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Soil type influences crop mineral composition in Malawi

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HIGHLIGHTS

- Plant samples collected across Malawi and analysed for elemental composition.
- Higher concentrations of Ca, Se and Zn in cereal grains from calcareous soils.
- Soil dust contributed 77% and 34% of Fe in leaf and grain samples, respectively.
- Nationally, average dietary supplies of Ca, Se and Zn are inadequate; Cu, Fe, Mg adequate.
- Estimated risks of Ca, Se and Zn deficiency lower in areas of calcareous soils.

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ABSTRACT

Food supply and composition data can be combined to estimate micronutrient intakes and deficiency risks among populations. These estimates can be improved by using local crop composition data that can capture environmental influences including soil type. This study aimed to provide spatially resolved crop composition data for Malawi, where information is currently limited.

Six hundred and fifty-two plant samples, representing 97 edible food items, were sampled from >150 sites in Malawi between 2011 and 2013. Samples were analysed by ICP-MS for up to 58 elements, including the essential minerals calcium (Ca), copper (Cu), iron (Fe), magnesium (Mg), selenium (Se) and zinc (Zn).

Maize grain Ca, Cu, Fe, Mg, Se and Zn concentrations were greater from plants grown on calcareous soils than those from the more widespread low-pH soils. Leafy vegetables from calcareous soils had elevated leaf Ca, Cu, Fe and Se concentrations, but lower Zn concentrations. Several foods were found to accumulate high levels of Se, including the leaves of *Moringa*, a crop not previously been reported in East African food composition data sets.

New estimates of national dietary mineral supplies were obtained for non-calcareous and calcareous soils. High risks of Ca (100%), Se (100%) and Zn (57%) dietary deficiencies are likely on non-calcareous soils. Deficiency risks on calcareous soils are high for Ca (97%), but lower for Se (34%) and Zn (31%). Risks of Cu, Fe and Mg deficiencies appear to be low on the basis of dietary supply levels.

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1. Introduction

Mineral micronutrient deficiencies (MNDs) are an important global health problem, affecting up to two billion people worldwide (WHO, 2002, 2004, 2008, 2009; Muthayya et al., 2013). Estimates of deficiency for some minerals can be based on direct measurement of mineral concentrations or indicators in blood and other tissues or in urine (e.g., Gibson, 2005; Zimmermann, 2008; Fairweather-Tait et al., 2011). Alternatively, food consumption or supply data can be used to calculate dietary mineral intakes and infer deficiency risks. These methods include direct intake assessments from duplicate dietary analyses (e.g., Hurst et al., 2013) or, in conjunction with food composition tables (FCTs), individual recall-based dietary surveys (e.g., Gibson and Huddle, 1998; Department of Health/Food Standards Agency, 2011), household consumption data (e.g., Ecker and Qaim, 2011) and national Food Balance Sheets (FBSs) available from the United Nations Food and Agriculture Organization (FAO, 2014) (e.g., Broadley et al., 2012; Joy et al., 2014).

Previous studies to estimate mineral deficiency risks in Malawi using dietary recall or food records in combination with local or regional composition data have reported a high risk of zinc (Zn) deficiency in children (Ferguson et al., 1989), calcium (Ca) and Zn in pregnant women

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Table 1

Summary of published estimates of dietary nutrient supplies in Malawi.

Nutrient	Supply (mg capita ⁻¹ d ⁻¹)	Population (n)	Region	Methodology	Reference
Ca Zn Phytate	410, 335, 350 6.8, 6.2, 6.4 1621, 1667, 1729	Females 4–6 years: harvest season (60), post-harvest (60), pre-harvest (62)	Rural area, southern Malawi	Mean from three-day food records and local/published composition data	Ferguson et al. (1989)
Ca	473, 342, 379	Males 4–6 years: at harvest (60),			
Zn Phytate	7.8, 7.0, 8.0 1921, 1857, 2161	post-harvest (60), pre-harvest (62)			
Ca	415	Pregnant women (141)	Rural area, Mangochi	Median from 24 h recall and local food	Gibson and Huddle (1998)
Cu	1.0			composition data	
Fe	14.8				
Zn	9.0				
Se	0.044, 0.046	TB patients (40) and controls (40)	Rural area, Mangochi	Median from 24 h recall and local composition data	Eick et al. (2009)
Fe	19.0	11,280 nationally representative	National	Mean from HH survey and regional	Ecker and Qaim (2011)
Zn	10.2	households		composition data	
Ca	306	National	National	Mean from FBSs and local/ regional	Broadley et al. (2012)
Mg	789			composition data	
Fe	16.6	Adult women (55)	Rural area, soil pH <4.5	Median from one-day-weighed duplicate	Hurst et al. (2013);
Se	0.0066			diet composites	Siyame et al. (2013)
Zn	4.8				
Fe	29.6	Adult women (58)	Rural area, soil pH >6.5		
Se	0.0553				
Zn	6.4				
Ca	592	National	National	Mean from FBSs and regional composition	Joy et al. (2014)
Cu	2.95			data	
Fe	29.1				
Mg	760				
Se	0.0336				
Zn	11.8				

(Gibson and Huddle, 1998) and selenium (Se) in adults (Eick et al., 2009; Table 1). Local food composition data are likely to improve estimates of mineral deficiency risks in Malawi and a strong influence of soil type on the concentration of Se in maize grain and dietary composites has been identified. Chilimba et al. (2011) observed mean and median concentrations of Se in maize grain of 0.022 and 0.016 mg kg⁻¹ on low pH soils (n = 72), but 0.298 and 0.342 mg kg⁻¹ on Eutric Vertisols (pH > 6.9, n = 16). Such variation is consistent with earlier findings as Donovan et al. (1991) reported a mean Se concentration in refined maize flour in Zomba District of 0.029 mg kg⁻¹ (n = 10), while Eick et al. (2009) reported a mean concentration in Mangochi District of 0.078 mg kg⁻¹ (n > 20; Supplementary Table 1). Analyses of composite diets showed that the median Se intake of women from villages with predominantly non-calcareous soils (pH <4,5) in Zombwe Extension Planning Area (EPA) was 6.6 μ g d⁻¹ (range 1.1-62.3, n = 55) but was eight-fold greater in villages with predominantly calcareous soils in Mikalango EPA (median 55.3 µg d⁻¹, range 5.8-192, n = 58); the estimated average requirement (EAR) for adult females is 45 $\mu g d^{-1}$ (IOM, 2000a). Women in Mikalango EPA had greater median concentration of Se in blood plasma (117 μ g L⁻¹, range 82.6-204, n = 60) compared to subjects from Zombwe EPA (53.7 μ g L⁻¹, range 32.4-78.4, n = 60; Hurst et al., 2013).

There is evidence of spatial variation in dietary supplies of other elements, with median intakes of iron (Fe) and Zn of 29.6 and 6.4 mg d⁻¹ diets, respectively, among Mikalango subjects compared to 16.6 and 4.8 mg d⁻¹ for subjects from Zombwe EPA (Siyame et al., 2013); for diets low in animal products, the adult female EARs of Fe and Zn are 13.4 mg d⁻¹ and 8.2 mg d⁻¹, respectively (WHO and FAO, 2004). Low Zn intakes and high phytate:Zn ratios are likely to be the cause of low plasma Zn status, with 92 and 95% of plasma Zn concentrations <10.7 µmol L⁻¹ in Zombwe and Mikalango EPAs, respectively. In contrast, the majority of subjects in Zombwe (70%) and Mikalango (78%) EPAs were Fe-sufficient (body Fe >0 mg kg⁻¹ and haemoglobin >120 g L⁻¹). Despite greater intakes of Fe on high-pH soils, body Fe and haemoglobin (Hb) concentrations were greater on low pH soils. Consistent with these findings, Dickinson et al. (2009, 2014) reported greater blood Fe and Hb,

but lower Se, concentrations among pregnant women living in villages on low- compared to high-pH soils in southern Malawi. Zinc deficiency was also apparent in the low- and high-pH soil groups, with mean concentrations in the plasma of 9.8 and 10.3 μ mol L⁻¹, respectively.

At a national level, Ecker and Qaim (2011) estimated dietary Fe and Zn supplies to be adequate although intakes are highly dependent on income, using food supply data from the Malawi Second Integrated Household Survey (NSO, 2005) combined with regional food composition data (Table 1). Broadley et al. (2012) found that risks of deficiency are likely to be high for Ca but low for Mg in Malawi based on FBS supply data and national maize composition data. Overall, published sources suggest that risks of dietary mineral deficiencies of calcium (Ca), selenium (Se) and zinc (Zn) are likely to be high in Malawi, as for many other countries in sub-Saharan Africa (Joy et al., 2014), and that environmental factors such as soil type are an important consideration.

Risk estimates are sensitive to the quality of composition data, especially for elements required in trace quantities (sub-milligram *per capita per* day). The FCTs of greatest potential relevance to Malawi, and which have been used previously (Joy et al., 2014), are those for Tanzania (Lukmanji et al., 2008), Mozambique (Korkalo et al., 2011) and South Africa (Wolmarans et al., 2010). Further, Malawian food composition data for commonly consumed items were reported by Ferguson et al. (1988, 1989, 1993), Donovan et al. (1991), Eick et al. (2009) and Dickinson et al. (2014). However, some elements and foodstuffs are not adequately represented in these data sets; for example, Gibson et al. (2011) were not able to assess dietary Se intakes because of limited data on the Se concentration of local foodstuffs.

