E. guineensis* production soils to accumulate TE contaminants to the point where production is impaired within 50 years. Copper, Zn, As and Pb are currently accumulating in *E. guineensis* production soils, however, these TEs have not yet reached concentrations where they would reduce soil fertility. Given that *E. guineensis* production is relatively new to Southeast Asia, excessive accumulation of elements has yet to be observed. Analyses of plantations in Nigeria, where *E. guineensis* has been produced for more than 100 years, may indicate the likelihood of these elements becoming an issue for sustainability. Due to the analytical difficulties of measuring F in environmental samples, concentrations of this TE were not measured in this thesis, however, modelling indicates that based on background concentrations and recommended inputs, within eight years under current production systems, concentrations of F and Cu may detrimentally affect productivity of *E. guineensis* plantation soils. In the *E. guineensis* production systems tested, production is unlikely to be economically viable without subsidies.

The palm oil analysed contained no elemental concentrations above food safety standards, however, the concentrations of Al may pose toxicity risks for children and should be further investigated. In addition to its value as animal fodder, PKE may be important source of low-Cd phosphate in New Zealand's agricultural systems. However, PKE contained concentrations of P, Mg, Al and Fe that exceeded maximum tolerable levels for these elements in cattle fodder and concentrations of K, S and Cu that were within 90% of the maximum tolerable levels. Given that many of these elements were deficient in the *E. guineensis* production soils tested, it is likely that PKE contamination occurred post-harvest or that the soils where the PKE was produced were distinct from the soils in this study.

There are several key questions that this thesis has highlighted that should be addressed with future research. Specific recommendations include:

1. An economic assessment to determine whether *E. guineensis* agriculture would be economically viable without subsidies, given TEs may limit this production system in the future.
2. A wider survey of *E. guineensis* plantation soils in other *E. guineensis* producing regions of Indonesia, including in high-input systems. This survey should include analysis of F in soils.
3. An assessment of the effects of elements in PKE that exceed or approach MTLs — including P, K, Mg, S, Fe, Cu and Al — in importing dairy systems, concerning animal health.
4. An assessment of whether elements in PKE may be entering human food chains through milk.
5. Assessment of the feasibility of biowastes to offset purchased nutrients on-farm.
6. An assessment of organic contaminants that may be contained in PKE.

7.8 References

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Appendices

Appendix A: Supplementary materials for Chapter 1

Table A1: Rare earth element concentrations in Aceraceae species

The following table provides reported rare earth element concentrations in Arecaceae species including *Phoenix dactylifera*, *Euterpe oleracea* and *Euterpe edulis*.

Element	**Mesocarp concentration *P. dactylifera* (mg/kg^{-1})**	**Seed concentration *P. dactylifera* (mg/kg^{-1})**	**Mesocarp concentration *E. oleracea* (mg/kg^{-1})**	**Mesocarp concentration *E. edulis* (mg kg^{-1})**
Sb	0.035 (0.004)[a]	—	—	—
Cs	0.010 (0.002)[a]	—	—	—
Ba	<0.009-0.20[b]	<0.009-0.160[b]	—	—
Pr	—	—	2.25-44.41[c]	3.78-38.01[c]
Nd	—	—	8.83-896.35[c]	45.05-490.55[c]
Sm	—	—	2.52-198.4[c]	13.40-111.85[c]
Eu	0.020 (0.003)[a]	—	0.66-15.79[c]	1.66-18.87[c]
Gd	—	—	2.10-89.00[c]	8.18-61.43[c]
Tb	—	—	0.11-21.91[c]	0.98-12.55[c]
Dy	—	—	0.73-41.51[c]	1.50-17.63[c]
Ho	—	—	0.09-10.23[c]	0.24-3.11[c]
Er	—	—	0.18-34.32[c]	0.65-8.78[c]
Tm	—	—	0.07-4.84[c]	0.04-0.82[c]
Yb	—	—	0.36-29.63[c]	0.51-5.60[c]
Lu	—	—	0.02-3.36[c]	0.03-0.73[c]

[a] Pakistan, number in brackets assumed to be standard deviation of the mean, n≥6 [1]

[b] Spain [2]

[c] [3]

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Appendix B: Supplementary materials for Chapter 2

Table B1: Trace element background concentrations, standards, limits and ranges for soil, plants, animal and human health

Table is in long data format. Table includes some compounds and ions of relevant TEs where appropriate. Units are mg kg^{-1} unless otherwise stated.

Atomic number	Element	Attribute group	Attribute 1	Attribute 2	Attribute 3	Unit notes	Value [+]	Range minimum [+]	Range maximum [+]	Notes
1	H	Biosphere	Average crustal concentration				1400			
3	Li	Biosphere	Average crustal concentration				20			
3	Li	Soil	Geogenic concentration				30			
3	Li	Soil	Deficient	Total TE concentration				15	25	
3	Li	Soil	Deficient	Soluble TE concentration				0.1	2	
3	Li	Plant	Sufficient	Leaves			3			
3	Li	Plant	Excessive					5	50	
3	Li	Animal	MTL [*]	Rodents			25			
3	Li	Animal	MTL	Poultry			25			
3	Li	Animal	MTL	Swine			25			
3	Li	Animal	MTL	Horse			25			
3	Li	Animal	MTL	Cattle			25			
3	Li	Animal	MTL	Sheep			25			
4	Be	Biosphere	Average crustal concentration				2.8			
4	Be	Plant	Sufficient	Leaves				1	7	
4	Be	Plant	Excessive					10	50	
5	B	Biosphere	Average crustal concentration				10			
5	B	Soil	Geogenic concentration				10			
5	B	Soil	Typical concentration					2	150	
5	B	Plant	Deficient					5	30	
5	B	Plant	Sufficient	Leaves				10	100	
5	B	Plant	Sufficient	Shoots			20			
5	B	Plant	Excessive					50	200	
5	B	Plant	Tolerable in agronomic crops				100			
5	B	Human	LOAEL [#]			mg kg^{-1} d^{-1}	9.6			
5	B	Human	Tolerable upper limit			mg d^{-1}	20			
5	B	Animal	MTL	Rodents			150			
5	B	Animal	MTL	Poultry			150			
5	B	Animal	MTL	Swine			150			
5	B	Animal	MTL	Horse			150			
5	B	Animal	MTL	Cattle			150			
5	B	Animal	MTL	Sheep			150			
6	C	Biosphere	Average crustal concentration				200			
6	C	Soil	Geogenic concentration				20000			
7	N	Biosphere	Average crustal concentration				20			
7	N	Soil	Geogenic concentration				1000			
7	N	Plant	Sufficient	Shoots			15000			
8	O	Biosphere	Average crustal concentration				466000			
8	O	Soil	Geogenic concentration				490000			
9	F	Biosphere	Average crustal concentration				625			
9	F	Soil	Geogenic concentration				200			
9	F	Plant	Sufficient	Leaves				5	30	

