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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

Hadee Thompson-Morrison[1,2] | **Elena Moltchanova**[3] | **Sally Gaw**[1] | **Brett Robinson**[1]

[1]School of Physical and Chemical Sciences, University of Canterbury, Christchurch, New Zealand

[2]Current address: Manaaki Whenua Landcare Research, Lincoln, New Zealand

[3]School of Mathematics and Statistics, University of Canterbury, Christchurch, New Zealand

Correspondence
Hadee Thompson-Morrison, School of Physical and Chemical Sciences, University of Canterbury, Christchurch, New Zealand.
Email: hadee.thompson-morrison@protonmail.com

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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 agriculture in Indonesia occurs on weathered, acidic soils that require substantial fertilizer inputs to maintain production. To understand whether TE accumulation in soils resulting from fertilizer 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 which occur in the soil–plant system and involve the transport of TEs. It was developed to be rationally convenient, that is, parsimonious, 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 soil- and-crop specific parameters. We validated our model using a data set from a 64-year fertilizer trial, which had known inputs and application rates of fluorine (F), cadmium (Cd) and uranium (U). We applied the validated model to a smallholder oil palm plantation in Indonesia, modelling accumulation of copper (Cu), F, 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 Cu and F were likely to reach phytotoxic concentrations in soils (289 and 719 mg kg^{-1}, respectively), within 50 years under these conditions. The future feasibility of intensive oil palm production on weathered, low fertility soils such as those in Indonesia should be assessed to avoid long-term negative impacts on soil quality.

KEYWORDS

mass-balance, model, oil palm, rhizosphere processes, trace elements

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1 | INTRODUCTION

Most agricultural production requires applications of agrichemicals and amendments to promote crop growth and aid soil fertility. These include fertilizers, pesticides and organic amendments, such as composts. Many agrichemicals contain trace elements (TEs) as either active ingredients (e.g. copper (Cu) fungicides) or contaminants. Phosphatic fertilizers may contaminate soil with TEs because of the impurities, especially fluorine (F), cadmium (Cd) and uranium (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, manures and composts may contain elevated levels of these and other TEs (Bolan et al., 2004; European Commission, 2004), while pesticides may contain TEs as active ingredients, particularly Cu, zinc (Zn) and manganese (Mn) (Gimeno-García et al., 1996; Rocha et al., 2014). Additionally, water used for irrigation may contain contaminants such as arsenic (As) and constitute a source of TEs to production soils (Bhatti et al., 2013).

Agrichemical application rates and type vary by system: for example, in dairy farms, the predominant input is nitrogen (N) at recommended rates (for New Zealand pasture) of 25–50 $kg\,ha^{-1}\,year^{-1}$, while in horticultural systems, the dominant agrichemicals are pesticides containing TEs such as Cu at rates of ≤16 $kg\,ha^{-1}\,year^{-1}$ (Bivi et al., 2016; Fungicide Resistance Action Committee, 2021; Morgan & Taylor, 2004; 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 systems, recommended rates of N, phosphorus (P) and potassium (K) are 218–286, 143–215 and 358–429 $kg\,ha^{-1}$, respectively (Woittiez et al., 2015). Varying rates of Cd are applied through P fertilizers because of 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 copper sulphate ($CuSO_4$) 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 owing 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). Cadmium is primarily added to soils through phosphate fertilizer application, along with F and U (Taylor et al., 2016). Other TEs that may accumulate in soils as a result of agrichemical use include Zn, As and lead (Pb). 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 & Kim, 2009). This produces unfavourable conditions for plant growth because of Al^{3+} toxicity (Taylor et al., 2012). 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 oil palm plantations from tropical forest in Southeast Asia often has low fertility and requires high fertilizer inputs, particularly P and K, to support production (Corley & Tinker, 2015; Woittiez et al., 2015). High applications of P fertilizers are associated with an increased soil Cd burden (Roberts, 2014). Pesticides, including Cu-based fungicides, are recommended to control plant pests common in oil palm production including *Ganoderma* spp. (Bivi et al., 2016). In contrast to traditional oil palm growing systems in parts of Africa (Corley & Tinker, 2015), oil palm production in Southeast Asia has undergone significant expansion since 1989 (Vijay et al., 2016) and most active plantations can be considered new systems that are naïve to fertilizer use. Soils from Indonesian oil palm growing regions are deficient in N, P and K, likely because of insufficient fertilizer application (Thompson-Morrison et al., 2022a; Woittiez et al., 2015). Oil palm agriculture and fertilizers are subsidized in Indonesia, and resultingly, fertilizer applications are often limited to subsidized products (Kissinger, 2016; Woittiez et al., 2018). This has led to low levels of key nutrients that are not available in subsidized forms, such as K, in Indonesian soils (Thompson-Morrison et al., 2022a;