This study aimed to provide crop composition data for a wide range of food items and investigate the influence of soil type on national-level dietary mineral supplies. As maize grain is commonly processed into flour in Malawi during preparation of the staple dish, *nsima*, flour samples were taken to test the effect of household processing on elemental composition. Composition data were obtained for up to 58 elements in 652 plant samples, representing 97 edible food items collected from >150 different locations in Malawi, including grains, roots, tubers, fruits and leaves. Coupled soil samples were also taken where possible and analysed to determine mineral concentration, pH and organic matter content. New estimates of dietary mineral supplies and deficiencies were calculated using FBS supply data.

2. Methods

2.1. Sample collection and analysis

Plant and soil samples were collected from farmers' fields and markets during 2012 and 2013. Plant samples were brushed and washed with distilled water to remove soil and dust particles before being oven-dried at 40 °C. Where possible, soil samples were collected close to where the plant samples were collected, although this could not be done for plant samples bought at markets. Soil samples were air-dried, crushed and sieved to 2 mm. Both plant and soil samples were further ground to <40 µm in an agate ball mill. Plant samples were passed through a food blender prior to milling, as appropriate. Soil samples (0.25 g) were digested in a mixed acid solution (HF:2.5 mL/HNO₃:2 mL/HClO₄:1 mL/H₂O₂:2.5 ml) on a programmable hot block; 0.5 g of plant samples were digested in HNO₃:10 mL/H₂O₂:1 mL mixed solution in a closed vessel microwave heating system (MARS Xpress). Subsequent total elemental analysis was carried out by inductively coupled plasma mass spectrometry (ICP-MS; Agilent 7500cx) using (i) collision cell mode (He-gas) for Li, Be, B, Na, Mg, Al, P, S, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Rb, Sr, Y, Zr, Nb, Mo, Ag, Cd, Sn, Sb, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, Ta, W, Tl, Pb, Bi, Th and U; and (ii) H₂-reaction cell mode for Si (in plant material) and Se. Soil pH was measured in water using 10 g soil and 25 mL water. Organic matter content was determined as loss on ignition (LOI) at 450 °C for 1 g of soil. Elemental composition data for maize samples collected in 2011 are also reported (Chilimba et al., 2011).

2.2. Soil Fe contamination of plant samples

Trace elements are normally poorly available to plants and can be used to estimate the likely proportion of soil contamination contributing to apparent plant sample element concentrations. Vanadium (V) may be a reliable indicator of extraneous contamination with soil dust. Although mobile forms of V exist in acid soils (e.g., vanadyl, VO^{2+}) and in calcareous soils (e.g., vanadate, HVO_4^{2-}), most soil V is bound predominantly within ferric oxides where (reduced) trivalent V^{III} (0.064 nm) isomorphically substitutes for Fe^{III} (0.065 nm; Schwertmann and Pfab, 1996). Vanadate is also likely to be retained as a specifically adsorbed anion on Fe oxides under acidic conditions. Thus, as vanadyl and vanadate are unlikely to be systemically absorbed by plants concurrently with Fe^{2+} or Fe^{3+} , it seems reasonable to assume that a high correlation between V and Fe in plants indicates contamination with Fe oxides present in soil dust. In the current study, the correlation coefficient between V and Fe concentrations in plant samples was strong (r = 0.981, p < 0.001). Thus, extraneous contamination of plant samples with Fe was estimated where coupled soil samples were taken. For example, the proportion of Fe in plant samples originating from contaminant soil (P_{Fe}) was estimated from Eq. 1:

$$P_{\rm Fe} = \frac{V_{\rm plant} \quad {\rm Fe}_{\rm soil}}{V_{\rm soil} \quad {\rm Fe}_{\rm plant}}$$

where V_{plant} and Fe_{plant} are elemental concentrations in the plant, and V_{soil} and Fe_{soil} are concentrations in the soil.

2.3. Assessing the influence of soil factors on the mineral composition of plants

Previous studies have shown that soil type may influence maize grain and diet composite Se, Fe and Zn concentrations. Thus, plant and soil sample locations were matched to FAO soil classifications available at a national scale (Green and Nanthambwe, 1992) using the 'spatial join' function in ArcGIS (v. 9.3, ESRI, Redlands, CA, USA). The FAO soil classes were assigned to two groups: 'calcareous' (Calcaric and Eutric classes) and 'non-calcareous' (all other classes) (Supplementary Table 2). Leptosols were not assigned because they cannot be classified on this basis. Using this classification, 69% of the total land area in Malawi is non-calcareous, and 26% is calcareous and the remainder is unassigned. The validity of the calcareous/non-calcareous groupings was tested by comparing their soil Ca concentrations and pH, which confirmed that the term 'calcareous' indicated the presence of calcium carbonate in the soil and pH >6.5. Soil type and sampling locations are shown in Fig. 1.

The influence of calcareous/non-calcareous soil types on maize grain and 'leafy vegetables' Ca, copper (Cu), Fe, magnesium (Mg), Se and Zn concentrations was assessed using analysis of variance (ANOVA,

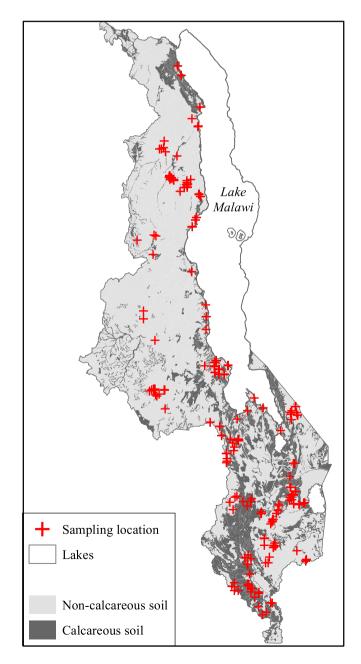


Fig. 1. A map of Malawi showing plant sampling locations and soil type.

Table 2

Summary statistics of plant sample Ca, Cu, Fe, Mg, Se and Zn concentrations (dry-weight, edible portion) by soil type (calcareous/non-calcareous) for 25 commonly consumed crops with >1 sample.