9	F	Plant	Excessive					50	500
9	F	Animal	MTL	Rodents			150		
9	F	Animal	MTL	Poultry			150		
9	F	Animal	MTL	Swine			150		
9	F	Animal	MTL	Horse			40		
9	F	Animal	MTL	Cattle			40		
9	F	Animal	MTL	Sheep			60		
9	F-	Human	DRI					3	4
9	F-	Human	LOAEL			mg kg^{-1} d^{-1}	0.1		
9	F-	Human	Tolerable upper limit				10		
11	Na	Biosphere	Average crustal concentration				28300		
11	Na	Soil	Geogenic concentration				6300		
11	Na	Human	DRI $^{\%}$					1300	1500
11	Na	Human	Tolerable upper limit				2300		
11	NaCl	Animal	MTL	Rodents			40000		
11	NaCl	Animal	MTL	Poultry			17000		
11	NaCl	Animal	MTL	Swine			30000		
11	NaCl	Animal	MTL	Horse			60000		
11	NaCl	Animal	MTL	Cattle	Lactating		30000		
11	NaCl	Animal	MTL	Cattle	Growing		45000		
11	NaCl	Animal	MTL	Sheep			40000		
12	Mg	Biosphere	Average crustal concentration				20900		
12	Mg	Soil	Geogenic concentration				5000		
12	Mg	Plant	Sufficient	Shoots			2000		
12	Mg	Human	DRI					310	420
12	Mg	Human	LOAEL			mg d^{-1}	360		
12	Mg	Human	Tolerable upper limit				350		
12	Mg	Animal	MTL	Rodents			5000		
12	Mg	Animal	MTL	Poultry	Growing		5000		
12	Mg	Animal	MTL	Poultry	Laying		7500		
12	Mg	Animal	MTL	Swine			2400		
12	Mg	Animal	MTL	Horse			8000		
12	Mg	Animal	MTL	Cattle			6000		
12	Mg	Animal	MTL	Sheep			6000		
12	Mg	Animal	MTL	Fish			3000		
13	Al	Biosphere	Average crustal concentration				81300		
13	Al	Soil	Geogenic concentration				71000		
13	Al	Animal	MTL	Rodents			200		
13	Al	Animal	MTL	Poultry			1000		
13	Al	Animal	MTL	Swine			1000		
13	Al	Animal	MTL	Horse			1000		
13	Al	Animal	MTL	Cattle			1000		
13	Al	Animal	MTL	Sheep			1000		
14	Si	Biosphere	Average crustal concentration				277200		
14	Si	Soil	Geogenic concentration				330000		
14	Si	Animal	MTL	Cattle			2000		
14	Si	Animal	MTL	Sheep			2000		
15	P	Biosphere	Average crustal concentration				1050		
15	P	Soil	Geogenic concentration				650		
15	P	Plant	Sufficient	Shoots			2000		
15	P	Human	DRI					580	700
15	P	Human	LOAEL			mg d^{-1}	10200		

15	P	Human	Tolerable upper limit				4000		
15	P	Animal	MTL	Rodents			6000		
15	P	Animal	MTL	Poultry	Growing		10000		
15	P	Animal	MTL	Poultry	Laying		8000		
15	P	Animal	MTL	Swine			10000		
15	P	Animal	MTL	Horse			10000		
15	P	Animal	MTL	Cattle			7000		
15	P	Animal	MTL	Sheep			6000		
15	P	Animal	MTL	Fish			10000		
16	S	Biosphere	Average crustal concentration				260		
16	S	Soil	Geogenic concentration				700		
16	S	Plant	Sufficient	Shoots			1000		
16	S	Animal	MTL	Rodents			5000		
16	S	Animal	MTL	Poultry			4000		
16	S	Animal	MTL	Swine			4000		
16	S	Animal	MTL	Horse			5000		
16	S	Animal	MTL	Cattle	High-concentrate diet		3000		
16	S	Animal	MTL	Cattle	High-forage diet		5000		
16	S	Animal	MTL	Sheep	High-concentrate diet		3000		
16	S	Animal	MTL	Sheep	High-forage diet		5000		
17	Cl	Biosphere	Average crustal concentration				130		
17	Cl	Soil	Geogenic concentration				100		
17	Cl	Soil	Typical concentration					20	900
17	Cl	Soil	Deficient	Total TE concentration				70	200
17	Cl	Plant	Deficient					0	2000
17	Cl	Plant	Sufficient	Leaves				2000	20000
17	Cl	Plant	Sufficient	Shoots			100		
17	Cl-	Human	DRI					2000	2300
17	Cl-	Human	Tolerable upper limit				3600		
19	K	Biosphere	Average crustal concentration				25900		
19	K	Soil	Geogenic concentration				14000		
19	K	Plant	Sufficient	Shoots			10000		
19	K	Human	DRI				4700		
19	K	Animal	MTL	Rodents			10000		
19	K	Animal	MTL	Poultry			10000		
19	K	Animal	MTL	Swine			10000		
19	K	Animal	MTL	Horse			10000		
19	K	Animal	MTL	Cattle			20000		
19	K	Animal	MTL	Sheep			20000		
20	Ca	Biosphere	Average crustal concentration				36300		
20	Ca	Soil	Geogenic concentration				13700		
20	Ca	Plant	Sufficient	Shoots			5000		
20	Ca	Human	DRI					1000	1200
20	Ca	Human	LOAEL			mg d^{-1}	5000		
20	Ca	Human	Tolerable upper limit				2500		
20	Ca	Animal	MTL	Rodents			20000		
20	Ca	Animal	MTL	Poultry	Growing		15000		
20	Ca	Animal	MTL	Poultry	Laying		50000		
20	Ca	Animal	MTL	Swine			10000		