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Woittiez et al., 2015). Soils analysed by Thompson-Morrison et al. (2022a) from active and abandoned oil palm plantations in Indonesia displayed characteristics typical of soils with limited fertilizer 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, Zn, As and Pb was noted and attributed to agrichemical use. There is potential that increased fertilizer application in oil palm 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 oil palm 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 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 which specifies the parameter. All 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 oil palm 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 1 were sufficient for our purposes or had been validated. Our desired model should also be 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. The objectives of this study were therefore to develop a whole-system model that met these requirements; to validate the model using data from long-term agricultural trials with known sources and rates of TE inputs to soils over time; and to use the model to calculate the TE concentrations likely to occur under oil palm production in Southeast Asian soils in order to understand likely timeframes and limiting TEs of any accumulation.

2 | METHODS

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; 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 1). The model takes a total of 17 input parameters specified by the user, divided into general, soil, plant and contaminant categories (Table 2). These processes are treated as one-dimensional, that is, 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.

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 1 over time at different depths. Below, we describe

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TABLE 1 Comparison of parameters considered in mass-balance models for the determination of soil fertilizer-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, fertilizer and 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), fertilizer application rate, Cd fertilizer content, crop yield, calculated crop concentration, literature-based Cd leaching estimates
De Vries and McLaughlin (2013), Australia	Cd	Numeric	Atmospheric deposition, fertilizer, biosolids and 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, run-off and pH *($CaCl_2$)*
Sheppard et al. (2009), Canada	Cu, Zn, As, selenium (Se), Cd and Pb	Analytical	Atmospheric deposition, fertilizer, biosolids and animal feed	Harvested crop offtake, leaching (using K_D), volatilization (Se only) and bioturbation	30 cm	K_D, soil bulk density (specified) and crop yield
ERM (2000), as cited in Cavanagh (2014), European Union	Cd (possibly others)	Analytical	Atmospheric deposition, fertilizer, biosolids and animal feed	Harvested crop offtake (estimated) and leaching (using K_D)	20–30 cm (plough depth)	K_D, soil bulk density (specified), soil background concentration, precipitation, infiltration, plough depth and crop TE uptake coefficient
Michaud et al. (2019), France	Cr, nickel (Ni), Cu, Zn, Cd, mercury (Hg) and Pb	(Not reported)	Organic waste products and atmospheric deposition	Leaching and harvested crop offtake (measured)	<45 cm	Bulk density
Chang et al. (2004), United States	As, Cd	Analytical	Fertilizer and irrigation	Leaching and 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 and root permeability coefficient

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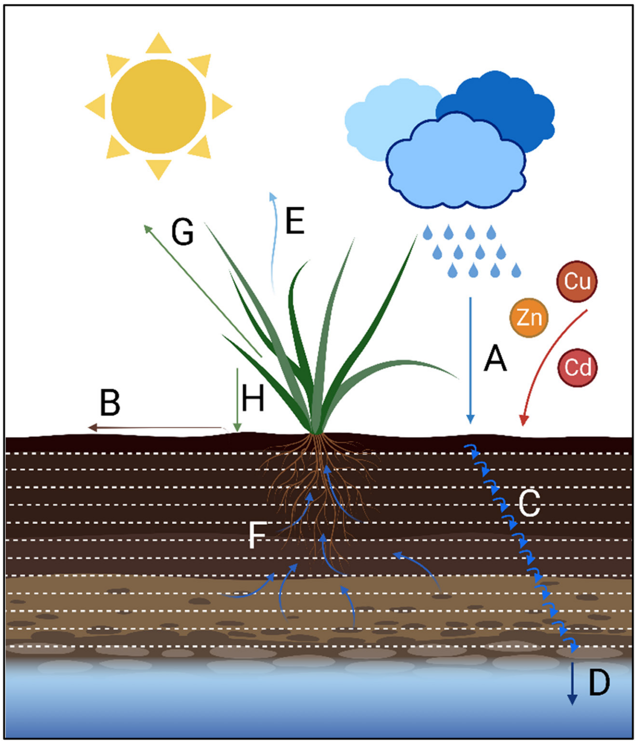

FIGURE 1 Key TE interactions in soil–plant systems. (A) Rainfall (and irrigation) water infiltrates the soil, while TEs are added to the soil surface through various mechanisms (fertilizer and amendment application, atmospheric deposition, presence in irrigation water). (B) Soil erosion may remove TEs 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 mobilizes and transports TEs downwards. The amount of contaminant mobilized 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).

the modelling process step by step. The full model code can be found in Appendix S1.