Amaranth,leaf y 3 23400 23800 23900 29300 2300 5.22 5.22 9.7 20.2 20.2 3 228 603 2950 2050 2100 3300 210.6 83 107 152 212 210 130 366 357.6 64.3 22 46.3 24 84.3 160 321.2 212 216 33 212 216	Plant_tissue Calcareous?		Ca ((mg kg ⁻¹	')				Cu ((mg kg [_]	¹)				Fe (mg kg ^{-1})					
Amarath Index 5 1760 19000 23200 25500 27.0 16.0 22.8 30.8 5 16.4 30.2 731 27.0 20.2 Banana,fruit n 4 126 131 160 331 384 4 2.71 3.02 4.68 7.93 8.76 4 8.12 8.54 1.24 2.07 2.3 7.66 8.58 16.4 1.02 2.2 1.02 1.25 1.25 1.25 1.25 1.25 1.25 1.25 1.25 1.2 1.2 1.2 5 1.06 8.08 8.7 1.1 1.5 6.0 8.08 8.8 1.07 1.55 1.6 1.6 1.06			n	Min	Q1	Median	Q3	Max	n	Min	Q1	Median	Q3	Max	n	Min	Q1	Median	Q3	Max
Banan_fruit n 4 126 131 160 331 384 4 2.71 3.02 4.68 7.93 8.76 4 8.12 8.54 12.4 20.7 22.7 Bean,seed y 3 1010 1010 1040 1200 1200 12.5 14.9 19.2 19.2 19.2 13 7.2.6 7.8.8 8.9.8 7.8.7 8.9.8 7.8.7 8.9.8 7.8.7 8.9.8 7.8.7 8.9.8 7.8.7 8.9.8 7.8.7 8.9.8 7.8.7 8.9.8 7.8.7 8.9 11.0 15.0 12.0	Amaranth_leaf	у	3	23400	23400	23800	29300	29300	3	5.22	5.22	9.7	20.2	20.2	3	228	228	603	2950	2950
Bean_seed y 3 1250 1250 1250 1710 1710 3 125 12.5 14.9 19.2 19.2 3 76.6 76.6 96.1 <td>Amaranth_leaf</td> <td>n</td> <td>5</td> <td>17600</td> <td>19000</td> <td>23200</td> <td>26500</td> <td>27200</td> <td>5</td> <td></td> <td>7.27</td> <td>10.6</td> <td>22.8</td> <td>30.8</td> <td>5</td> <td>164</td> <td>302</td> <td>731</td> <td>2710</td> <td>3040</td>	Amaranth_leaf	n	5	17600	19000	23200	26500	27200	5		7.27	10.6	22.8	30.8	5	164	302	731	2710	3040
Bean_seed n 3 1010 1010 1040 1200 1200 3 5.24 7.13 17.2 17.2 3 69.8 60.8 70.8 64.3 20 61.0 70.8 61.0 70.8 61.0 70.8 61.0 70.8 70.3 70.8 70.3 70.8 70.3 70.8 70.3 70.8 70.3 70.8 70.3 70.8 70.3 70.8 70.3 70.3 70.3 70.3 70.4 70.3 70.3 70.3 70.3 70.3 70.3 70.3 70.3 70.3 70.3 <th70.3< th=""> 70.3 <th70.3< th=""></th70.3<></th70.3<>	Banana_fruit	n	4	126	131	160	331	384	4	2.71	3.02	4.68	7.93	8.76	4	8.12	8.54	12.4	20.7	22.7
Black jack leaf n 5 7470 7510 12700 14100 14100 5 8.8 107 15.2 21 22 5 102 116 184 691 118 Cabbage_leaf n 4 3780 4110 6550 22000 3700 4 1.06 * 55 7.62 4 61.2 63.3 112 166 170 361 489 527 Cassava_leaf n 4 3780 4110 6550 15200 1700 10 60 8.2 8.7 11.1 15.6 61.0 71.0 10 489 527 Cassava_root n 6 518 599 75.3 1560 160 12 3.7 7.5 6.3 61.2 46.7 52.6 58.6 63 Cassava_root n 6 518 599 73.3 1560 120 145.0 12 3.9 7.7 58.6 12 46.7 52.6 58.6 63 Growpea_seed n 5	Bean_seed	У	3	1250	1250	1560	1710	1710	3	12.5	12.5	14.9	19.2	19.2	3	72.6	72.6	98.1	251	251
Cabbage_leaf y 2 5160 * 18700 * 32300 2 10.6 * 37.5 * 64.3 2 8.8 * 71.3 * 13.8 Cabbage_leaf n 4 3780 4110 650 25000 30700 6 8.2 8.5 11.1 6.6 7.62 4 6.12 6.33 1.12 1.68 1.72 Cassava_leaf n 10 4440 7420 9570 1700 170 6.05 8.8 8.7 6.03 3 3 2 1.2 2.8 3.8 3.8 3.8 3.8 3.8 7.8 1.60 2.8 8.8 7.6 6.3 6.3 3.4 3.9 6.5 5.8 7.5 8.4 1.2 1.8 1.4 2.2 1.03 1.6 7.2 4.6 5.8 7.5 8.4 1.2 1.8 1.4 2.4 2.0 6.58 8.7 1.6 3.4 1.2 1.6 1.4 1.2 1.6 1.4 1.2 1.6 <t< td=""><td>Bean_seed</td><td>n</td><td>3</td><td>1010</td><td>1010</td><td>1040</td><td>1200</td><td>1200</td><td>3</td><td>5.24</td><td>5.24</td><td>7.13</td><td>17.2</td><td>17.2</td><td>3</td><td>69.8</td><td>69.8</td><td>76.8</td><td>96.1</td><td>96.1</td></t<>	Bean_seed	n	3	1010	1010	1040	1200	1200	3	5.24	5.24	7.13	17.2	17.2	3	69.8	69.8	76.8	96.1	96.1
Cathong-Leal y z 3100 1100 52.300 z 100 51.3 61.3 z 46 71.5 110 51.3 61.3 z 46 71.5 110 51.3 61.3 2 46 71.5 110 51.3	Black jack_leaf	n	5	7470	7510	12700	14100	14100	5	8.8	10.7	15.2	21	22	5	102	116	184	691	1180
Cassav_leaf y 6 5230 6400 9550 15200 19300 6 8.2 8.5 11.1 56.1 67.2 6 129 170 361 489 527 Cassav_leaf n 10 4440 7420 9570 17400 17400 10 60 6.8 8.74 11.1 15.6 10 11 193 348 188 56 Cassav_root n 6 518 599 753 1560 22 6.6 7.57 6.23 7.05 8.46 12 4.67 5.98 7.32 6.46 7.4 3.05 10.8 1.3 5 8.8 5.10 8.13 1.3 5.2 5.26 5.86 6.3 6.47 7.99 3.3 1.1 1.61 1.4 1.5 9.0 7.0 1.3 1.3 5.20 5.20 5.26 5.86 6.3 6.47 7.99 3.1 1.1 1.5 1.4 1.5 1.5 1.5 6.6 1.21 1.3 1.6 1.3 1.4	Cabbage_leaf	У	2	5160	*	18700	*	32300	2	10.6	*	37.5	*	64.3	2	48	*	713	*	1380
Cassava_leaf n 10 4440 7420 9570 14700 10 666 6.89 8.74 11.1 15.6 10 71 101 193 348 188 Cassava_noot y 3 328 328 784 5160 5160 2.8 2.8 5.7 60.3 60.3 3.2 2.2 2.2 36.8 36.3 32.4 2.2 2.2 36.8 36.4 2.2 2.0 3.6 3.2 2.2 2.0 3.5 13.4 2.2 2.0 3.5 13.4 2.2 2.0 3.5 13.4 2.2 2.00 4.55 5.6 6.9 7.84 8.3 5 4.67 7.09 8.3 10.8 11.8 1.8 1.9 1.0 1.9 3.0 1.5 1.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5 2.5 3.5 3.6 1.1 1.1 1.5 1.9 3.1 1.5 1.5 1.5 1.5 </td <td>Cabbage_leaf</td> <td>n</td> <td>4</td> <td>3780</td> <td>4110</td> <td>6550</td> <td>25000</td> <td>30700</td> <td>4</td> <td>1.09</td> <td>1.56</td> <td>3.05</td> <td>6.5</td> <td>7.62</td> <td>4</td> <td>61.2</td> <td>63.3</td> <td>112</td> <td>166</td> <td>170</td>	Cabbage_leaf	n	4	3780	4110	6550	25000	30700	4	1.09	1.56	3.05	6.5	7.62	4	61.2	63.3	112	166	170
Cassav_root y 3 328 328 784 5160 5160 3 2.8 2.8 5.7 60.3 60.3 3 21.2 21.2 22 36.8 36.4 Cassav_root n 6 518 599 753 1560 250 120 12 3.9 5.7 6.03 6.03 8.0 12 4.7.5 7.6 5.7 6.45 5.7 6.45 5.7 6.45 5.7 6.45 5.7 6.45 5.8 7.35 13.4 22 Cowpe_ased n 5 4.81 565 692 7.84 833 5 4.67 5.99 8.35 1.08 1.13 5 4.32 4.05 5.2	Cassava_leaf	У	6	5230	6400	9550	15200	19300	6	8.2	8.5	11.1	56.1	67.2	6	129	170	361	489	527
Cassav_root n 6 518 599 753 1560 2950 6 0.508 1.12 2.38 3.49 3.79 6 5.87 5.98 7.35 13.4 22. Cowpea_seed y 12 549 923 1050 1200 1450 12 3.9 5.7 6.23 7.05 8.46 12 4.67 5.9 8.45 12.0 4.65 5.99 8.35 10.8 11.3 5 4.81 5.6 5.26	Cassava_leaf	n	10	4440	7420	9570	14700	17400	10	6.06	6.89	8.74	11.1	15.6	10	71	101	193	348	1880
Cowpea_seed y 12 549 923 1050 120 1450 12 3.9 5.7 6.23 7.05 8.46 12 6.4.5 5.4 5.6 5.6 5.6 6.22 7.84 833 5 4.67 5.99 8.35 10.8 11.3 5. 4.32 6.45 5.6 6.35 6.35 7.05 8.46 7.2 6.45 7.2 6.45 5.6 6.5 6.5 6.92 7.84 8.33 5 6.64 7.09 9.3 12.1 13.4 8.1 9.1 12.0 14.4 14.0<	Cassava_root	У	3	328	328	784	5160	5160	3	2.8	2.8	5.7	60.3	60.3	3	21.2	21.2	22	36.8	36.8
Cowpa-seed n 5 481 565 692 784 833 5 4.67 5.99 8.35 10.8 11.3 5 43.2 46.5 52.6 58.6 63.7 Finger millet_grain n 2 2000 * 3800 * 513 553 8 6.44 * 9.3 * 12.2 2 103 * 152 * 201 Groundnut_seed n 8 300 37.9 445 513 553 8 6.44 * 9.3 12.1 13.4 8 19.4 19.6 20.5 25.7 29.3 Maize_grain n 94 <30 34.3 43.1 61.1 87.1 43 1.2 2.06 2.48 3.41 11.9 19.3 17.8 17.8 18.1 18.1 18.1 18.1 18.1 18.1 18.1 18.1 18.1 18.1 18.1 18.3 13.4 18.4 18.4 18.4 18.4 18.4 18.4 18.4 18.4 18.4 18	Cassava_root	n	6	518	599	753	1560	2950	6	0.508	1.12	2.38	3.49	3.79	6	5.87	5.98	7.35	13.4	22.1
Finger millet_grain n 2 2900 * 3800 * 4700 2 6.44 * 9.3 * 12.2 2 103 * 152 * 201 Groundnut_seed n 8 300 379 445 513 553 8 6.4 7.09 9.3 12.1 13.4 8 19.4 19.6 20.5 25.7 29.0 Maize_grain y 40 <30	Cowpea_seed	V	12	549	923	1050	1200	1450	12	3.9	5.7	6.23	7.05	8.46	12	46.7	57	64.5	72.4	324
Inite grain In	Cowpea_seed	n	5	481	565	692	784	833	5	4.67	5.99	8.35	10.8	11.3	5	43.2	46.5	52.6	58.6	63
Maize_grain y 40 <30 34.3 43.1 61.3 87.1 43 1.2 2.06 2.48 3.43 1.94 33 10.8 16.8 21.5 31.5 96.7 Maize_grain n 94 <30 <30 30.9 41 163 94 0.72 1.39 1.67 2.41 421 90 9.06 1.28 1.42 16.6 34.4 Mango_fruit n 93 707 707 1360 2080 2080 3 3.72 6.31 11.9 11.9 3 17.8 17.8 18.1 18.1 Mango_fruit n 9 305 580 911 1350 2270 9 2.69 6.6 8.06 10.2 14.8 9 17.8 17.8 18.1 12.0 12.0 12.0 13.0 14.0 18.0 13.0 13.0 13.0 13.0 13.0 13.0 13.0 13.0 14.0 13.0 13.0 13.0 13.0 13.0 13.0 13.0 13.0 13.0<	Finger millet_grain	n	2	2900	*	3800	*	4700	2	6.44	*	9.3	*	12.2	2	103	*	152	*	201
Maize_grain n 94 <30 <30 94.1 163 94 0.72 1.39 1.67 2.41 421 90 9.06 1.28 1.42 1.66 3.4.4 Mango_fruit y 3 707 707 1360 2080 2080 3 3.72 3.72 6.31 11.9 1.9 3 1.78 1.78 1.78 1.81 18.1 18.1 Mango_fruit n 9 305 580 911 1350 2270 9 2.69 6.6 8.06 10.2 14.8 9 11.2 13.8 15.4 19.3 23 Mushroom_fruit n 3 266 266 6.9 11.3 40.4 40.4 3 32 34 140 130 140 130 140 33 140 130 140 30.7 36.0 36.0 36.0 36.9 6.9 13.4 40.4 4 31 2 24.3 39 73 130 140 40 30.00 30.00 30.00	Groundnut_seed	n	8	300	379	445	513	553	8	6.4	7.09	9.3	12.1	13.4	8	19.4	19.6	20.5	25.7	29.3