20	Ca	Animal	MTL	Horse			20000		
20	Ca	Animal	MTL	Cattle			15000		
20	Ca	Animal	MTL	Sheep			15000		
20	Ca	Animal	MTL	Fish			9000		
21	Sc	Biosphere	Average crustal concentration				22		
21	Sc	Animal	MTL	Rodents			100		
22	Ti	Biosphere	Average crustal concentration				4400		
22	Ti	Soil	Geogenic concentration				5000		
22	Ti	Plant	Excessive					50	200
23	V	Biosphere	Average crustal concentration				135		
23	V	Soil	Geogenic concentration				100		
23	V	Plant	Sufficient	Leaves				0.2	1.5
23	V	Plant	Excessive					5	10
23	V	Human	LOAEL			mg kg^{-1} d^{-1}	707		
23	V	Human	Tolerable upper limit				1.8		
23	V	Animal	MTL	Poultry	Growing		25		
23	V	Animal	MTL	Poultry	Laying			0	5
23	V	Animal	MTL	Swine			10		
23	V	Animal	MTL	Horse			10		
23	V	Animal	MTL	Cattle			50		
23	V	Animal	MTL	Sheep			50		
24	Cr	Biosphere	Average crustal concentration				100		
24	Cr	Soil	Geogenic concentration				100		
24	Cr	Plant	Sufficient	Leaves				0.1	0.5
24	Cr	Plant	Excessive					5	30
24	Cr	Plant	Tolerable in agronomic crops				2		
24	Cr	Human	DRI					0.02	0.035
24	Cr3+	Animal	MTL	Rodents			100		
24	Cr3+	Animal	MTL	Poultry			500		
24	Cr3+	Animal	MTL	Swine			100		
24	Cr3+	Animal	MTL	Horse			100		
24	Cr3+	Animal	MTL	Cattle			100		
24	Cr3+	Animal	MTL	Sheep			100		
24	CrO	Animal	MTL	Rodents			30000		
24	CrO	Animal	MTL	Poultry			3000		
24	CrO	Animal	MTL	Swine			3000		
24	CrO	Animal	MTL	Horse			3000		
24	CrO	Animal	MTL	Cattle			3000		
24	CrO	Animal	MTL	Sheep			3000		
25	Mn	Biosphere	Average crustal concentration				950		
25	Mn	Soil	Geogenic concentration				850		
25	Mn	Soil	Typical concentration					7	10000
25	Mn	Soil	Deficient	Total TE concentration				1000	3000
25	Mn	Soil	Deficient	Soluble TE concentration			43952		
25	Mn	Plant	Deficient					10	30
25	Mn	Plant	Sufficient	Leaves				20	300
25	Mn	Plant	Sufficient	Shoots			50		
25	Mn	Plant	Excessive					400	1000
25	Mn	Plant	Tolerable in agronomic crops				300		

25	Mn	Human	DRI				1.8	2.3
25	Mn	Human	LOAEL		mg d^{-1}	11		
25	Mn	Human	Tolerable upper limit			11		
25	Mn	Animal	MTL	Rodents		2000		
25	Mn	Animal	MTL	Poultry		2000		
25	Mn	Animal	MTL	Swine		1000		
25	Mn	Animal	MTL	Horse		400		
25	Mn	Animal	MTL	Cattle		2000		
25	Mn	Animal	MTL	Sheep		2000		
26	Fe	Biosphere	Average crustal concentration			50000		
26	Fe	Soil	Geogenic concentration			38000		
26	Fe	Soil	Typical concentration				200	500000
26	Fe	Soil	Deficient	Soluble TE concentration			2.5	5
26	Fe	Plant	Deficient				0	50
26	Fe	Plant	Sufficient	Leaves			50	500
26	Fe	Plant	Sufficient	Shoots		100		
26	Fe	Human	DRI	Female			8	18
26	Fe	Human	DRI	Male			5	8
26	Fe	Human	LOAEL		mg d^{-1}	70		
26	Fe	Human	Tolerable upper limit			45		
26	Fe	Animal	MTL	Rodents		500		
26	Fe	Animal	MTL	Poultry		500		
26	Fe	Animal	MTL	Swine		3000		
26	Fe	Animal	MTL	Horse		500		
26	Fe	Animal	MTL	Cattle		500		
26	Fe	Animal	MTL	Sheep		500		
27	Co	Biosphere	Average crustal concentration			25		
27	Co	Soil	Geogenic concentration			8		
27	Co	Plant	Sufficient	Leaves			0.02	1
27	Co	Plant	Excessive				15	50
27	Co	Plant	Tolerable in agronomic crops			5		
27	Co	Animal	MTL	Rodents		25		
27	Co	Animal	MTL	Poultry		25		
27	Co	Animal	MTL	Swine		100		
27	Co	Animal	MTL	Horse		25		
27	Co	Animal	MTL	Cattle		25		
27	Co	Animal	MTL	Sheep		25		
28	Ni	Biosphere	Average crustal concentration			75		
28	Ni	Soil	Geogenic concentration			40		
28	Ni	Soil	Typical concentration				3	1000
28	Ni	Plant	Deficient				0	0.1
28	Ni	Plant	Sufficient	Leaves			0.01	10
28	Ni	Plant	Sufficient	Shoots		0.1		
28	Ni	Plant	Excessive				10	100
28	Ni	Plant	Tolerable in agronomic crops				1	10
28	Ni	Human	LOAEL		mg kg^{-1} d^{-1}	5		
28	Ni	Human	Tolerable upper limit			1		
28	Ni	Animal	MTL	Rodents		50		
28	Ni	Animal	MTL	Poultry		250		
28	Ni	Animal	MTL	Swine		250		
28	Ni	Animal	MTL	Horse		50		

28	Ni	Animal	MTL	Cattle		100		
28	Ni	Animal	MTL	Sheep		100		
28	Ni	Animal	MTL	Fish		50		
29	Cu	Biosphere	Average crustal concentration			55		
29	Cu	Soil	Geogenic concentration			20		
29	Cu	Soil	Typical concentration				2	250
29	Cu	Soil	Deficient	Soluble TE concentration			0.1	2.5
29	Cu	Plant	Deficient				2	5
29	Cu	Plant	Sufficient	Leaves			3	30
29	Cu	Plant	Sufficient	Shoots		6		
29	Cu	Plant	Excessive				20	100
29	Cu	Plant	Tolerable in agronomic crops				5	20
29	Cu	Human	DRI				0.7	0.9
29	Cu	Human	LOAEL		mg d^{-1}	10		
29	Cu	Human	Tolerable upper limit			10		
29	Cu	Animal	MTL	Leaves for livestock consumption		300		
29	Cu	Animal	MTL	Rodents		500		
29	Cu	Animal	MTL	Ducks		500		
29	Cu	Animal	MTL	Poultry		250		
29	Cu	Animal	MTL	Swine		250		
29	Cu	Animal	MTL	Horse		250		
29	Cu	Animal	MTL	Cattle		40		
29	Cu	Animal	MTL	Sheep		15		
29	Cu	Animal	MTL	Fish		100		
30	Zn	Biosphere	Average crustal concentration			70		
30	Zn	Soil	Geogenic concentration			50		
30	Zn	Soil	Typical concentration				1	900
30	Zn	Soil	Deficient	Soluble TE concentration			0.1	2
30	Zn	Plant	Deficient				10	20
30	Zn	Plant	Sufficient	Leaves			15	150
30	Zn	Plant	Sufficient	Shoots		20		
30	Zn	Plant	Excessive				100	400
30	Zn	Plant	Tolerable in agronomic crops				50	100
30	Zn	Human	DRI	Female			6.8	8
30	Zn	Human	DRI	Male			9.4	11
30	Zn	Human	LOAEL		mg d^{-1}	60		
30	Zn	Human	Tolerable upper limit			40		
30	Zn	Animal	MTL	Leaves for livestock consumption		1000		
30	Zn	Animal	MTL	Rodents		500		
30	Zn	Animal	MTL	Poultry		500		
30	Zn	Animal	MTL	Swine		1000		
30	Zn	Animal	MTL	Horse		500		
30	Zn	Animal	MTL	Cattle		500		
30	Zn	Animal	MTL	Sheep		300		
30	Zn	Animal	MTL	Fish		250		
31	Ga	Biosphere	Average crustal concentration			15		
31	Ga	Soil	Geogenic concentration			30		
32	Ge	Biosphere	Average crustal concentration			1.5		