2.2 | Model processes

2.2.1 | Initialization

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 initialized as follows.

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

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

and the initial TE mass at depth d as

$$M_{0,d} = S_d C_0, \tag{2}$$

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

$$M_{0,\bullet} = \sum_{d=1}^{D} M_{0,d}. \tag{3}$$

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

$$M^+_{y,d} = \begin{cases} M^+_{y,\bullet}/I & \text{for } d = 1, \ldots, D_I \\ 0 & \text{for } d > D_I, \end{cases} \tag{4}$$

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

$$M_{y,d} = M_{y-1,d} + M^+_{y,d}. \tag{5}$$

In the current model, we assume the amount of added contaminant to be constant over the years, that is, $M^+_{y,\bullet} = \text{constant}$. 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).

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 run-off. 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 because of run-off in the top layer is evaluated as

$$E^{(f)} = \min\left(\frac{E}{S_1}, 1\right). \tag{6}$$

The TE mass in the top layer is then updated as

$$M_{y,1} = M_{y,1}\left(1 - E^{(f)}\right). \tag{7}$$

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TABLE 2 Model input parameters with relevant units and associated information.

Notation	Code ID	Parameters	Unit	Notes
General parameters				
y	iTime	Simulation time	year	
D	iSimulationDepth	Simulation depth	cm	Must be ≥ root depth
T	iRainfall	Rainfall and irrigation	mm year^{-1}	Rainfall infiltrates soil and is taken up by plants down to root depth
V	iET	Evapotranspiration	mm year^{-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} year^{-1} (equiv)	Mass of soil lost to erosion and run-off
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		$0 \le K < 0.5$
Contaminant parameters				
M	iConApplied	Mass applied	g ha^{-1} year^{-1}	Mass of contaminant applied in amendments, irrigation water etc
[NA]	iAtmosDeposition	Atmospheric deposition	g ha^{-1} year^{-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

The annual run-off soil loss is thus

$$M_y^{(R)} = M_{y,1} E^{(f)}. \tag{8}$$

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 reaches 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

$$T^{(m)} = 10 * \max(T - V, 0). \tag{9}$$

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

$$L = \left\lfloor T^{(m)} / W \right\rfloor, \tag{10}$$

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 mobilized by the downward movement of water and is thus removed from that soil layer d and added to the next one $d + 1$:

$$M_{y,d} = M_{y,d} - M_{y,d}^{(s)} \qquad \text{for } d = 1, \ldots, D \tag{11}$$

and

$$M_{y,d+1} = M_{y,d+1} + M_{y,d}^{(s)} \qquad \text{for } d = 1, \ldots, D, \tag{12}$$

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

$$M_{y,d}^{(s)} = \frac{M_{y,d} / S_d}{K} W. \tag{13}$$

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).

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

$$\varnothing = \frac{i}{C_0 / K}. \tag{14}$$

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 following Robinson et al. (2003):

$$\varnothing(H) = \frac{\varnothing_1 H_1}{H_1 + N\left(H - H_1\right)} \tag{15}$$

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

$$M_{y,d}^{-} = \begin{cases} \dfrac{M_{y,d}^{(s)} \varnothing(H) B}{D_R W} & \text{for } d = 1, \ldots, D_R \\ 0 & \text{for } d > D_R \end{cases} \tag{16}$$

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, that is, essentially the pore space of the soil. The values for the contaminant mass in the soil can be updated as

$$M_{y,d} = M_{y,d} - M_{y,d}^{-}. \tag{17}$$

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

$$P_y^{(s)} = \sum_d M_{y,d}^{-}. \tag{18}$$

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

$$M_{y,d} = M_{y,d} - (1 - B) P_y^{(s)} / D_I \quad \text{for } d = 1, \ldots, D_I, \tag{19}$$

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

$$M_y^{(P)} = P_y(s) B. \tag{20}$$

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

$$M_{y,d} = M_{y,d} - M_{y,d}^{-}. \tag{21}$$

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:

$$\sum_d M_{Y,d} = \sum_d M_{0,d} + \sum_y M_{y,\bullet}^{+} - \sum_y \left(M_y^{(P)} + M_y^{(L)} + M_y^{(R)} \right). \tag{22}$$

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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.

