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Historical variation in the mineral composition of edible horticultural products

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SUMMARY
Historical variation in the mineral composition of edible horticultural products was determined from UK and USA food survey data. From these data, it was possible to measure the variation in the mineral composition of edible horticultural products in general, and in edible horticultural products grouped as vegetables, fruits or nuts, in the 1930s and in the 1980s (or later) for both countries. Thus, the hypothesis that the mineral composition of edible horticultural products had altered since the 1930s was tested. The average concentrations of Cu, Mg and Na in the dry matter of vegetables, and the average concentrations of Cu, Fe and K in fruits decreased significantly between the 1930s and the 1980s in the UK. The same hypothesis was tested with comparable data from the USA, whose historical horticultural and consumer practices have paralleled those of the UK. Data from the USA showed that the average Ca, Cu and Fe concentrations in the dry matter of vegetables, and the average concentrations of Cu, Fe and K in fruits had decreased significantly since the 1930s. There were insufficient data to determine if the mineral composition of any single edible horticultural species had altered significantly over time either in the UK or in the USA. The nutritional implications of this study are discussed. Since horticultural products in general, and fruits and nuts in particular, are relatively small contributors of minerals to the average UK diet, historical changes in mineral composition are unlikely to be significant in overall dietary terms.

Mayer (1997) presented evidence, based on a comparison of the data from McCance and Widdowson (1960) and Holland *et al*. (1991b), which indicated that the average dry matter content and mineral concentrations in fresh vegetables and fruits available in the UK had decreased since the 1930s. Since Mayer's paper, this sentiment has been repeated in the popular press, where the decrease in mineral concentrations in UK vegetables and fruits is often attributed to modern breeding and/or cultural practices. Thus, it is important to ascertain the statistical validity of these claims, since, if they are correct, there are opportunities to improve crop genotypes and cultural practices.

Here, we test the hypothesis that the mineral composition of vegetables, fruits and nuts available in the UK has altered since the 1930s. Data were obtained from the original notebooks of McCance and Widdowson (1929–1944) and from the Sixth Summary Edition of McCance and Widdowson's *The Composition of Foods* (Food Standards Agency, 2002). In contrast to the study by Mayer (1997), mineral concentration data were expressed on a dry weight (DW) basis to remove the variations caused by tissue hydration. Further, the same hypothesis was tested with comparable data from the USA, whose historical horticultural and consumer practices have paralleled those in the UK.

Since horticultural products in general, and fruits and nuts in particular, are relatively small contributors of minerals to most UK diets, historical changes in their mineral composition are unlikely to impact significantly on dietary intakes. However, if considered desirable, crop genotypes with higher mineral concentrations could be selected or bred, whilst agronomic interventions based on micro-nutrient fertilisation strategies could also be used to alter crop mineral compositions.

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MATERIALS AND METHODS

Primary data

Data on Ca, Cl, Cu, Fe, K, Mg and Na concentrations in horticultural produce available in the UK between 1934–1935 were taken from the laboratory notebooks of McCance and Widdowson (*Analysis of Foodstuffs, 1929–1944*. The Wellcome Library for the History and Understanding of Medicine, London. GC/97/A). Data on P concentrations were taken from McCance *et al*. (1938). These data were compared with the most recent analyses of UK produce, contained in the Sixth Summary Edition of McCance and Widdowson's *The Composition of Foods* (Food Standards Agency, 2002). However, it should be noted that the data for mineral concentrations in raw vegetables, fruits and nuts in this publication were taken from supplements to the Fourth and Fifth Editions published by Holland *et al*. (1991a; 1992), which are mostly derived from analyses undertaken between 1984–1987. The Ca, Cu, Fe, K, Mg and P concentrations in produce available in the USA in the 1940s were taken from Beeson (1941). These data were compared with values abstracted from the United States Department of Agriculture (USDA) National

Nutrient Database for Standard Reference, Release 16 (http://www.nal.usda.gov/fnic/foodcomp/Data), which represent the mineral concentrations in present-day produce. All data were expressed on a DW basis (Tables I and II). Clearly, the data must be interpreted subject to the *caveat* that analytical techniques have changed and become more sensitive since the 1930s.