32	Ge	Animal	MTL	Rodents		30		
33	As	Biosphere	Average crustal concentration			1.8		
33	As	Soil	Geogenic concentration			6		
33	As	Plant	Sufficient	Leaves			0.01	7
33	As	Plant	Excessive				5	20
33	As	Plant	Tolerable in agronomic crops		FW	0.2		
33	As	Animal	MTL	Leaves for livestock consumption		50		
33	As	Animal	MTL	Rodents		30		
33	As	Animal	MTL	Poultry		30		
33	As	Animal	MTL	Swine		30		
33	As	Animal	MTL	Horse		30		
33	As	Animal	MTL	Cattle		30		
33	As	Animal	MTL	Sheep		30		
33	As	Animal	MTL	Fish		5		
34	Se	Biosphere	Average crustal concentration			0.05		
34	Se	Soil	Geogenic concentration			0.2		
34	Se	Plant	Sufficient	Leaves			0.01	2
34	Se	Plant	Excessive				5	30
34	Se	Human	DRI				0.045	0.055
34	Se	Human	LOAEL		mg d^{-1}	0.8		
34	Se	Human	Tolerable upper limit			0.4		
34	Se	Animal	MTL	Rodents		5		
34	Se	Animal	MTL	Poultry		3		
34	Se	Animal	MTL	Swine		4		
34	Se	Animal	MTL	Horse		5		
34	Se	Animal	MTL	Cattle		5		
34	Se	Animal	MTL	Sheep		5		
34	Se	Animal	MTL	Fish		2		
35	Br	Biosphere	Average crustal concentration			2.5		
35	Br	Soil	Geogenic concentration			5		
35	Br	Animal	MTL	Rodents		300		
35	Br	Animal	MTL	Poultry		2500		
35	Br	Animal	MTL	Swine		200		
35	Br	Animal	MTL	Horse		200		
35	Br	Animal	MTL	Cattle		200		
35	Br	Animal	MTL	Sheep		200		
37	Rb	Biosphere	Average crustal concentration			90		
37	Rb	Soil	Geogenic concentration			100		
37	Rb	Animal	MTL	Rodents		200		
38	Sr	Biosphere	Average crustal concentration			375		
38	Sr	Soil	Geogenic concentration			300		
38	Sr	Animal	MTL	Rodents		1000		
38	Sr	Animal	MTL	Poultry		2000		
38	Sr	Animal	MTL	Swine		2000		
38	Sr	Animal	MTL	Horse		2000		
38	Sr	Animal	MTL	Cattle		2000		
38	Sr	Animal	MTL	Sheep		2000		
38	Sr	Animal	MTL	Fish		33		
39	Y	Animal	MTL	Leaves for livestock consumption		100		
40	Zr	Biosphere	Average crustal concentration			165		

40	Zr	Soil	Geogenic concentration			300		
40	Zr	Plant	Excessive			15		
41	Nb	Biosphere	Average crustal concentration			20		
42	Mo	Biosphere	Average crustal concentration			1.5		
42	Mo	Soil	Geogenic concentration			2		
42	Mo	Soil	Typical concentration				0.1	40
42	Mo	Soil	Deficient	Total TE concentration			2	13
42	Mo	Soil	Deficient	Soluble TE concentration			0.1	0.3
42	Mo	Plant	Deficient				0.03	0.3
42	Mo	Plant	Sufficient	Leaves			0.1	5
42	Mo	Plant	Sufficient	Shoots		0.1		
42	Mo	Human	DRI				0.034	0.045
42	Mo	Human	LOAEL		mg kg^{-1} d^{-1}	0.9		
42	Mo	Human	Tolerable upper limit			2.7		
42	Mo	Animal	MTL	Rodents		7		
42	Mo	Animal	MTL	Poultry		100		
42	Mo	Animal	MTL	Swine		150		
42	Mo	Animal	MTL	Horse		5		
42	Mo	Animal	MTL	Cattle		5		
42	Mo	Animal	MTL	Sheep		5		
42	Mo	Animal	MTL	Fish		10		
44	Ru	Biosphere	Average crustal concentration			0.01		
45	Rh	Biosphere	Average crustal concentration			0.005		
46	Pd	Biosphere	Average crustal concentration			0.01		
47	Ag	Biosphere	Average crustal concentration			0.07		
47	Ag	Soil	Geogenic concentration			0.1		
47	Ag	Plant	Sufficient	Leaves		0.5		
47	Ag	Plant	Excessive				5	10
47	Ag	Animal	MTL	Poultry		100		
47	Ag	Animal	MTL	Swine		100		
47	Ag	Animal	MTL	Fish		3		
48	Cd	Biosphere	Average crustal concentration			0.2		
48	Cd	Soil	Geogenic concentration			0.06		
48	Cd	Plant	Sufficient	Leaves			0.05	0.2
48	Cd	Plant	Excessive				5	20
48	Cd	Plant	Tolerable in agronomic crops				0.05	0.5
48	Cd	Animal	MTL	Leaves for livestock consumption		0.5		
48	Cd	Animal	MTL	Rodents		10		
48	Cd	Animal	MTL	Poultry		10		
48	Cd	Animal	MTL	Swine		10		
48	Cd	Animal	MTL	Horse		10		
48	Cd	Animal	MTL	Cattle		10		
48	Cd	Animal	MTL	Sheep		10		
48	Cd	Animal	MTL	Fish		10		
49	In	Biosphere	Average crustal concentration			0.1		
50	Sn	Biosphere	Average crustal concentration			2		
50	Sn	Plant	Excessive			60		
50	Sn	Animal	MTL	Rodents		100		
50	Sn	Animal	MTL	Poultry		100		