Mango_fruit y 3 707 707 1360 2080 2080 3 3.72 3.72 6.31 11.9 11.9 3 17.8 17.8 17.8 18.1 18.1 18.1 Mango_fruit n 9 305 580 911 1350 2270 9 2.69 6.6 8.06 10.2 14.8 9 11.2 13.8 15.4 19.3 23 Mushroom_fruit n 3 256 256 461 501 501 3 3.1.1 41.4 49.6 57.3 57.3 3 19.7 19.7 485 1320 132 Onion_bulb n 3 1640 160 307 3650 3 650 6.9 6.9 6.11.3 40.4 40.4 3 2 44 47.8 47.4 Papaya_fruit y 2 6830 170 256 362 1410 38 2.45 4.92 5.44 6.3 12.2 38 393 783 188 2	Maize_grain	V	40	<30	34.3	43.1	61.3	87.1	43	1.2	2.06	2.48	3.43	19.4	33	10.8	16.8	21.5	31.5	96.7
Mangefruit n 9 305 580 911 1350 2270 9 2.69 6.6 8.06 10.2 14.8 9 11.2 13.8 15.4 19.3 23 Mushroom_fruit n 3 256 256 461 501 501 3 34.1 34.1 49.6 57.3 57.3 3 197 197 485 1320 132 Onion_bulb n 3 1640 1640 3070 3650 3650 3 6.9 6.9 11.3 40.4 40.4 3 32 32 44 47.8 47.4 Papaya_fruit y 2 6890 * 9630 * 12400 2 8.74 * 13.4 * 18.1 2 24.3 * 30.9 26.6 Pumpkin_leaf y 3 14100 15000 21500 38000 5170 5.3 7.33 7.35 7.3 3 147 147 30.8 75.7 75.3 7.3 7.3 7.35 <	Maize_grain	n	94	<30	<30	30.9	41	163	94	0.72	1.39	1.67	2.41	421	90	9.06	12.8	14.2	16.6	34.4
Mushroom_fruit n 3 256 256 461 501 501 3 34.1 34.1 49.6 57.3 57.3 3 197 197 485 1320 132 Onion_bulb n 3 1640 1640 3070 3650 3650 3 6.9 6.9 11.3 40.4 40.4 3 32 32 44 47.8 47.3 Papaya_fruit y 2 6890 * 9630 * 12400 2 8.74 * 13.4 * 18.1 2 24.3 * 39.4 * 54.4 Papaya_fruit y 3 14100 15000 21500 38000 5170 5 6.69 8.5 10.6 32.0 38 38.0 200 2000 8.28 8.85 12.3 19 9 284 487 1800 2570 268 Pumpkin_leaf n 3 12300 1900 2000 2900 35.5 5.35 7.33 7.35 3 147	Mango_fruit	V	3	707	707	1360	2080	2080	3	3.72	3.72	6.31	11.9	11.9	3	17.8	17.8	17.8	18.1	18.1
Onion-bullb n 3 1640 1640 3070 3650 3650 3 6.9 11.3 40.4 40.4 3 32 32 32 44 47.8 47.8 Papaya_fruit y 2 6890 * 9630 * 12400 2 8.74 * 13.4 * 18.1 2 24.3 * 39.4 * 54.4 Pearl millet_grain y 3 126 179 256 362 1410 38 2.45 4.92 5.44 6.3 12.2 38 56.6 81.5 166 309 22.6 Pumpkin_leaf y 5 14100 15000 21500 38000 51700 5 6.69 8.5 10.6 22.9 35 5 334 393 783 1880 204 Pumpkin_leaf n 3 12300 12900 29000 29000 2900 35.5 5.35 7.33 7.35 3 147 30.8 27.5 26.8 27.0 26.8 2	Mango_fruit	n	9	305	580	911	1350	2270	9	2.69	6.6	8.06	10.2	14.8	9	11.2	13.8	15.4	19.3	23
Papaya_fruit y 2 6890 * 9630 * 12400 2 8.74 * 13.4 * 18.1 2 24.3 * 39.4 * 54.4 Pearl millet_grain y 38 126 179 256 362 1410 38 2.45 4.92 5.44 6.3 12.2 38 56.6 81.5 166 309 226 Pumpkin_leaf y 5 14100 15000 21500 38000 51700 5 6.69 8.5 10.6 22.9 35 5 334 393 783 1880 204 Pumpkin_leaf n 9 4810 1800 26000 3800 5 5.55 7.33 7.35 3 147 18.00 2570 268 Rape_leaf n 3 12.00 12300 19000 29000 2900 3.55 5.35 7.33 7.35 3 147 47.09 9.84 21.0 47.4 Sorghum_grain y 40 <30	Mushroom_fruit	n	3	256	256	461	501	501	3	34.1	34.1	49.6	57.3	57.3	3	197	197	485	1320	1320
Paga Intri y 2 0630 1000 12400 2 6.74 13.4 13.4 16.1 2 24.3 35.4 35.4 35.4 35.4 92.4 Pearl millet_grain y 38 126 179 256 362 1410 38 2.45 4.92 5.44 6.3 12.2 38 56.6 81.5 166 309 22.6 Pumpkin_leaf y 5 14100 15000 21500 3800 51700 5 6.69 8.5 10.6 22.9 35 5 33.4 393 783 1880 204 Pumpkin_leaf n 9 4810 11800 18100 26000 38900 5.35 5.35 7.33 7.35 7.3 147 147 308 753 753 Rape_leaf n 3 12300 12010 1202 562 2420 40 6.08 3.41 4.93 6.33 17.4 4 7.09 9.84 21.0 47.4 Sorghum_grain n	Onion_bulb	n	3	1640	1640	3070	3650	3650	3	6.9	6.9	11.3	40.4	40.4	3	32	32	44	47.8	47.8
Pumpkin_leaf y 5 14100 15000 21500 38000 51700 5 6.69 8.5 10.6 22.9 35 5 334 393 783 1880 204 Pumpkin_leaf n 9 4810 11800 18100 26000 38900 9 6.91 8.28 8.85 12.3 19 9 284 487 1850 2570 268 Rape_leaf n 3 12300 12300 19900 29000 29000 3 5.35 5.35 7.33 7.35 7.35 3 147 147 308 753 753 Rice_grain * 21 <30	Papaya_fruit	v	2	6890	*	9630	*	12400	2	8.74	*	13.4	*	18.1	2	24.3	*	39.4	*	54.4
Pumpkin_leaf n 9 4810 11800 18100 26000 38900 9 6.91 8.28 8.85 12.3 19 9 284 487 1850 2570 268 Rape_leaf n 3 12300 12300 19900 29000 29000 3 5.35 5.35 7.33 7.35 7.35 3 147 147 308 753 753 Rice_grain * 21 <30	Pearl millet_grain	v	38	126	179	256	362	1410	38	2.45	4.92	5.44	6.3	12.2	38	56.6	81.5	166	309	2260
Pumpkin_leaf n 9 4810 11800 18100 26000 38900 9 6.91 8.28 8.85 12.3 19 9 284 487 1850 2570 268 Rape_leaf n 3 12300 12300 19900 29000 29000 3 5.35 5.35 7.33 7.35 7.35 3 147 147 308 753 753 Rice_grain * 21 <30	Pumpkin leaf	v	5	14100	15000	21500	38000	51700	5	6.69	8.5	10.6	22.9	35	5	334	393	783	1880	2040
Rap-leaf n 3 12300 12300 19900 29000 3 5.35 5.35 7.33 7.35 7.35 3 147 147 308 753 753 Rice_grain * 21 <30 35.7 40.1 65.3 75.7 21 0.853 1.88 2.28 3.58 13.8 21 <4 7.09 9.84 21.0 47.4 Sorghum_grain y 40 <30 151 202 562 2420 40 <0.83 1.44 4.93 6.33 17.1 40 <4 7.63 155 2.68 250 Sorghum_grain n 5 87.9 96.9 140 234 260 5 3.56 4.1 5.09 12.5 17 5 47.6 5.36 64.3 77.4 85.95 Sorghum_grain n 2 2450 * 2660 * 2870 2 11.6 * 13.2 * 14.8 2 94.7 55.7 * 97.4 Sorghum_grain <	1 –	5	9	4810	11800	18100	26000	38900	9	6.91		8.85	12.3	19	9	284	487	1850	2570	2680
Rice_grain * 21 <30 35.7 40.1 65.3 75.7 21 0.853 1.88 2.28 3.58 13.8 21 <4 7.09 9.84 21.0 47.4 Sorghum_grain y 40 <30	1 –	n	3	12300	12300	19900	29000	29000	3	5.35	5.35	7.33	7.35	7.35	3	147	147	308	753	753
Sorghum_grain y 40 <30 151 202 562 2420 40 <0.08 3.41 4.93 6.33 17.1 40 <4 76.3 155 268 250 Sorghum_grain n 5 87.9 96.9 140 234 260 5 3.56 4.1 5.09 12.5 17 5 47.6 53.6 64.3 77.4 85.9 Soya_seed n 2 2450 * 2660 * 2870 2 11.6 * 13.2 * 14.8 2 94 * 95.7 * 97.9 Sweet potato_leaf n 4 5600 5820 7250 10200 1080 4 11.7 12.1 13.9 19.8 21.6 4 208 23 729 2030 232 Sweet potato_leaf n 4 5600 6570 10100 20600 23900 4 12.9 13.1 14.5 24.5 27.7 4 99 139 279 794 95.9 <td>1 -</td> <td></td> <td>21</td> <td></td> <td></td> <td></td> <td></td> <td></td> <td>21</td> <td>0.853</td> <td></td> <td></td> <td></td> <td></td> <td>21</td> <td></td> <td></td> <td></td> <td></td> <td>47.8</td>	1 -		21						21	0.853					21					47.8
Sorghungrain n 5 87.9 96.9 140 234 260 5 3.56 4.1 5.09 12.5 17 5 47.6 53.6 64.3 77.4 85.9 Soya_seed n 2 2450 * 2660 * 2870 2 11.6 * 13.2 * 14.8 2 94 * 95.7 * 97.4 Sweet potato_leaf y 4 5660 5820 7250 10200 10800 4 11.7 12.1 13.9 19.8 21.6 4 208 232 729 2030 232 Sweet potato_leaf n 4 5600 6570 10100 20600 23900 4 12.9 13.1 14.5 24.5 27.7 4 99 139 279 794 95 Sweet potato_tuber y 2 682 * 711 * 740 2 7.68 * 12.4 * 17.1 2 9.84 * 10 * 10.7	-0	v														<4				2500
Soya_seed n 2 2450 * 2660 * 2870 2 11.6 * 13.2 * 14.8 2 94 * 95.7 * 97.4 Sweet potato_leaf y 4 5660 5820 7250 10200 10800 4 11.7 12.1 13.9 19.8 21.6 4 208 232 729 2030 232 Sweet potato_leaf n 4 5600 6570 10100 20600 23900 4 12.9 13.1 14.5 24.5 27.7 4 99 139 279 794 955 Sweet potato_tuber y 2 682 * 711 * 740 2 7.68 * 12.4 * 17.1 2 9.84 * 10 * 10.2 Tomato_fruit y 2 3600 * 5500 * 7390 2 18.5 * 43.3 * 68.2 2 88.8 * 98.1 * 107 </td <td></td> <td></td> <td></td> <td></td> <td></td> <td></td> <td></td> <td></td> <td>5</td> <td></td> <td></td> <td></td> <td></td> <td>17</td> <td></td> <td>47.6</td> <td></td> <td></td> <td></td> <td>85.9</td>									5					17		47.6				85.9
Sweet potato_leaf y 4 5660 5820 7250 10200 10800 4 11.7 12.1 13.9 19.8 21.6 4 208 232 729 2030 232 Sweet potato_leaf n 4 5600 6570 10100 20600 23900 4 12.9 13.1 14.5 24.5 27.7 4 99 139 279 794 955 Sweet potato_tuber y 2 682 * 711 * 740 2 7.68 * 12.4 * 17.1 2 9.84 * 10 * 10.7 Tomato_fruit y 2 3600 * 5500 * 7390 2 18.5 * 43.3 * 68.2 2 88.8 * 98.1 * 107	0 0		-						-						-					97.4
Sweet potato_leaf n 4 5600 6570 10100 20600 23900 4 12.9 13.1 14.5 24.5 27.7 4 99 139 279 794 955 Sweet potato_tuber y 2 682 * 711 * 740 2 7.68 * 12.4 * 17.1 2 9.84 * 10 * 10.7 Tomato_fruit y 2 3600 * 5500 * 7390 2 18.5 * 43.3 * 68.2 2 88.8 * 98.1 * 107	5 -		_		5820		10200				12.1		19.8				232		2030	2320
Sweet potato_tuber y 2 682 * 711 * 740 2 7.68 * 12.4 * 17.1 2 9.84 * 10 * 10.2 Tomato_fruit y 2 3600 * 5500 * 7390 2 18.5 * 43.3 * 68.2 2 88.8 * 98.1 * 107	1 –	5	-						-						-					959
Tomato_fruit y 2 3600 * 5500 * 7390 2 18.5 * 43.3 * 68.2 2 88.8 * 98.1 * 107	1 –		•						-											10.2
	1 –	5	-		*		*		_		*		*				*		*	10.2
יו דע דע געט געט געט גער	Tomato fruit	n	5	1990	2000	3150	5560	7680	5	11.7	12.4	17.6	26.1	30	5	56.8	59	76.3	110	113