2.3 | Other relevant model assumptions and limitations

The model has several key assumptions to facilitate parameterization. Volatilization of TEs 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: that is, 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, for example, a change in soil bulk density occurring over the simulation time. All input variables are assumed constant throughout the course of the simulation.

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 fertilizer 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 fertilizer regimes over time (Rickard & Moss, 2012). The Winchmore trials have a known fertilizer history, including known contaminant concentrations of the fertilizers 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 the best of our knowledge, no such sites with known histories exist in tropical regions where oil palm is grown. Therefore, we used the Winchmore data set to validate the model before applying it to tropical oil palm systems.

At the Winchmore trial site, two trials were run with single superphosphate applied to pastoral soil plots at rates of 188 and 376 $\text{kg ha}^{-1} \text{year}^{-1}$ (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 fertilizers 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 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 Table S2.

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: Cd was measured and hence modelled 16 times over 64 years, while F was measured 11 times over 63 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 fertilizer trial (Figure 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 64 years, on average, the model produced likely concentrations of Cd in soils of 92% (range: 84%–108%) and 78% (range: 66%–87%) of the measured concentration for the 188 and 376 trial plots, respectively. Likely, F soil concentrations over 63 years simulation time on average were 90% (range: 80%–97%) and 106% (range: 95%–113%) of measured concentrations in 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

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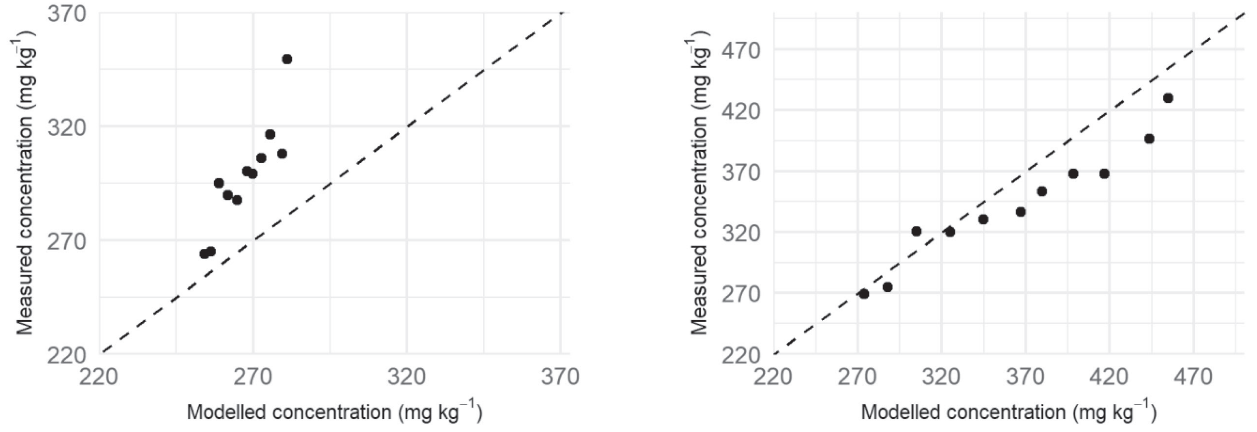
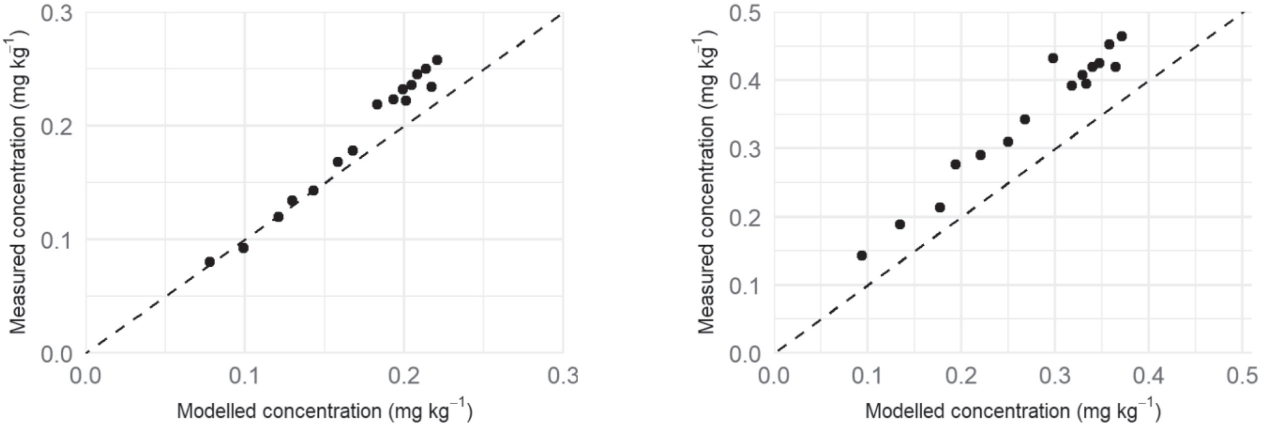