50	Sn	Animal	MTL	Swine		100			
50	Sn	Animal	MTL	Horse		100			
50	Sn	Animal	MTL	Cattle		100			
50	Sn	Animal	MTL	Sheep		100			
51	Sb	Biosphere	Average crustal concentration			0.2			
51	Sb	Plant	Sufficient	Leaves			0.005 7	0.1 50	Different authors report different concentration ranges
51	Sb	Plant	Excessive			150			
51	Sb	Animal	MTL	Rodents			70	150	
52	Te	Biosphere	Average crustal concentration			0.01			
53	I	Biosphere	Average crustal concentration			0.5			
53	I	Soil	Geogenic concentration			5			
53	I	Human	DRI				0.095	0.150	
53	I	Human	LOAEL		mg d^{-1}	1.7			
53	I	Human	Tolerable upper limit			1.1			
53	I	Animal	MTL	Poultry		300			
53	I	Animal	MTL	Swine		400			
53	I	Animal	MTL	Horse		5			
53	I	Animal	MTL	Cattle		50			
53	I	Animal	MTL	Sheep		50			
55	Cs	Biosphere	Average crustal concentration			3			
55	Cs	Soil	Geogenic concentration			6			
56	Ba	Biosphere	Average crustal concentration			425			
56	Ba	Soil	Geogenic concentration			500			
56	Ba	Plant	Excessive			500			
56	Ba	Animal	MTL	Rodents		250			
56	Ba	Animal	MTL	Poultry		100			
56	Ba	Animal	MTL	Swine		100			
56	Ba	Animal	MTL	Horse		100			
57	La	Biosphere	Average crustal concentration			30			
57	La	Soil	Geogenic concentration			30			
57	La	Animal	MTL	Rodents		100			
58	Ce	Biosphere	Average crustal concentration			60			
58	Ce	Animal	MTL	Rodents		100			
59	Pr	Biosphere	Average crustal concentration			8.2			
59	Pr	Animal	MTL	Rodents		100			
60	Nd	Biosphere	Average crustal concentration			28			
60	Nd	Animal	MTL	Rodents		100			
61	Pm	Animal	MTL	Rodents		100			
62	Sm	Animal	MTL	Poultry		6			
62	Sm	Human	DRI			100			
63	Eu	Biosphere	Average crustal concentration			1.2			
63	Eu	Animal	MTL	Rodents		100			
64	Gd	Biosphere	Average crustal concentration			5.4			
64	Gd	Animal	MTL	Rodents		100			
65	Tb	Biosphere	Average crustal concentration			0.9			
65	Tb	Animal	MTL	Rodents		100			
66	Dy	Biosphere	Average crustal concentration			3			
66	Dy	Animal	MTL	Rodents		100			
67	Ho	Biosphere	Average crustal concentration			1.2			
67	Ho	Animal	MTL	Rodents		100			
68	Er	Biosphere	Average crustal concentration			2.8			

68	Er	Animal	MTL	Rodents		100		
69	Tm	Biosphere	Average crustal concentration			0.5		
69	Tm	Animal	MTL	Rodents		100		
70	Yb	Biosphere	Average crustal concentration			3.4		
70	Yb	Animal	MTL	Rodents		100		
71	Lu	Biosphere	Average crustal concentration			0.5		
71	Lu	Animal	MTL	Rodents		100		
72	Hf	Biosphere	Average crustal concentration			3		
73	Ta	Biosphere	Average crustal concentration			2		
74	W	Biosphere	Average crustal concentration			1.5		
74	W	Soil	Geogenic concentration			1		
74	W	Animal	MTL	Rodents		20		
74	W	Animal	MTL	Poultry		20		
74	W	Animal	MTL	Swine		20		
74	W	Animal	MTL	Horse		20		
74	W	Animal	MTL	Cattle		20		
74	W	Animal	MTL	Sheep		20		
75	Re	Biosphere	Average crustal concentration			0.001		
76	Os	Biosphere	Average crustal concentration			0.005		
77	Ir	Biosphere	Average crustal concentration			0.001		
78	Pt	Biosphere	Average crustal concentration			0.01		
79	Au	Biosphere	Average crustal concentration			0.004		
80	Hg	Biosphere	Average crustal concentration			0.08		
80	Hg	Soil	Geogenic concentration			0.03		
80	Hg	Plant	Excessive				1	3
80	Hg	Plant	Tolerable in agronomic crops		FW	0.2		
80	Hg	Animal	MTL	Rodents		1		
80	Hg	Animal	MTL	Poultry		1		
80	Hg	Animal	MTL	Swine		2		
80	Hg	Animal	MTL	Horse		1		
80	Hg	Animal	MTL	Cattle		2		
80	Hg	Animal	MTL	Sheep		2		
80	Hg	Animal	MTL	Fish		1		
80	Hg inorganic	Animal	MTL	Rodents		0.2		
80	Hg inorganic	Animal	MTL	Poultry		0.2		
80	Hg inorganic	Animal	MTL	Swine		0.2		
80	Hg inorganic	Animal	MTL	Horse		0.2		
81	Tl	Biosphere	Average crustal concentration			0.5		
81	Tl	Plant	Sufficient	Leaves		0.05		
81	Tl	Plant	Excessive			20		
82	Pb	Biosphere	Average crustal concentration			13		
82	Pb	Soil	Geogenic concentration			10		
82	Pb	Plant	Sufficient	Leaves			2	10
82	Pb	Plant	Excessive				30	300
82	Pb	Plant	Tolerable in agronomic crops				0.5	10
82	Pb	Animal	MTL	Leaves for livestock consumption		30		
82	Pb	Animal	MTL	Rodents		10		
82	Pb	Animal	MTL	Poultry		10		
82	Pb	Animal	MTL	Swine		10		
82	Pb	Animal	MTL	Horse		10		
82	Pb	Animal	MTL	Cattle		100		

82	Pb	Animal	MTL	Sheep	100
82	Pb	Animal	MTL	Fish	10
83	Bi	Biosphere	Average crustal concentration		0.2
83	Bi	Plant	Sufficient	Leaves	0.06
83	Bi	Animal	MTL	Rodents	500
83	Bi	Animal	MTL	Poultry	1000
83	Bi	Animal	MTL	Swine	500
83	Bi	Animal	MTL	Horse	500
90	Th	Biosphere	Average crustal concentration		7.2
92	U	Biosphere	Average crustal concentration		1.8
92	U	Animal	MTL	Rodents	100
92	U	Animal	MTL	Fish	100

[+] All figures in mg kg^{-1} DW unless stated in column 'Unit notes'

[*] MLT = Maximum tolerable level

[#] LOAEL = Lowest (or no) observed adverse effect level

[%] DRI = Dietary reference intake

References

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Appendix C: Supplementary materials for Chapter 3

Table C1: SRM recoveries of elements

Element	SRM recovery of certified value (%) – EPA method recoveries
Mg	136
Al	142
P	82
S	149
K	164
Ca	84
Cr	133
Mn	101
Fe	99
Co	137
Ni	107
Cu	99
Zn	104
Ge	No SRM – concentrations were comparable to crustal average of 1.5 mg kg^{-1} (Mason & Moore, 1982)
As	88
Sr	102
Mo	37
Pd	No SRM – concentrations were comparable to crustal average of 0.01 mg kg^{-1} (Mason & Moore, 1982)
Cd	68
Te	No SRM – concentrations were comparable to crustal average of 0.01 mg kg^{-1} (Mason & Moore, 1982)
Cs	118
Ce	99
Re	No SRM – concentrations were comparable to crustal average of 0.001 mg kg^{-1} (Mason & Moore, 1982)
Ir	No SRM – concentrations were comparable to crustal average of 0.001 mg kg^{-1} (Mason & Moore, 1982)
Hg	74
Pb	102

Table C2: $\mathrm{Ca(NO_3)_2}$ extractable concentrations of trace elements in soils under palm production

Table C2. Mean $\mathrm{Ca(NO_3)_2}$ extractable TE concentrations of soils at each site (SE in parentheses). Mean concentrations in mg kg^{-1}. Sites which share the same letter(s) for a single variable are not significantly different from each other.