Statistical analyses

The hypothesis that DW-based mineral concentrations in vegetables, fruits and nuts had altered since the 1930s was tested for each mineral element. A "new/old" quotient was calculated for each crop and mineral element (Table III). The natural logarithm of the "new/old" quotient was taken and a one sample, two-tailed, *t*-test was performed with the null hypothesis that the logarithm of the quotient was equal to zero (Tables IV and V). A value significantly less than zero indicated a decline in mineral concentration, and a value significantly greater than zero indicated an increase in mineral concentration. Since each comparison was based on independent data for each mineral, no *post hoc* Type I error corrections were applied. All analyses were performed using GenStat (Release 6.1.0.200; VSN International, Oxford, UK).

RESULTS

The data of McCance and Widdowson (1929–1944), McCance *et al*. (1938) and Holland *et al*. (1991a; 1992) were not originally intended to be used to document historical trends in the mineral composition of horticultural produce. Instead, they are "representative" values of mineral concentrations for different types of produce (Tables I and II). Occasionally, data are given for different varieties of a vegetable or fruit, but this is not common. The sampling procedure for each vegetable, fruit or nut was to purchase a representative sample from several sources and to bulk these together prior to mineral analysis. Multiple analyses of the composite samples were then performed. Unfortunately, this sampling procedure does not allow the biological or genetic variation within a particular crop to be assessed. Without knowledge of this variation, it cannot be determined whether the representative values of mineral concentrations cited for a particular vegetable, fruit or nut differed significantly between the 1930s and the 1980s. Thus, there are insufficient data in these publications to test the hypothesis that the mineral composition of any *individual* horticultural crop product has declined. However, values for "produce in general" can be compared between the 1930s and 1980s, and therefore an estimate of variation can be derived for the changes in mineral concentrations in many different crops (Tables IV and V).

The "new/old" quotient for different crops and mineral elements varied considerably, with the concentrations of some minerals in some crops declining, and the concentrations of some minerals in some crops increasing. Statistical analyses indicated that the average concentrations of Cu, Mg and Na in the dry matter of vegetables, and the average concentrations of Cu, Fe and K in the dry matter of fruits available in the UK decreased significantly between the 1930s and the 1980s (Table IV). This is consistent with the observations of Mayer (1997), who undertook an analysis of the mineral concentrations in 20 raw vegetables and 20 fresh fruits. On a fresh, tissue weight basis, reductions in dry matter content and concentrations of Ca, Cu, Mg and Na in raw vegetables and Cu, Fe, K and Mg in fresh fruits available in the UK were noted over this period (Mayer, 1997). Interestingly, although the average Mg concentration in dried fruits available in the UK had decreased, the average Cu concentration in dried fruits had increased, and a significant increase in the average Cu concentration in nuts was also observed (Table IV).

The average concentrations of Ca, Cu and Fe in vegetables available in the USA have decreased significantly since the 1930s (Table V). The decline in Cu is consistent with the decline in Cu in vegetables in the UK over the same period. A decline in Ca and Fe in vegetables is also consistent with a recent analysis of USDA data on horticultural products between 1950 and 1999 (Davis *et al*., 2004). Furthermore, our data indicate that the average concentrations of Cu, Fe and K in fruits available in the USA have decreased significantly since the 1930s (Table V), which is entirely consistent with data for fruits available in the UK (Table IV). A decrease in Fe concentrations in dried fruits available in the USA since the 1930s was also observed (Table V). However, there was no significant difference in mineral concentrations in nuts in the USA since the 1930s.