F 188 trial plot

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

F 376 trial plot

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

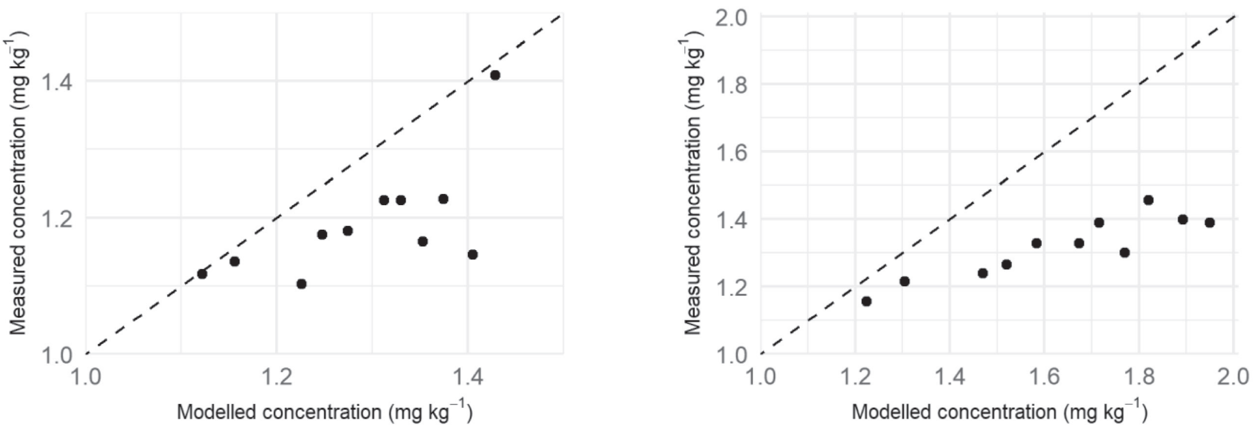


Cd 188 trial plot

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

Cd 376 trial plot

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



U 188 trial plot

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

U 376 trial plot

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

FIGURE 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} year^{-1} using Winchmore trial data. Dashed lines represent a 1:1 relationship between measured and modelled concentrations.

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TABLE 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

log-transformed values so as to make errors comparable between scenarios (Table 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 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)

4 | MODELLED TRACE ELEMENTS IN PALM PRODUCTION SOILS

We applied the model to an active oil palm 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, magnesium (Mg) and molybdenum (Mo). Four TEs were modelled: Cu, because of its application in Cu fungicide and F, Cd and U because of their presence in phosphate fertilizers used in oil palm production.

4.1 | Model input parameters

Two timeframes of 8 and 50 years were chosen for the modelled scenario (Table 4). These represented both a mid- and long-term analysis. The 8-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 oil palm production is expected to continue and expand within this timeframe (Murphy et al., 2021).

TABLE 4 Site-specific model input parameters for the modelling of TE inputs to an oil palm plantation.