	C (n=10)	D (n=25)	A (n=25)	B (n=25)
Cr	0.0093(0.0023) ^{a}	0.0086(0.0015) ^{a}	0.011(0.0017) ^{a}	0.0094(0.0019) ^{a}
Mn	53(11) ^{c}	4.5(0.85) ^{a}	27(7.1) ^{b}	54(8.5) ^{c}
Co	0.0068(0.0015) ^{a}	0.022(0.0044) ^{a}	0.067(0.015) ^{b}	0.095(0.014) ^{b}
Ni	0.0096(0.0052) ^{a}	0.30(0.057) ^{a}	0.61(0.092) ^{b}	1.1(0.13) ^{c}
Cu	<0.000 ^{a}	0.0012(0.0008) ^{a}	0.021(0.0050) ^{a}	0.11(0.016) ^{b}
Zn	0.43(0.11) ^{a}	0.36(0.062) ^{a}	1.3(0.19) ^{b}	2.1(0.36) ^{c}
Ge	0.0022(0.00042) ^{ab}	0.0036(0.00089) ^{ab}	0.0013(0.00041) ^{a}	0.0078(0.0020) ^{b}
As	0.060(0.024) ^{ab}	0.060(0.015) ^{ab}	0.030(0.012) ^{a}	0.092(0.015) ^{b}
Sr	0.57(0.24) ^{ab}	0.17(0.037) ^{a}	1.0(0.13) ^{c}	0.52(0.099) ^{b}
Mo	0.0030(0.0030) ^{ab}	0.010(0.0028) ^{b}	0.0036(0.0020) ^{ab}	0.0012(0.0012) ^{a}
Cd	0.013(0.0029) ^{ab}	0.013(0.0010) ^{a}	0.019(0.0036) ^{ab}	0.027(0.0044) ^{b}
Ce	0.089(0.018) ^{ab}	0.16(0.030) ^{b}	0.033(0.0089) ^{a}	0.22(0.052) ^{b}
Pb	0.11(0.016) ^{ab}	0.15(0.023) ^{ab}	0.089(0.011) ^{a}	0.22(0.041) ^{b}

Table C3: Additional trace elements analysed

Table C3: Concentrations of total and $\mathrm{Ca(NO_3)_2}$ extractable Pd, Te, Re, Ir and Hg at each site, with total Ag and Cs at Site E

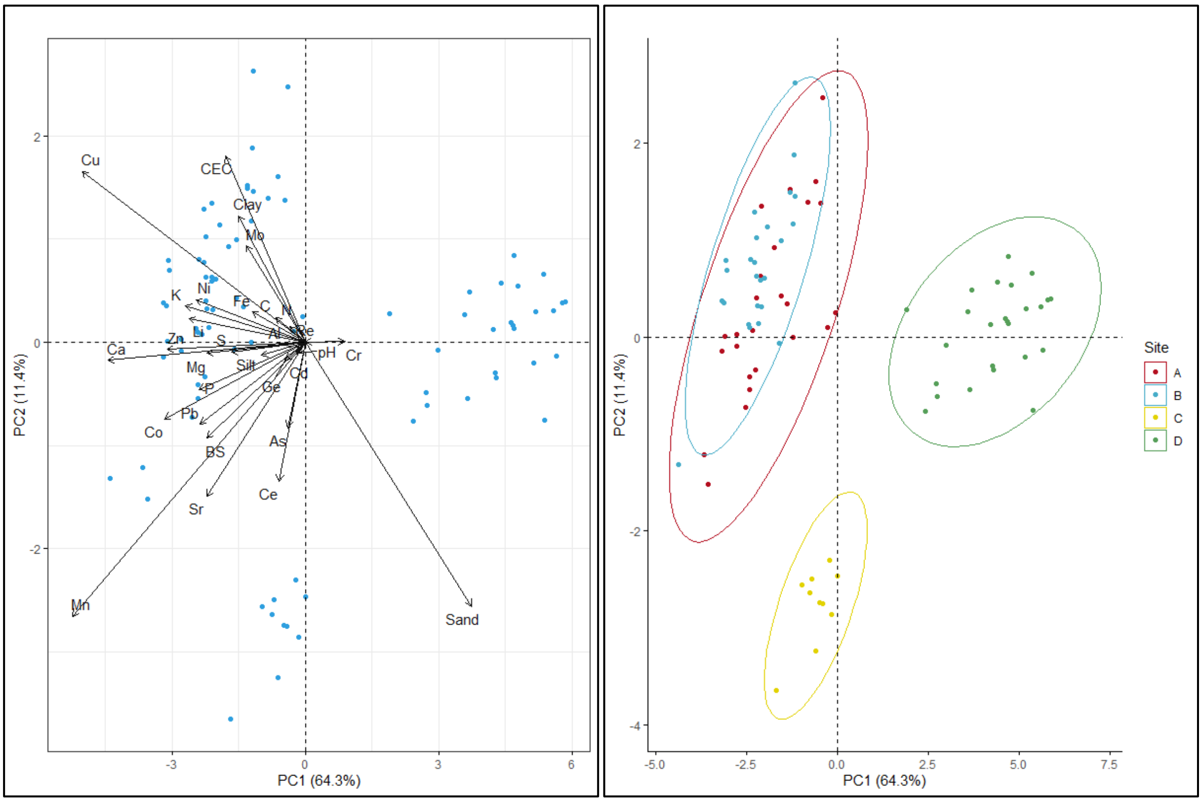
Site		**A (n=25)**	**B (n=25)** $^{+}$	**C (n=10) ***	**D (n=25)**	**E (n=32)**
Pd	**Total (mg kg^{-1})**	0.074(0.012) ^{b}	0.051(0.015) ^{b}	0.039(0.0052) ^{ab}	0.013(0.0015) ^{a}	—
	$\mathrm{Ca(NO_3)_2}$ extractable (mg kg^{-1})	<0.01 ^{a}	<0.01 ^{b}	<0.01 ^{ab}	<0.01 ^{a}	—
Te	**Total (mg kg^{-1})**	0.14(0.098) ^{a}	0.030(0.0033) ^{a}	0.019(0.0010) ^{a}	<0.01 ^{a}	—
	$\mathrm{Ca(NO_3)_2}$ extractable (mg kg^{-1})	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	—
Cs		—	—	—	—	8.2(0.10)
Re	**Total (mg kg^{-1})**	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{b}
	$\mathrm{Ca(NO_3)_2}$ extractable (mg kg^{-1})	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	—
Ir		<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	0.0013(0.0004) ^{a}
Hg	**Total (mg kg^{-1})**	0.054(0.0090) ^{c}	0.030(0.0038) ^{b}	0.031(0.0045) ^{abc}	0.0092(0.0014) ^{a}	0.10(0.003) ^{d}
	$\mathrm{Ca(NO_3)_2}$ extractable (mg kg^{-1})	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	<0.01 ^{a}	—

$^{+}$ Site B Pd, Te, Ir, Hg n=23

* Site C Pd, Te, Ir, Hg n = 8

Figure C1: PCA results of four palm sites including general soil parameters

Principal component one explains 64.3% of variance and is dominated by Mn, Cu, Ca, Sand, Co, Zn, K, Ni, P and Pb in descending order, while principal component two is dominated by Mn, Sand, CEC, Cu, Sr, Ce and Clay and explains 11.4% of variance. A mix of edaphic and anthropogenic factors contribute to the soil chemistry at these sites.