DISCUSSION

It can be concluded that the average concentrations of Cu, Mg and Na in the dry matter of vegetables and the average concentrations of Cu, Fe and K in the dry matter of fruits available in the UK have decreased significantly between the 1930s and the 1980s (Table IV). Since the average concentrations of Cu in the dry matter of vegetables, and the average concentrations of Cu, Fe and K in the dry matter of fruits, have also decreased since the 1930s in the USA (Table V), these phenomena might reflect modern agronomic practices. It is noteworthy that modern fertilisers have lower levels of contaminating metals, and that there has been a major reduction in the use of Cu-containing pesticides in conventional agricultural and horticultural production systems since the 1930s. However, historical changes are unlikely to have significant dietary impacts in the UK.

In dietary terms, the UK population consumes more Na per head d^{-1} than the recommended daily intake (Food Standards Agency, 2003), while table salt can be replaced by sodium salt substitutes containing KCl. Thus, any decrease in the K composition of horticultural produce is unlikely to be an important dietary issue. However, decreases in the average concentration of Cu in vegetables, and in the average concentrations of Cu and Fe in fruits, warrant further consideration.

In animals, tissue Cu concentrations are under tight homeostatic control. Although Cu is an essential element, it is toxic at high concentrations. The estimated safe and adequate dietary intake of Cu is 1.2 – 3.0 mg d^{-1} (Food Standards Agency, 2003). Copper is necessary for the activity of several key enzymes, such as cytochrome *c* oxidase, amino acid oxidase, superoxide dismutase and monoamine oxidase, and Cu has been implicated in host

TABLE I

Mineral concentrations of UK vegetables, fruits and nuts recorded in the laboratory notebooks of McCance and Widdowson (1929-1944) and published by McCance et al. *(1938), or published by the Food Standards Agency (2002)*