GenStat v.16, VSN International, Hemel Hempstead, UK). A log_{10} transformation was used to reduce skew in the data. The influence of soil type was assessed for maize grain and leafy vegetable as these categories had adequate sample numbers and represent different plant tissues.

2.4. New estimates of national dietary mineral supplies

Dietary mineral supplies were estimated using FBS food supply data (FAO, 2014). Analysed plant samples were assigned to a best-fit FBS category according to the authors' discretion. A maximum of four croptypes were chosen to represent each FBS item. Where more than four crop-types could be matched, the most commonly consumed were chosen on the basis of household survey data (NSO, 2012; Supplementary Table 3). Samples were assigned by soil type to create two composition tables: calcareous and non-calcareous. Calcium, Cu, Fe, Mg, Se and Zn composition values for each FBS item were calculated as the median of assigned plant samples. Median values were used as they are robust to outliers. In the absence of relevant data for one soil type, composition data from the other soil type was used. For FBS items not represented at all in the survey (e.g., animal products), published food composition data were used from a previously compiled data set for the East Africa region (Joy et al., 2014). Because the FBS supply data are reported on a fresh weight basis, per capita supply was first converted to dry weight (DW) equivalents using estimated moisture content values (USDA-ARS, 2013; Supplementary Table 4). Estimated rates of deficiency were calculated using the EAR cut-point method (cf. Joy et al., 2014 for methodology and detailed discussion).

3. Results and discussion

3.1. Categorisation of soils

Results of soil chemical and physical analyses are presented in Supplementary Table 5. Mean and median total Ca concentrations for the calcareous soil group were 14,800 and 13,600 mg kg⁻¹ (range 193– 43,200, n = 57), while mean and median soil pH values were 6.79 and 6.63. These values compare to mean and median Ca concentrations for the non-calcareous soil group of 5,910 and 2,210 mg kg⁻¹ (range 43.5–35,100, n = 101), and mean and median soil pH of 5.67 and 5.54. Both Ca concentration (log₁₀ transformed to account for nonnormality) and pH were greater in calcareous soils (one-way ANOVA; p < 0.001). The use of FAO soil classifications as defined by national scale maps is therefore likely to provide a useful prediction of whether plant samples were grown on calcareous or non-calcareous soil type, provided that the plant was grown at or near the sampling location. For example, the majority of plant samples were collected directly from farmers' fields and samples bought from markets were generally grown in the local area, as confirmed by the vendor. Rice is likely to be an exception as it is commonly grown as a cash crop and traded over relatively long distances. Therefore, the location where rice is purchased is unlikely to provide a good prediction of the soil type on which it was grown; the data for rice are therefore reported independent of soil type.