Figures C1a and b: Loading plot (a) and PCA plot (b) of palm sites including general soil parameters

References

Mason, B. H., & Moore, C. B. (1982). Principles of geochemistry. Wiley. https://books.google.co.nz/books?id=K75gIAAACAAJ

Appendix D: Supplementary materials for Chapter 4

Table D1. SRM recoveries of elements

Element	SRM recovery of certified value (%)
B	105
Na	119
Mg	88
Al	79
Si	No SRM — concentrations were comparable to land plants average of 200 mg kg^{-1} (Mason & Moore, 1982)
P	95
S	134
K	98
Ti	No SRM — concentrations were comparable to land plants average of 1.0 mg kg^{-1} (Mason & Moore, 1982)
Cr	97
Mn	109
Fe	103
Co	99
Co	116
Ni	99
Cu	100
Zn	97
As	102
Sr	110
Zr	No SRM
Mo	98
Ag	134
Cd	94
Te	76
Cs	107
Ce	84
Au	153
Hg	97
Pb	No SRM — concentrations were comparable to land plants average of 2.7 mg kg^{-1} (Mason & Moore, 1982)

References

Mason, B. H., & Moore, C. B. (1982). Principles of geochemistry. Wiley. https://books.google.co.nz/books?id=K75gIAAACAAJ

Appendix E: Supplementary materials for Chapter 5

Table E1. Sampling locations for pasture samples

Sample ID	Latitude	Longitude
1	172.5183	-43.4047
2	172.5199	-43.4037
3	172.5178	-43.4025
4	172.0957	-43.7039
5	172.1018	-43.6971
6	172.0933	-43.6991
7	172.0758	-43.6864
8	172.0799	-43.6862
9	172.093	-43.7429
10	172.084	-43.7463
11	172.0857	-43.7427
12	171.882	-43.5804
13	171.8667	-43.5852
14	171.8672	-43.5908
15	171.787	-43.7693
16	171.7775	-43.7717
17	171.7627	-43.7621
18	171.5686	-43.9918
19	171.5648	-43.993
20	171.5565	-43.9849
21	171.4787	-43.9227
22	171.4773	-43.9248
23	171.4744	-43.9243
24	171.7498	-43.8461
25	171.7599	-43.8548
26	171.7575	-43.8495
27	171.8912	-43.8051
28	171.8961	-43.8144
29	171.8973	-43.8159
30	172.4548	-43.63
32	172.4587	-43.631
33	172.4614	-43.6411
34	172.4576	-43.6413
35	171.2907	-43.9328
36	171.2882	-43.8997
37	171.2825	-43.9046
38	171.7394	-43.7456
39	171.7356	-43.7501
40	171.7303	-43.753
41	170.872	-43.8581

Table E1 continued

Sample ID	Latitude	Longitude
42	170.8934	-43.8741
43	170.9024	-43.8931
44	171.1665	-44.1245
45	171.1632	-44.126
46	171.1605	-44.1249
47	170.9692	-44.3635
48	170.9697	-44.3641
49	170.9594	-44.5147
50	170.9573	-44.5186
51	170.9589	-44.5203
52	171.0192	-44.4341
53	171.0191	-44.4365
54	171.0095	-44.4392
55	171.2962	-44.2946
56	171.2953	-44.2984
57	171.29	-44.2979
58	171.2706	-43.9858
59	171.2744	-43.9899
60	171.2766	-43.9932
61	173.0189	-43.6863
62	173.0255	-43.6804
63	173.0316	-43.6804
64	173.0351	-43.6789
65	170.8765	-44.4172
66	170.874	-44.4222
67	170.8759	-44.4294
68	170.7899	-45.4791
69	170.7845	-45.4808
70	170.7773	-45.4882
71	170.6736	-45.4478
78	170.7055	-45.0085
79	170.7049	-45.0008
80	170.7107	-45.0027
81	170.6299	-44.9629
82	170.6303	-44.9645
83	170.6178	-44.9656
84	171.0486	-44.908
85	171.0404	-44.9037
86	171.0397	-44.9111

Table E2. SRM recoveries of elements

Element	SRM recovery of certified value (%)
B	84
Na	95
Mg	101
Al	97
P	91
S	105
K	117
Ti	No SRM
Cr	100
Mn	104
Fe	102
Co	99
Co	109
Ni	108
Cu	110
Zn	106
As	109
Sr	100
Zr	No SRM – concentrations were comparable to land plants average of 0.64 mg kg^{-1} [1]
Mo	101
Ag	150
Cd	102
Te	73
Cs	112
Ce	73
Au	156
Hg	91
Pb	100

References

1. B. H. Mason and C. B. Moore, *Principles of geochemistry*, Wiley, 1982.

Appendix F: Supplementary materials for Chapter 6

F1: Model R code

Input variables

The following input variables are required to run the model. The values entered here are purely examples of values which may be used.

```
# Global Variables
iTime <- 50
iSimulationDepth <- 100
iRootDepth <- 60
iIncorporationDepth <- 1
iRainfall <- 800
iET <- 500
iSoilDensity <- 1.2
iSoilLoss <- 3
iSliceWaterMass <- 50
iAtmosDeposition <- 0
iDryBiomassProduced <- 10
iPercentRemoved <- 50
iK <- 0.1

# Contaminant-specific variables
iInitialSoilCon <- 0.5
iInitialCropCon <- 0.2
iKdSoil <- 500
iConApplied <- 400

iConAdded <- iAtmosDeposition + iConApplied

#Output variables
SoilCon <<- c(rep(0,iTime))
PlantData <<- c(rep(0,iTime))
Leached <<- c(rep(0,iTime))
FinalProfile <<- c(rep(0,iSimulationDepth))

#Mass Balance variables
MassAdded <- 0
MassInPlants <- 0
MassLeached <- 0
MassRunoff <- 0
MassInSoilStart <- 0
MassInSoilFinish <- 0
```