	McCance and Widdowson (1929-1944)								Food Standards Agency (2002)							
	K	Ca	Mg	Fe	Cu	Na	Cl	P	K	Ca	Mg	Fe	Cu	Na	Cl	P
Vegetables																
Potato (old)	2326[1]	32	100	3.9	0.60	27	324	167	1714	24	81	1.9	0.38	3	314	176
Butter bean	1867	93	180	6.5	1.30	67	51	348	1923	96	215	6.7	0.84	45	53	362
Haricot bean	1260	195	199	7.2	0.70	47	2	336	1308	203	203	7.6	0.69	48	2	349
Lentil	720	41	82	8.1	0.60	39	68	259	926	69	108	10.5	0.90	27	85	376
Runner bean	3236	391	270	8.7	1.10	76	267	304	2500	375	216	13.6	0.23	1	239	386
Pea	1536	70	152	8.2	0.90	4	172	482	1299	83	134	11.0	0.20	4	154	512
Aubergine	3601	157	145	5.8	1.30	38	925	183	2958	141	155	4.2	0.14	28	197	225
Cabbage (red)	2932	517	160	5.5	0.90	307	432	312	2688	645	97	4.3	0.11	86	484	398
Carrot (old)	2118	455	113	5.3	0.80	900	649	199	1667	245	29	2.9	0.20	245	324	147
Celery	4257	799	131	6.5	1.60	2098	2795	485	6531	837	102	8.2	0.20	1224	2653	429
Chicory	4789	485	332	18.0	3.60	193	363	551	2982	368	105	7.0	0.88	18	439	474
Cucumber	3930	634	253	8.4	2.60	362	683	672	3889	500	222	8.3	0.28	83	472	1361
Lettuce	4308	535	201	15.2	3.10	63	818	626	4490	571	122	14.3	0.20	61	959	571
Mushroom	5517	34	155	12.1	7.60	107	998	1607	4324	81	122	8.1	9.73	68	932	1081
Mustard and cress	4490	880	364	63.2	1.50	254	1188	874	2340	1064	468	21.3	0.21	404	830	702
Onion	1881	428	104	4.1	1.10	140	281	411	1455	227	36	2.7	0.45	27	227	273
Parsnip	1149	314	128	3.3	0.50	94	232	395	2174	198	111	2.9	0.24	48	237	357
Pumpkin	5821	735	154	7.2	1.40	25	689	366	2600	580	200	8.0	0.40	–	740	380
Radish	3590	654	170	28.0	1.90	883	280	405	5217	413	109	13.0	0.22	239	804	435
Spring onion	1711	1023	83	9.4	1.00	98	269	179	3333	500	154	24.4	0.77	90	397	372
Swede	1574	653	124	3.2	0.40	604	353	220	1932	602	102	1.1	0.11	170	352	455
Tomato	4338	200	166	6.5	1.60	42	769	321	3623	101	101	7.2	0.14	130	797	348
Turnip	3563	880	110	5.0	0.60	868	1048	412	3182	545	91	2.3	0.11	170	443	466
Watercress	3529	2499	191	18.2	1.50	675	1756	585	3067	2267	200	29.3	0.13	653	2267	693
Endive	6062	698	165	44.1	1.40	161	1122	1058	6032	698	159	44.4	0.16	159	1127	1063
Horseradish	2287	470	141	8.0	0.60	31	74	277	2292	474	142	7.9	0.91	32	75	277
Fruits																
Apples, eating	754	23	30	1.9	0.70	15	8	49	774	26	32	0.6	0.13	19	1	71
Apples, cooking	852	25	20	2.0	0.70	15	31	112	715	33	24	0.8	0.16	16	16	57
Apricots	2394	129	92	2.8	0.90	0	0	159	2109	117	86	3.9	0.47	16	23	156
Avocado pear	2118	82	157	2.8	1.10	86	32	165	1636	40	91	1.5	0.69	22	22	142
Bananas	1187	23	142	1.4	0.60	4	271	96	1606	24	137	1.2	0.40	4	317	112
Blackberries	1150	350	163	4.7	0.60	20	122	132	1067	273	153	4.7	0.73	13	147	207
Cherries, eating	1489	86	52	2.0	0.40	15	0	91	1221	76	58	1.2	0.41	6	1	122
Cherries, cooking	1510	100	57	1.5	0.50	20	0	103	–	–	–	–	–	–	–	–
Cranberries	922	114	65	8.6	1.10	14	0	87	731	92	54	5.4	0.38	15	1	85
Currants, black	1646	267	76	5.6	0.60	12	65	191	1637	265	75	5.8	0.62	13	66	190
Currants, red	1595	207	74	7.1	0.70	13	81	171	1628	209	76	7.0	0.70	12	81	174
Currants, white	1738	134	76	5.5	0.80	9	64	167	1737	132	78	5.4	0.84	12	66	168
Custard Apple	2165	45	90	2.0	0.60	52	150	191	1082	78	127	2.2	0.56	26	–	131