General parameters	
Simulation time (year)	8; 50
Simulation depth (cm)	50
Rainfall and irrigation (mm year^{-1})	2950[a]
Evapotranspiration (mm year^{-1})	1538[b]
Soil parameters	
Initial contaminant concentration (mg kg^{-1})	[contaminant specific]
Erosive loss (t ha^{-1} year^{-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} year^{-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} year^{-1})	[Contaminant specific]
Atmospheric deposition (g ha^{-1} year^{-1})	[Contaminant specific]
Incorporation depth (cm)	1

[a] Rochayati et al. (2010).

[b] Takizawa et al. (2009).

[c] Aflizar et al. (2018). A total of 48% of area in a measured watershed had erosion up to 5 t ha year. Erosion up to >200 t ha^{-1} year^{-1} was reported in this study. However, at rates this high, the issue in questions 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).

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TABLE 5 Contaminant-specific parameters for the modelling of TE inputs to an oil palm plantation.

Contaminant name	Initial soil concentration (mg kg^{-1})	Initial plant concentration (mg kg^{-1})	K_D	Decay constant	Mass of contaminant added (g ha^{-1} year^{-1})
Fluorine	321	0.5	365	0	32,890
Copper	5.6	15	5643	0.1	14,432
Cadmium	0.085	0.012	227	0	54
Uranium	4	0.014	10,000	0	51

TABLE 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 (year)	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 run-off (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

We modelled scenarios for F, Cd and U because of their presence in phosphate fertilizers, and Cu because of its application in Cu-fungicides which are used in oil palm plantations. Site-specific and contaminant-specific model input parameters were used to run each scenario (Tables 4 and 5, respectively).

Where possible, site-specific data was used to model the TE fluxes in the oil palm plantation. Where this data were not available, the 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 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 fertilizers was calculated from TE contaminant rates in NPK (15–15-15) fertilizer (Oosterhuis et al., 2000; Taylor et al., 2016). NPK fertilizer was used as this was identified as the most commonly used fertilizer 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 oil palm 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 fertilizer every year. Applications of initial capital fertilizer were not modelled because of the high application rates of maintenance fertilizer 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} year^{-1} added as NPK contaminants (Taylor et al., 2016).

4.2 | Model outputs

For each scenario modelled, a mass-balance output was produced (Table 6). We also produced four figures from modelling results, corresponding with contaminant concentration in the final soil profile, the plant concentration of contaminant over the simulation time, the mass

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of contaminant leached over the simulation time and the contaminant concentration in soil over the simulation time (Figure 3).

4.3 | Implications for oil palm production

The modelling results indicate that accumulation of Cu, Cd and F above initial soil concentrations can be expected over time in the oil palm plantation because of NPK fertilizer and Cu-based fungicide applications (Figure 4). The amount of U added in fertilizer applications was low enough to effect no substantial change to background soil concentrations. The modelled concentrations of Cu surpassed SGVs within 8 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 8 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 oil palm 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 mg kg^{-1} after 8 and 50 years, respectively, while Cu accumulated to average concentrations down to 10 cm depth of 77 and 289 mg kg^{-1} after 8 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 et al., 2012; Taylor & Kim, 2009). The use of phosphate fertilizers and Cu fungicides in *E. guineensis* plantations presents risks to the sustainability of this system because of the presence of TE contaminants. We expect that SGVs for Cu and F will be exceeded within 8 years and production will become unviable because of 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 oil palm plantations in