Several soil samples that mapped as 'non-calcareous' had unexpectedly high Ca concentrations ($>20,000 \text{ mg kg}^{-1}$). It is likely that this discrepancy resulted from the different scales of mapping used in this

Mg ($Mg (mg kg^{-1})$					Se (mg kg ⁻¹)						Zn (mg kg ⁻¹)					
n	Min	Q1	Median	Q3	Max	n	Min	Q1	Median	Q3	Max	n	Min	Q1	Median	Q3	Max
3	12700	12700	14800	19300	19300	3	0.0615	0.0615	0.0835	0.173	0.173	3	41.8	41.8	41.9	49.6	49.6
5	11200	11400	12200	13500	13900	5	0.0454	0.052	0.069	0.101	0.111	5	42.8	45.2	63.6	69.5	70.2
4	973	1010	1210	1880	2060	4	< 0.01	< 0.01	0.0126	0.155	0.2	4	4.16	4.25	5.21	8.49	9.35
3	1660	1660	1830	1930	1930	3	0.0292	0.0292	0.0445	0.104	0.104	3	34.1	34.1	38	43.4	43.4
3	1600	1600	1630	1690	1690	3	< 0.01	< 0.01	0.163	0.48	0.48	3	21.3	21.3	39.1	39.2	39.2
5	4040	4090	4350	4530	4670	5	< 0.01	0.0122	0.0309	0.133	0.221	5	33.1	44.1	56.9	82.9	91.8
2	2910	*	5380	*	7840	2	0.0122	*	0.0669	*	0.122	2	34.9	*	40.3	*	45.8
4	1740	1950	3100	4780	5170	4	0.017	0.029	0.09	0.384	0.473	4	15.9	19.4	30.2	48.8	54.9
6	3110	3690	4130	5020	5870	6	0.107	0.126	0.327	0.859	1.37	6	50.9	53.3	63.1	79.8	91.1
10	2230	3270	4390	4830	6690	10	0.0134	0.0339	0.0913	0.17	0.717	10	31	46.6	72.1	83.7	129
3	771	771	837	3220	3220	3	0.0104	0.0104	0.0106	0.0329	0.0329	3	5.31	5.31	13.5	24.6	24.6
6	465	567	776	1060	1390	6	< 0.01	< 0.01	< 0.01	0.0467	0.168	6	3.41	6.32	9.11	15.2	17.6
12	1680	1810	1930	2070	2250	12	0.035	0.0461	0.0816	0.184	0.453	12	23.4	31	33	35.5	46.6
5	1670	1790	1960	2030	2090	5	0.0145	0.0146	0.034	0.139	0.241	5	21.9	22.5	26	29	31.8
2	1560	*	1610	*	1650	2	0.0331	*	0.0595	*	0.0858	2	20.1	*	20.2	*	20.2
8	1920	1950	2000	2250	2490	8	0.0124	0.0221	0.0289	0.066	0.0736	8	18.3	21.1	23.2	27.2	31
43	767	925	1040	1230	1530	43	< 0.01	0.0154	0.044	0.248	0.533	43	15.8	19.2	21.7	24.9	32.8
94	554	789	864	980	1500	94	< 0.01	< 0.01	0.0146	0.0258	0.347	94	9.63	13.7	16.5	18.8	29.1
3	477	477	794	1530	1530	3	0.0124	0.0124	0.0467	0.0473	0.0473	3	3.06	3.06	5.02	8.05	8.05
9	642	901	1110	1180	1330	9	0.0105	0.0173	0.0353	0.0632	0.186	9	3.11	4.6	5.76	6.91	8.88
3	627	627	1460	2070	2070	3	0.106	0.106	0.256	2.17	2.17	3	57.8	57.8	98.7	147	147
3	1160	1160	1840	2020	2020	3	0.0138	0.0138	0.0297	0.0477	0.0477	3	10.6	10.6	36.2	49.4	49.4
2	2550	*	4320	*	6090	2	0.102	*	0.108	*	0.114	2	7.17	*	12.9	*	18.6
38	848	1200	1320	1500	1750	38	0.0934	0.233	0.303	0.406	0.735	38	17.7	24.3	28.6	31	44.1
5	4720	5910	7750	12100	16300	5	0.0695	0.0742	0.138	0.37	0.44	5	19.9	25.6	36.6	60.3	68.6
9	4550	5420	6760	10400	12500	9	0.0443	0.0562	0.0815	0.296	0.461	9	21.2	30.4	44.9	62.8	124
3	2980	2980	4250	6370	6370	3	0.0451	0.0451	0.0816	0.169	0.169	3	49.7	49.7	59.1	61.2	61.2
21	191	219	286	961	1200	21	0.0128	0.0215	0.0277	0.0449	0.138	21	12.0	13.0	14.9	16.6	23.3
40	<3	1510	1730	1920	4290	40	0.0144	0.161	0.276	0.362	0.768	40	< 0.5	18.5	21.3	24.4	49
5	1310	1350	1760	1880	1890	5	< 0.01	0.0162	0.0276	0.0672	0.083	5	16.2	17.4	19.3	22.8	25.5
2	2310	*	2450	*	2600	2	0.0487	*	0.0745	*	0.1	2	34.7	*	35	*	35.4
4	3100	3180	3840	4770	4940	4	0.115	0.115	0.176	0.647	0.785	4	24.8	25.3	26.8	29.8	30.8
4	2550	3080	5090	5760	5840	4	< 0.01	< 0.01	0.0186	0.0565	0.0666	4	16.3	16.6	22.1	29.7	30.7
2	770	*	806	*	841	2	< 0.01	*	< 0.01	*	< 0.01	2	7.52	*	8.22	*	8.92
2	2350	*	2430	*	2520	2	0.0822	*	0.126	*	0.17	2	29.3	*	33	*	36.7
5	1890	1960	2190	3360	3830	5	0.0451	0.051	0.0569	0.128	0.178	5	22.4	23.6	25.3	31.4	37.5

survey (generally ± 25 m) to the necessary generalisation of national soil maps at the 1:250 k scale, with localised variation in soil types either not being mapped at the national scale or the boundaries generalised for clarity. For example, Ca concentrations for samples S3016 and S3017 were 25,700 and 29,600 mg kg⁻¹, respectively (Supplementary Table 5). The samples were noted in the field as yellow-grey sandy soils but were mapped as Luvisols, which normally have a reddish colour. Overall, despite some inaccuracies in mapping, the higher pH and Ca concentrations of calcareous compared to non-calcareous soils provides confidence in the approach.

3.2. Plant composition, estimates of dietary mineral supplies and deficiency risks and the influence of soil type

The elemental composition of plant samples is presented in Supplementary Table 6 and summary statistics according to plant and soil type for Ca, Cu, Fe, Mg, Se and Zn are shown in Supplementary Table 7. Samples that have been processed in a way likely to influence mineral composition (milling of grain, boiling of leaves, etc.) are distinguished. Commonly consumed items with n > 1 are presented in Table 2.

3.2.1. Calcium

Total Ca concentration in maize grain from calcareous soils (median 43.1 mg kg⁻¹, range 7.53–87.1, n = 40) was greater than in grain from non-calcareous soils (median 30.9 mg kg⁻¹, range 10.7–163, n = 94; p = 0.008; Table 3). Similarly, Ca concentration in vegetable leaf samples from calcareous soils (median 19,700 mg kg⁻¹, range 5,160–51,700, n = 57) was greater than in samples from non-calcareous

soils (median 14,100, range 3,780–38,900, *n* = 61; *p* = 0.017; Supplementary Table 8).

The significant positive correlation (r = 0.646, p < 0.001) between measured plant concentrations of Ca in the present study and published data when matched by FBS item (Supplementary Table 9) suggests that published Ca composition data are relevant for dietary assessments in Malawi. Estimated Ca supplies were 368, 430 and 367 mg capita⁻¹ d⁻¹ for non-calcareous soils, calcareous soils and published composition data, respectively. These estimates are much lower than the adult female and national mean EARs of 625 and 632 mg *capita*⁻¹ d⁻¹, respectively, and the estimated risk of dietary Ca deficiency was >97% for all data sources (Table 4, Supplementary Table 9). Insufficient dietary supply and a high risk of Ca deficiency in Malawi have been reported in previous studies based on FBSs (Broadley et al., 2012; Joy et al., 2014) and dietary recall or food records (Ferguson et al., 1989; Gibson and Huddle, 1998; Table 1). Estimated supplies of Ca are lower and deficiency risks are greater in this study than in Joy et al. (2014), mainly due to updated information on the supply of the FBS item 'Freshwater Fish'. Previously, a moisture content based on processed fish had been used, whereas 'live weight equivalent' should be used (FAOSTAT, 2014).

Agronomic options to address Ca deficiency are limited because Ca is restricted to xylem transport and therefore grain generally has low Ca concentrations, i.e., up to two orders of magnitude less than those in leaves (Fig. 2; White and Broadley, 2009; Broadley and White, 2010). For example, the median Ca concentration in cabbage leaves from all soil types in this study was 6,580 mg kg⁻¹ (range 3,780–32,300, n = 6) compared to 34.2 mg kg⁻¹ (range 7.53–392, n = 152) in maize grain from all soil types. However, finger millet grain has a relatively high Ca

Table 3

One-way ANOVA (log₁₀ transformed data) to test influence of calcareous/non-calcareous soil type on maize grain elemental concentrations.

	Source	d.f.	S.S	m.s.	v.r.	F pr.
Calcium	Soil type	1	0.35	0.353	7.29	0.008
	Residual	132	6.40	0.049		
	Total	133	6.75	0.051		
Copper	Soil type	1	0.50	0.495	4.88	0.029
	Residual	135	13.70	0.101		
	Total	136	14.20	0.104		
Iron	Soil type	1	0.99	0.985	43.5	< 0.00
	Residual	121	2.74	0.023		
	Total	122	3.73	0.031		
Magnesium	Soil type	1	0.17	0.171	24.8	< 0.001
	Residual	135	0.93	0.007		
	Total	136	1.10	0.008		
Selenium	Soil type	1	7.69	7.690	36.3	< 0.001
	Residual	135	28.60	0.212		
	Total	136	36.30	0.267		
Zinc	Soil type	1	0.46	0.463	59.4	< 0.001
	Residual	135	1.19	0.009		
	Total	136	1.66	0.012		

concentration of 3,290 mg kg⁻¹ (range 2,310–4,700, n = 7) and could be a good source of dietary Ca as it can be used in place of maize flour to make the staple dish *nsima*.