Damsons	1289	104	49	1.8	0.40	12	0	73	1289	107	49	1.8	0.36	9	0	71
Figs	1739	222	130	2.7	0.40	10	119	209	1299	247	97	1.9	0.39	19	117	97
Gooseberries, unripe	2087	281	70	3.2	1.30	19	65	337	2121	283	71	3.0	0.61	20	71	343
Gooseberries, ripe	1041	113	53	3.5	0.90	7	66	117	1717	192	91	6.1	0.61	10	111	192
Grapes	1414	58	26	1.7	0.50	8	5	95	1154	71	38	1.6	0.66	11	1	99
Grapefruit	2509	184	111	2.8	0.70	14	14	167	1818	209	82	0.9	0.18	27	27	182
Greengages	1397	77	35	1.7	0.40	6	0	104	1722	94	44	2.2	0.44	6	6	128
Lemons, whole	1099	725	78	2.4	1.80	40	34	140	1095	620	88	3.6	1.90	36	36	131
Loganberries	1711	234	166	9.1	0.90	16	105	162	1733	233	167	9.3	0.93	20	107	160
Medlars	963	118	41	1.9	0.70	24	12	110	980	118	43	2.0	0.67	24	12	110
Melons, cantaloupe	4906	300	317	12.7	0.70	212	684	478	2658	253	139	3.8	0.01	101	557	165
Mulberries	1718	238	101	10.5	0.40	14	24	319	1733	240	100	10.7	0.40	13	27	320
Nectarines	1354	20	64	2.3	0.30	46	24	121	1532	63	90	3.6	0.54	9	45	198
Oranges	1409	296	92	2.3	0.50	21	23	170	1079	338	72	0.7	0.36	36	22	151
Passion Fruit	1303	58	145	4.2	0.40	106	137	203	797	44	116	5.2	–	76	–	255
Peaches	1872	35	57	2.8	0.30	20	0	134	1441	63	81	3.6	0.54	9	1	198
Pears, eating	705	43	37	1.2	0.80	14	3	67	926	68	43	1.2	0.37	19	6	80
Pineapple	1566	77	107	2.7	0.50	10	181	50	1185	133	119	1.5	0.81	15	215	74
Plums, Victoria Dessert	1184	69	45	2.2	0.60	11	0	103	1357	79	50	2.9	0.71	14	–	114
Plums, cooking	1313	93	53	2.0	0.60	13	0	98	1491	81	50	2.5	0.62	12	1	143
Pomegranate	1406	20	22	1.0	0.50	8	362	52	–	–	–	–	–	–	–	–
Quinces	1284	88	38	2.0	0.80	20	12	120	1266	89	38	1.9	0.82	19	13	120
Raspberries	1333	242	129	7.2	1.30	15	132	171	1308	192	146	5.4	0.77	23	169	238
Rhubarb	7253	1754	232	6.7	2.20	37	1485	358	5000	1603	224	5.2	1.21	52	1500	293
Strawberries	1444	197	105	6.4	1.10	14	157	206	1524	152	95	3.8	0.67	57	171	229
Tangerines	1175	315	85	2.0	0.70	17	18	127	1203	316	83	2.3	0.08	15	15	128
Watermelon	3610	239	231	4.2	0.60	338	781	151	1299	91	104	3.9	0.39	26	–	117
Dried fruits																
Apricots	2208	108	76	4.8	0.30	66	40	138	2204	108	76	4.8	0.47	66	41	141
Currants	908	122	46	2.3	0.60	25	20	52	854	110	36	1.5	0.96	17	19	84
Dates	882	79	68	1.9	0.20	6	339	75	820	53	48	1.5	0.30	12	433	70
Figs	1220	341	111	5.0	0.30	104	200	110	1165	301	96	5.1	0.36	75	207	107
Peaches	1307	42	64	8.0	0.70	7	12	137	1302	43	64	8.0	0.75	7	13	142
Prunes	1125	49	35	3.8	0.20	16	3	108	1103	49	35	3.8	0.20	16	4	106
Raisins	1096	77	53	2.0	0.30	66	11	42	1175	53	40	4.4	0.45	69	10	88
Sultanas	1047	64	43	2.2	0.40	64	19	116	1250	75	37	2.6	0.47	22	19	101
Nuts																
Almonds	898	259	270	4.4	0.10	6	2	464	814	251	282	3.1	1.04	15	19	574
Barcelona	991	180	214	3.1	1.00	3	36	317	997	180	212	3.2	1.02	3	36	318
Brazil	831	192	449	3.1	1.20	2	67	647	679	175	422	2.6	1.81	3	59	607
Chestnut	1029	95	69	1.8	0.50	23	31	153	1035	95	68	1.9	0.48	23	31	153
Hazelnut	580	74	94	1.8	0.40	2	10	385	765	147	168	3.4	1.29	6	19	314
Coconut (solid)	751	23	90	3.6	0.50	28	196	162	673	24	75	3.8	0.58	31	200	171
Peanut	712	64	189	2.1	0.30	6	7	382	715	64	224	2.7	1.09	2	7	459
Walnut	898	79	172	3.1	0.40	3	30	667	463	97	165	3.0	1.38	7	25	391