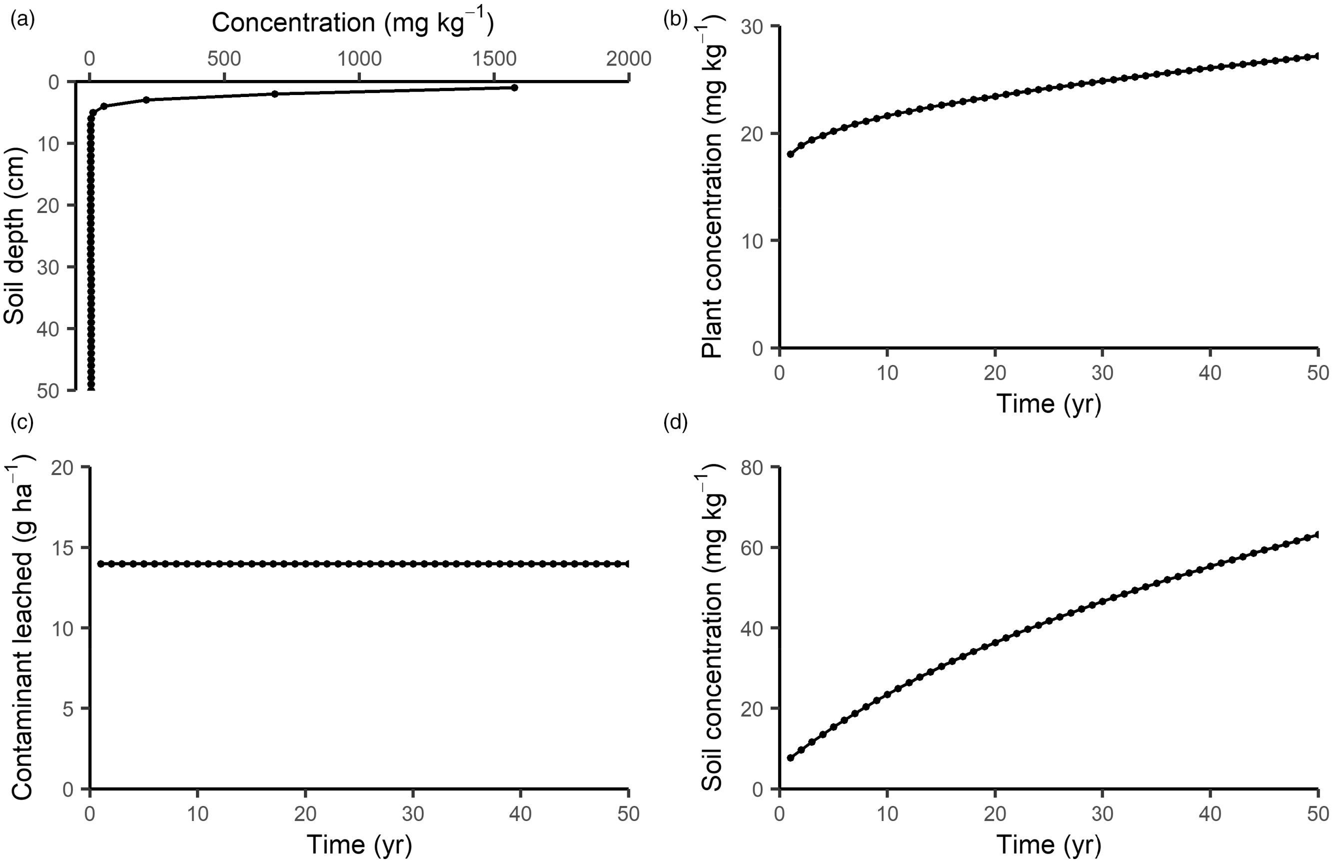

FIGURE 3 Output plots for Cu modelled in an oil palm plantation after 50 years, showing (a) Cu concentrations in the soil profile, (b) Cu concentrations in plants over time, (c) Cu concentrations leached over time and (d) total soil concentrations over time.