3.2.2. Copper

Total Cu concentration in maize grain from calcareous soils (median 2.48 mg kg⁻¹, range 1.2–19.4, n = 43) was greater than in grain from non-calcareous soils (median 1.67 mg kg⁻¹, range 0.72–421, n = 94; p = 0.029; Table 3). Similarly, Cu concentration in leafy vegetables from calcareous soils (median 10.8 mg kg⁻¹, range 4.97–67.2, n = 57) was greater than in those from non-calcareous soils (median 9.39 mg kg⁻¹, range 1.09–40.2, n = 61; p = 0.034, Supplementary Table 8). No significant correlation between composition data presented here and published sources was found for either soil type using either linear or non-linear analysis methods. Compared to an adult female EAR of 0.7 mg capita⁻¹ d^{-1} , estimated Cu supplies were 2.0, 2.6 and 3.0 mg capita⁻¹ d⁻¹ based on non-calcareous soils, calcareous soils and published composition data, respectively, with estimated risks of dietary deficiency being <1% in each case (Table 4, Supplementary Table 9). Estimates of low Cu deficiency risk are consistent with previous studies (Joy et al., 2014).

3.2.3. Iron

Total Fe concentration in maize grain from calcareous soils (median 21.5 mg kg⁻¹, range 10.8–96.7, n = 33) was greater than in that from non-calcareous soils (median 14.2 mg kg⁻¹, range 9.06–34.4, n = 90; p = 0.003; Table 3). Similarly, Fe concentration in leafy vegetables from calcareous soils (median 783 mg kg⁻¹, range 48.0–13,300, n = 57) was greater than in samples from non-calcareous soils (median 308 mg kg⁻¹, range 61.2–3,040, n = 61; p < 0.001; Supplementary

Table 4

National mean dietary supply and deficiency risk estimates in Malawi using FAO food supply data matched to plant composition data for calcareous and non-calcareous soils. Results using published composition data are presented for comparison.

Composition data source	Ca	Cu	Fe	Mg	Se	Zn
	Supply	v (mg ca	pita ⁻¹ d ⁻	1)		
Non-calcareous	368	2.0	18.4	632	0.0188	10.1
Calcareous	430	2.6	23.2	712	0.0409	12.0
Published	367	3.0	29.2	753	0.0302	11.7
	Risk o	Risk of deficiency (%)				
Non-calcareous	100	0.2	3.8	0.1	100	57
Calcareous	97	0.1	1.2	0.1	34	31
Published	100	0.1	0.5	0.1	80	34

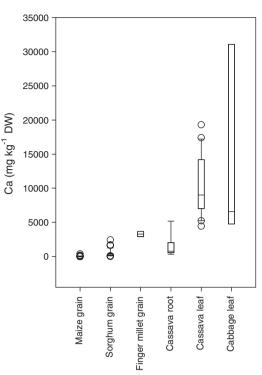


Fig. 2. Concentration (mg kg⁻¹ dry-weight, DW) of calcium (Ca) in selected leaf, grain and root crop samples. Boxes represent Q1, median and Q3; whiskers represent 90th and 10th percentiles; circles represent outliers.

Table 8). No significant correlation between composition data presented here and published sources was found for either soil type using either linear or non-linear analysis methods. Estimated Fe supplies were 18.4, 23.2 and 29.2 for non-calcareous soils, calcareous soils and published composition data, respectively (Supplementary Table 9). Risks of dietary Fe deficiency are <4% using the EAR cut-point method (Table 4; Supplementary Table 9). However, requirements are not normally distributed so this is likely to underestimate risk of deficiency; estimated supplies appear adequate for the majority of the population (EAR for adult women is 13.4 mg *capita*⁻¹ d⁻¹) but not for pregnant women (EAR 41.2 mg *capita*⁻¹ d⁻¹).

The much greater estimated supply obtained using published composition data is mainly due to the FBS items 'maize' and 'potatoes'. Concentrations of Fe in maize (including refined flour) were 19.6, 25.2 and 39.0 mg kg⁻¹ in non-calcareous, calcareous and published data sources, respectively, while the corresponding values for potato were 32.1 and 155 mg kg⁻¹ in non-calcareous and published sources (no data for calcareous soils).

In a sample of adult women in rural Malawi, Siyame et al. (2013) reported most Fe intakes to be adequate following analysis of dietary composites and blood samples, and that the Fe content of diets containing food crops grown on calcareous soils (median 29.6 mg *capita*⁻¹ d⁻¹, range 9.10–90.1, n = 58) was higher than in those grown on non-calcareous soils (median 16.6 mg capita⁻¹ d⁻¹, range 3.80–49.3, n = 55), consistent with the findings of the present study. However, Fe status was significantly better in the noncalcareous area, as measured by plasma ferritin and body Fe levels. This apparently contradictory finding might be explained by greater levels of inhibitors of Fe-absorption in Mikalango diets compared to those in Zombwe (Sivame et al., 2013). In addition, further investigation is required to determine whether Fe from soil dust is available for absorption in humans and whether this depends on the soil type, particularly in view of the significant contribution of soil to concentrations of Fe in samples of seed (33.8%) and leaf (76.7%; Section 3.3).

3.2.4. Magnesium

Total Mg concentration in maize grain from calcareous soils (median 1040 mg kg⁻¹, range 767–1,530, n = 43) was greater than in grain from non-calcareous soils (median 864 mg kg⁻¹, range 554–1,500, n = 94; p < 0.001; Table 3). However, there was no significant influence of soil type on Mg concentration in leafy vegetables (Supplementary Table 8). There was no significant correlation between Mg concentrations of plant samples from non-calcareous soils and published data. However, the correlation between Mg concentrations of plant samples from calcareous soils and published data was significant using nonlinear analysis (p = 0.011, Supplementary Table 9). Estimated Mg supplies were 632, 712 and 753 mg *capita*⁻¹ d⁻¹ for non-calcareous soils, calcareous soils and published composition data, respectively. These estimates are greater than adult female and national mean EARs of 183 and 155 mg capita⁻¹ d⁻¹, respectively, and estimated risks of deficiency were <1% for both soil types and using published composition data (Table 4; Supplementary Table 9). Low risks of dietary Mg deficiency in Malawi have been reported previously (Broadley et al., 2012; Joy et al., 2014).

3.2.5. Selenium

Total Se concentration in maize grain from calcareous soils (median 0.044 mg kg⁻¹, range 0.005–0.533, n = 43) was greater than in grain from non-calcareous soils (median 0.015, range 0.004-0.347, n = 94; p < 0.001; Table 3; Fig. 3). Similarly, Se concentration in leafy vegetables from calcareous soils (median 0.572 mg kg⁻¹, range 0.012–3.82, n = 57) was greater than in samples from noncalcareous soils (median 0.082, range 0.007–1.72, n = 61; *p* < 0.001; Fig. 3; Supplementary Table 8). There was no significant correlation between Se concentrations in plant samples grown on non-calcareous soils and published data, but there was a strong positive correlation for plant samples grown on calcareous soils (Supplementary Table 9). This observation suggests that published composition data may be relevant for estimating Se supplies in areas of calcareous soils in Malawi but are unlikely to be relevant in areas of non-calcareous soils. Estimated Se supplies were 0.019, 0.041 and 0.030 mg capita⁻¹ d⁻¹ for non-calcareous soils, calcareous soils and published composition data, respectively, compared to adult female and national mean EAR values of 0.045 and 0.037 mg $capita^{-1} d^{-1}$, respectively. Estimated risks of dietary Se deficiency were 100%, 34% and 80% when using non-calcareous, calcareous and published composition data, respectively (Table 4; Supplementary Table 9). The lower concentration of Se in plant samples grown on non-calcareous soils compared to calcareous soils is expected due to the limited phyto-availability of Se at soil pH <6.5 and substantiates similar findings in a national survey of Se concentrations in maize grain (Fordyce, 2005; Chilimba et al., 2011). Furthermore, the wide geographic coverage of this survey reinforces the findings of Hurst et al. (2013) that dietary Se deficiency is likely to be widespread in Malawi where non-calcareous soils represent 69% of total land area.

The fortification of maize grain through application of Se-enriched fertilisers is a promising strategy to alleviate Se deficiencies in Malawi (Chilimba et al., 2012). For diet-based strategies, animal products may contain higher concentrations of Se, while the leaves of moringa (*Moringa oleifera* Lam., Moringaceae) had the greatest Se concentration of all commonly consumed crop types, with median concentrations of 0.772 and 0.504 mg kg⁻¹ on calcareous and non-calcareous soil types, respectively. Median Se concentration in mushroom (n = 3) was 0.256 mg kg⁻¹, although one sample had a concentration of 2.17 mg kg⁻¹, c. 10-fold greater than the median total Se concentration in Malawian soils. A great variety of Se concentrations in the fruiting body of different edible wild-grown mushrooms has been reported (Falandysz, 2008), possibly due to variation in the accumulation of Se-containing proteins.