Model processes

The section headings below correspond to numbered sections in the body of the text.

```
# 2.2.1 Initialisation

SoilMass <- iSoilDensity*100
ConMass <- SoilMass*iInitialSoilCon

for (i in 1:iSimulationDepth)
{
  SoilMass[i] <- iSoilDensity * 100
  TotalSoilMass <- sum(SoilMass)
  ConMass[i] <- SoilMass[i] * iInitialSoilCon
  MassInSoilStart <- sum(ConMass)
}

# Solution mass function

# Used in drainage and plant uptake
SolutionMass <- function(i)
{
  ((ConMass[i] / SoilMass[i]) / iKdSoil) * iSliceWaterMass
}

InitSolnConc <- SolutionMass(1)/iSliceWaterMass
RAF1 <- iInitialCropCon / (iInitialSoilCon / iKdSoil)

# 2.2.2 Incorporation

Incorporate <- function(ContaminantMass,IncorporationDepth)
{
  SliceConAdded <- rep(c(ContaminantMass / IncorporationDepth, 0),
                    c(IncorporationDepth, iSimulationDepth-IncorporationDepth))
  ConMass <<- ConMass + SliceConAdded
}


# 2.2.3 Soil loss

SoilLoss <- function()
{
  SoilLossFract <- pmin(iSoilLoss / SoilMass[1],1)
  SoilConLoss <- ConMass[1] * SoilLossFract
  ConMass[1] <<- ConMass[1] - SoilConLoss
  MassRunoff <<- MassRunoff + SoilConLoss
```

```
}

# 2.2.4 Drainage & leaching

Drainage <- function(HoldX)
{
 Leached <- 0
 MobileCon <-0
 RainfallMass <- 10*pmax(iRainfall-iET,0)
 Loops <- trunc(RainfallMass / iSliceWaterMass)
 for (steps in 1: Loops)
 {

  for (i in 1: iSimulationDepth)
  {
   SLNMS <- SolutionMass(i)
   ConMass[i] <<- ConMass[i] - SLNMS
   if (i < iSimulationDepth)
   {
    ConMass[i + 1] <<- ConMass[i + 1] + SLNMS
   }
   else
   {
    Leached <- Leached + SLNMS
   }

  }
 }
 Leached[HoldX] <<- Leached
 MassLeached <<- MassLeached + Leached
}

# 2.2.5 Plant Uptake

PlantUptake <- function(HoldX)
{
 BiomassRemoved <- iPercentRemoved / 100
 PlantConMass <- 0
 RemovedFromSlice <- 0
 for (i in 1: iRootDepth)
 {
  SolnConc <- SolutionMass(i)/iSliceWaterMass
  RAF <- (RAF1 * InitSolnConc) / (InitSolnConc + iK * (SolnConc - InitSolnConc))
  RemovedFromSlice <- (SolnConc * RAF * iDryBiomassProduced) / iRootDepth
  PlantConMass <- PlantConMass + RemovedFromSlice
```

```
   ConMass[i] <<- ConMass[i] - RemovedFromSlice
  }
  PlantData[HoldX] <<- (PlantConMass / iDryBiomassProduced)
  MassInPlants <<- MassInPlants + PlantConMass * BiomassRemoved
  Incorporate(PlantConMass * (1 - BiomassRemoved), iIncorporationDepth)
}
```

The main loop

The following loop combines all of the model processes and runs the code for the specified scenario.

```
# The main loop

MainLoop <- function(iRootDepth, iRainfall, iET, iSoilDensity, iKdSoil,
                iConApplied, iSoilLoss, iInitialCropCon, iInitialSoilCon,
                iDryBiomassProduced, iPercentRemoved, iSliceWaterMass, iAtmosDeposition)
{
  for (year in 1: iTime)
  {
   #Incorporation
   MassAdded <<- MassAdded + iConAdded
   Incorporate(iConAdded, iIncorporationDepth)
   SoilLoss()
   Drainage(year)
   PlantUptake(year)
   #annual mass sum
   Mass <- sum(ConMass)
   for (i in 1: iSimulationDepth)
   {
    SoilCon[year] <<- (Mass / TotalSoilMass)
   }
  }
  MassInSoilFinish <<- sum(ConMass)
  for (i in 1: iSimulationDepth)
  {
   FinalProfile[i] <<- (ConMass[i] / SoilMass[i])
  }
  checksum <<- (MassInSoilFinish + MassLeached + MassInPlants + MassRunoff) - (MassInSoilStart +
MassAdded)
}

MainLoop()
```

F2: Inputs used for model validation specific to the Winchmore fertiliser trials

Table F1: Site-specific model input parameters

General parameters	
Simulation time (yr)	[contaminant specific]
Simulation depth (cm)	100
Rainfall and irrigation (mm yr^{-1})	1175 [a, b]
Evapotranspiration (mm yr^{-1})	919 [c]
Soil parameters	
Initial contaminant concentration (mg kg^{-1})	[contaminant specific]
Erosive loss (t ha^{-1} yr-^{1})	1.67 [d]
KD	[contaminant specific]
Bulk density (t m3 $^{-1}$)	1.06 [e]
Porosity (%)	50 [f]
Plant parameters	
Dry biomass produced (t ha^{-1} yr^{-1})	188 trial plots: 11 [g]
	376 trial plots: 12 [g]
Biomass removed from site (%)	43 [h]
Root depth (cm)	56 [i]
Initial contaminant concentration (mg kg^{-1})	[contaminant specific]
Decay constant	[contaminant specific]
Contaminant parameters	
Mass applied (g ha^{-1} yr^{-1})	[contaminant specific]
Atmospheric deposition (g ha^{-1} yr^{-1})	[contaminant specific]
Incorporation depth (cm)	1

[a] Irrigation data from McDowell et al. (2021)

[b] Rainfall data from Gray and Moss (2021).

[c] Macara (2016)

[d] Stats NZ (2018)

[e] Srinivasan and McDowell (2009)

[f] Typical soil porosity (Negassa et al., 2015)

[g] Site-specific information measured in trial, sourced from the Winchmore dataset

[h] Pasture removed through grazing (Betteridge et al., 1994)

[I] Weighted average of ryegrass white clover pasture (Birendra et al., 2020; Reiser et al., 2014; Roberts & Longhurst, 2002)

Table F2: Contaminant-specific model input parameters

Contaminant name	Trial plot	Initial soil concentration (mg kg^{-1}) [a]	Initial plant concentration (mg/kg)	K_D	Decay constant	Mass of contaminant added (g ha^{-1} yr^{-1})
Fluorine	188	251	0.5 [b]	365 [c]	0	2660 [d]
	376	251	0.5 [b]	365 [c]	0	5320 [d]
Cadmium	188	0.055	0.077 [e]	3000 [f]	0	3.0 [a]
	376	0.049	0.077 [e]	3000 [f]	0	6.0 [a]
Uranium	188	1.1	0.014 [g]	4500 [h]	0	9.8 [d]
	376	1.2	0.014 [g]	4500[h]	0	20 [d]

[a] Site-specific information measured in trial, sourced from the Winchmore dataset

[b] Mason and Moore (1982)

[c] Mean K_D of F in silt loam soils (Nakamura et al., 2017)

[d] Taylor et al. (2016)

[e] Weighted mean of pasture species from Stafford et al. (2016)

[f] Reiser et al. (2014)

[g] Average U concentration of 40 crops converted to mg kg^{-1} (Evans & Eriksson, 1983)

[h] USEPA (1999)

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