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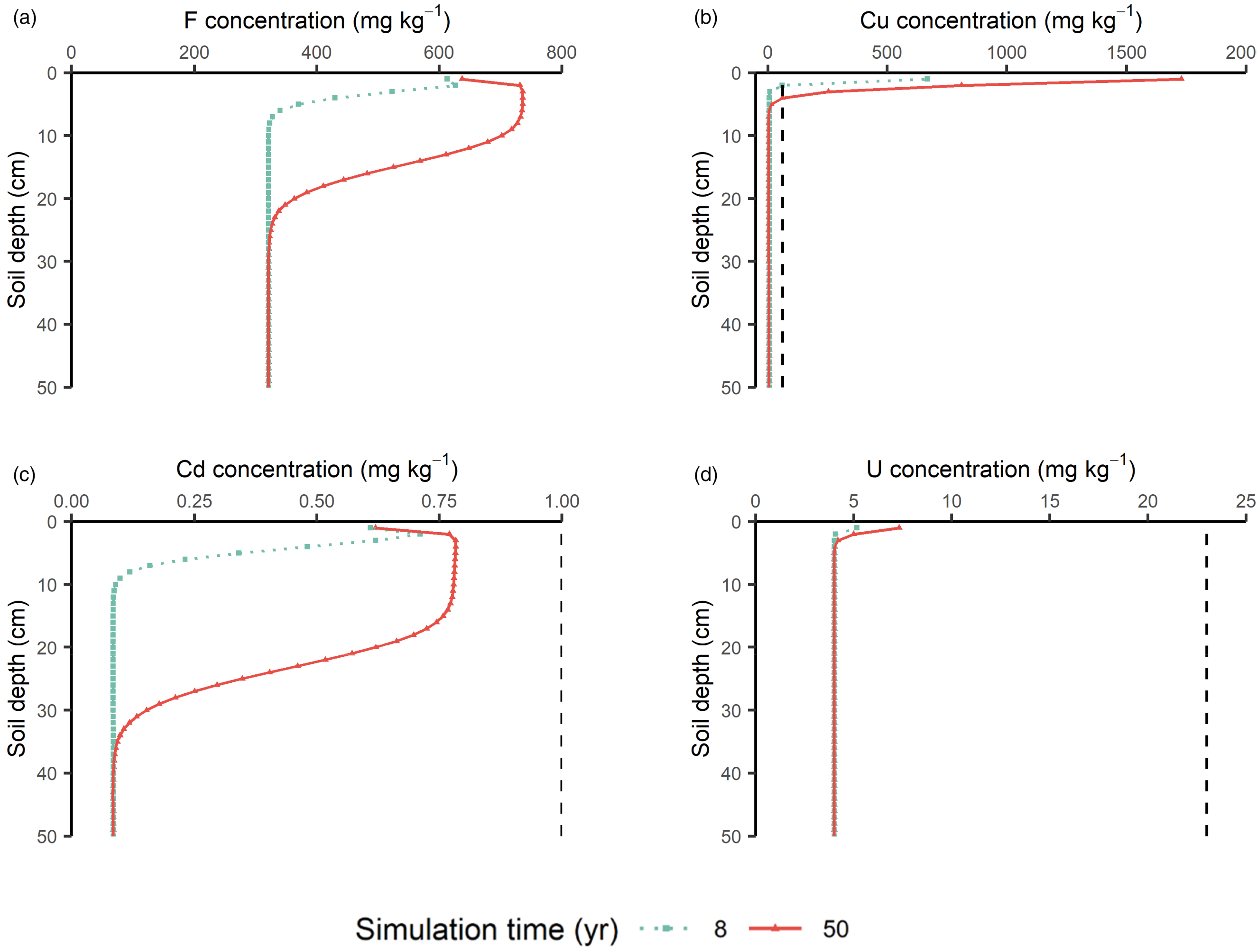

FIGURE 4 Final soil profiles for (a) F, (b) Cu, (c) Cd and (d) U concentrations after 8 and 50 years of inputs associated with oil palm production in a Sumatran plantation. Dashed vertical lines represent SGVs. The lower concentration in the top-most layer of profiles for F and Cd relative to the layer immediately below is attributed to the downward movement of the majority of TE in solution in the top-most layer, because of the K_D of these TEs.

Nigeria (Olafisoye et al., 2016, 2020) and Malaysia (Aini Azura et al., 2012), we see this as an emerging issue for the industry. Management practices which minimize TE inputs to soil may be researched and adopted in oil palm cultivation to minimize this risk.

Our modelling results indicate that if sufficient nutrients were applied to oil palm soils, accumulation of Cu and F would be much greater than it currently is. This current oil palm 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 because of 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, fertilizer subsidies that incentivize oil palm 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 oil palm 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.

5 | CONCLUSIONS

To calculate likely timeframes and limiting elements associated with TE accumulation in oil palm plantation soils owing to agrichemical applications, we have developed a mass-balance model to calculate the likely

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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 which receive some form of TE input and is applied at the hectare scale. This model has been validated with the Winchmore data set for Cd, F and U over timeframes of up to 64 years. The results of our modelling of oil palm plantation soils in Indonesia with recommended fertilizer inputs indicate that production will become unviable at this site because of phytotoxic concentrations of Cu and F within 50 years. In the modelled scenario, topsoil concentrations reached up to 289 and 719 mg kg^{-1} of Cu and F, respectively, within 50 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 data sets. It may also be possible to incorporate year-specific TE application rates which 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 oil palm soils into the future, this work will be carried out in future, separate publications.

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CONFLICT OF INTEREST STATEMENT

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

DATA AVAILABILITY STATEMENT

The data that supports the findings of this study are available in the supplementary material of this article.

ORCID

Hadee Thompson-Morrison https://orcid.org/0000-0002-4509-152X
Sally Gaw https://orcid.org/0000-0002-9556-3089

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