3.2.6. Zinc

Total Zn concentration was significantly greater in leafy vegetables grown on non-calcareous soils (median 44.9, range 10.1–129, n = 61) than calcareous soils (median 24.8, range 9.32–91.1, n = 57; p < 0.001; Fig. 3; Supplementary Table 8). This is as expected since the binding of Zn to calcite and co-precipitation in Fe oxides is likely to reduce the availability of Zn to roots in calcareous soils and the solubility and lability of Zn in soil both systematically decline as pH increases (Tye et al., 2003). In contrast, Zn concentration in maize grain from calcareous soils (median 21.7, range 15.8–32.8, n = 43) was greater than in grain from non-calcareous soils (median 16.5, range 9.63-29.1; p < 0.001; Table 3; Fig. 3); the cause of this effect is unknown but is likely to be a result of plant physiology. Dickinson et al. (2014) also found a greater concentration of Zn in maize grain from alluvial soils in Malawi with mean pH 6.5 (32.6 mg kg⁻¹, n = 16) than grain from leached plateau soils with mean pH 3.9 (30.5 mg kg⁻¹, n = 38), although the difference was not significant (p = 0.145).

There was no significant correlation between the Zn concentrations of plant samples grown on non-calcareous soils and published data, but there was a strong positive correlation for plant samples grown on calcareous soils (p = 0.005, Supplementary Table 9). This suggests that published composition data may be relevant for estimating Zn supplies of populations living on calcareous soils in Malawi, but not non-calcareous soils. Estimated Zn supplies were 10.1, 12.0 and 11.7 mg *capita*⁻¹ d⁻¹ for non-calcareous soils, calcareous soils and published composition data, respectively, compared to adult female and national mean EARs of 8.2 and 10.5 mg *capita*⁻¹ d⁻¹, respectively, with estimated risks of dietary Zn deficiency of 57, 31 and 34% (Table 4; Supplementary Table 9). The substantial risks of dietary deficiency exist despite supply estimates being greater than national mean EAR because the

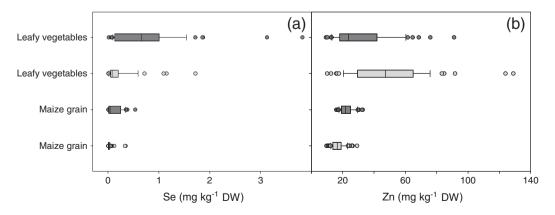


Fig. 3. Concentrations (mg kg⁻¹ dry-weight, DW) of (a) selenium, Se and (b) zinc, Zn in maize grain and leafy vegetables grown on non-calcareous (light grey boxes) and calcareous (dark grey boxes) soils. Boxes represent Q1, median and Q3; whiskers represent 90th and 10th percentiles; circles represent outliers.

EAR defines an intake level adequate for 50% of individuals in an age-sex group, and additionally there is an assumed 25% coefficient of variation in Zn intake. These results also demonstrate the sensitivity of the EAR cut-point approach, whereby a difference in dietary supply of 22% alters the estimated risk of deficiency by 44%.

3.3. Soil contamination of plant samples

There was strong evidence that presence of soil particles on plant samples influenced elemental concentrations. The greatest effect was on the Fe concentrations in leaves, with a mean contribution from soil of 76.7% (n = 116; Fig. 4). The mean contribution of soil particles to the Fe concentrations in seed was 33.8% (n = 223), while it also contributed to leaf Cu (6.2%), Se (5.5%) and Zn (5.6%) concentrations, and to root Se concentrations (9.6%). Contributions of soil particles to other mineral/ crop tissue combinations was <5%. Leaf of tove had the greatest level of soil contamination; tove (Ceratotheca sesamoides Endl. Pedaliaceae) is a creeping plant that grows wild and is commonly found in the Shire valley region in southern Malawi. Tove samples were collected during the dry season and had been dried by households for preservation, thus preventing washing and wiping of leaf fragments after sample collection. The median Fe concentration was 2,820 mg kg⁻¹ (range 878–13,300, n = 20) with a mean contribution from soil particles of 97.4%. Median Zn concentration was 19.7 mg kg⁻¹ (range 12.8–28.6) with mean contribution from soil particles of 19.7% (range 4.22–68.5; Supplementary Table 10). Other than tove, only eight individual samples contained >10% of Zn from soil particles, including three samples of pumpkin leaves and one of courgette leaves. These samples are susceptible to contamination by soil particles from dust and rain splash as they grow low to the ground. Presence of soil particles has the greatest effect on Fe as concentrations of Fe in the soil are c. 40,000 mg kg⁻¹ compared to c. 50 mg kg^{-1} for Zn.

The level of care taken to remove soil from samples in this study is likely to exceed that of households during food preparation, and it would therefore appear that a substantial proportion of total dietary Fe intake comes from soil rather than being endogenous to the plant.

3.4. Caveats

Food supply and composition data only provide a proxy for dietary mineral intakes, and estimates are subject to the accuracy of underlying data. FBS data can over-estimate supply due to non-accounting of household-level waste, or under-estimate supply when subsistence production or foods gathered from the wild are not captured. Moreover, the use of the EAR cut-point method to estimate deficiency risks is subject to major caveats such as requiring a normal and uni-modal distribution of intakes and requirements (IOM, 2000b).

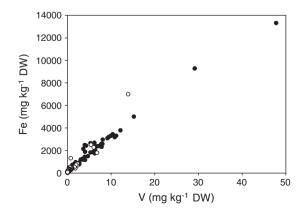


Fig. 4. Concentrations (mg kg⁻¹dry-weight, DW) of vanadium (V) and iron (Fe) in plant samples: open circles seed; closed circles leaf. Concentration of V is a proxy for level of contamination from soil particles.

Estimates of dietary mineral supplies in this study are generally higher than estimates derived from analyses of composite diets (e.g., Gibson and Huddle, 1998; Siyame et al., 2013). In addition, the finding of low deficiency risks for Fe is surprising considering the high rates of anaemia associated with Fe-deficiency in sub-Saharan Africa (WHO, 2008) but is consistent with the reporting of adequate dietary Fe supplies by Siyame et al. (2013), as is the finding of high risks of Zn deficiency (Siyame et al., 2013; Dickinson et al., 2014), and Se deficiency on non-calcareous soils (Hurst et al., 2013; Dickinson et al., 2014).

3.4.1. Influence of processing

Preparation of the most commonly consumed form of maize flour in Malawi (*ufa woyera*) involves soaking the grain in water for several days before drying, winnowing and pounding or milling. This process removes the bran and germ leaving the starch-rich endosperm. The effect of processing on elemental concentrations was studied using *ufa woyera* and maize grain samples from Zombwe EPA in northern Malawi (Table 5).

The increase in Ca concentration during processing is likely to be due to the soaking process as further studies revealed a very high Ca concentration (209 mg L^{-1}) in water from a borehole near the sampling location. Decreases in Cu, Mg and Zn concentrations due to processing are likely to result from the loss of bran and germ, which have higher concentrations of minerals than the endosperm (Tang et al., 2008; Cakmak et al., 2010). However, as Se has a more even distribution across different grain fractions (Hart et al., 2011), the decrease during processing might be due to the soaking process.

4. Conclusions

Soil type influences the mineral concentrations of food crops in Malawi as the concentrations of Ca, Cu, Fe, Mg, Se and Zn were higher in maize grain from calcareous than non-calcareous soils, and concentrations of Ca, Cu, Fe and Se were higher, but Zn concentration was lower, in leafy vegetables grown on calcareous than non-calcareous soils. Use of soil-specific composition data can improve estimates of dietary mineral supplies considering that the majority of households in Malawi engage in subsistence agriculture. We provide new nationalscale estimates of dietary mineral supplies using FBS food supply data which show that the risk of deficiency due to inadequate dietary supply is likely to be high for Ca (100%), Se (100%) and Zn (57%) among subsistence populations living on non-calcareous soils, which cover 69% of land area in Malawi. Deficiency risks on calcareous soils are high for Ca (97%), but lower for Se (34%) and Zn (31%). Risks of Cu, Fe and Mg deficiencies due to inadequate dietary supply are likely to be low, based on supply data. Further work is required to understand the influence of contaminant soil particles on Fe intake and its bioavailability during digestion. Results are dependent on the accuracy of supply data and might be improved through use of household survey data, expansion of food items sampled and determination of the influence of food processing on mineral concentrations.

Table 5

Elemental concentrations in maize grain and flour samples from Zombwe EPA, northern Malawi and results of a one-way ANOVA.

		Ca	Cu	Fe	Mg	Se	Zn
		mg kg	- 1				
Grain $(n = 15)$	Median	41.1	2.85	24.5	974	0.0181	16.30
	Min	14.2	0.52	14.6	150	< 0.001	1.95
	Max	392.0	13.20	58.4	1420	0.0472	24.70
Flour $(n = 14)$	Median	134.0	0.76	24.4	153	0.0070	2.50
	Min	5.5	0.45	10.5	96	< 0.001	1.15
	Max	189.0	1.64	76.0	929	0.0160	15.60
	р	0.151	< 0.001	0.656	< 0.001	0.038	< 0.001

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx. doi.org/10.1016/j.scitotenv.2014.10.038.

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