[1]All mineral concentrations are quoted in mg 100 g^{-1} DW.

TABLE II

Mineral concentrations of US vegetables, fruits and nuts listed by Beeson (1941) and those available from the USDA[1]

	Beeson (1941)						USDA (2004)					
	K	Ca	Mg	Fe	Cu	P	K	Ca	Mg	Fe	Cu	P
Vegetables												
Potato	2280[2]	49	130	10.5	0.80	250	2137	55	112	3.6	0.58	295
Butter (Lima) bean	1890	104	200	11.2	0.90	412	1569	114	195	10.6	1.07	457
Haricot (Navy) bean	1410	170	190	13.9	1.10	630	1472	72	484	9.3	1.71	480
Lentil	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Pea	1410	190	180	12.6	0.90	570	1154	118	156	7.0	0.83	511
Aubergine (Eggplant)	NA	NA	NA	14.8	1.30	NA	3030	119	184	3.2	1.08	329
Cabbage	2710	730	160	9.5	1.40	380	2843	513	218	6.9	0.37	348
Carrot	2100	400	170	17.9	1.00	330	2733	282	102	2.6	0.38	299
Celery	3920	2360	390	15.9	16.30	640	5689	875	241	4.4	0.77	525
Chicory, witloof	NA	NA	NA	NA	NA	NA	3850	347	182	4.4	0.93	474
Cucumber	4480	740	NA	27.0	2.40	750	3082	335	273	5.9	0.86	503
Lettuce	5980	770	240	103.4	1.90	560	3368	433	184	4.6	0.58	396
Mushroom	NA	NA	NA	10.9	6.20	NA	4164	40	119	6.9	4.23	1127
Mustard	NA	2130	50	49.7	NA	710	3848	1120	348	15.9	1.60	467
Onion	1520	390	130	13.0	1.20	260	1257	192	87	1.7	0.33	236
Parsnip	1850	280	120	11.4	0.70	370	1832	176	142	2.9	0.59	347
Pumpkin	2470	500	260	NA	0.40	240	4048	250	143	9.5	1.51	524
Radish	NA	NA	NA	43.6	2.90	NA	4926	529	211	7.2	1.06	423
Spring onion	NA	NA	NA	NA	NA	NA	2714	708	197	14.6	0.82	364
Swede (Rutabaga)	1910	490	140	27.5	0.80	270	3259	455	222	5.0	0.39	561
Tomato	4800	240	300	20.2	1.40	550	4309	182	200	4.9	1.07	436
Turnip	2770	510	240	9.2	0.90	360	2349	369	135	3.7	1.05	332
Watercress	NA	NA	NA	92.9	1.20	NA	6748	2454	429	4.1	1.57	1227
Endive	NA	NA	NA	70.0	NA	NA	5056	837	242	13.4	1.59	451
Fruits												
Apples	740	77	NA	1.5	0.60	71	675	38	30	0.5	0.23	83
Apricots	NA	NA	NA	4.3	NA	NA	1897	95	73	2.9	0.57	168
Avocado pear	NA	NA	NA	26.4	NA	NA	1812	45	108	2.1	0.71	194
Bananas	NA	NA	NA	2.8	0.90	NA	1427	20	108	1.0	0.31	88
Blackberries	NA	NA	NA	6.3	1.00	NA	1367	245	169	5.2	1.39	186
Cherries, eating	NA	NA	NA	6.9	1.40	NA	1251	73	62	2.0	0.34	118
Cherries, cooking	NA	NA	NA	NA	NA	NA	1247	115	65	2.3	0.75	108
Cranberries	NA	NA	NA	1.9	0.80	NA	660	62	47	1.9	0.47	101
Currants, black	NA	NA	NA	NA	1.70	NA	1785	305	133	8.5	0.48	327
Currants, red	NA	NA	NA	NA	NA	NA	1713	206	81	6.2	0.67	274
Currants, white	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Custard Apple	NA	NA	NA	NA	NA	NA	1340	105	63	2.5	NA	74
Figs	NA	NA	NA	3.7	0.60	NA	1111	168	81	1.8	0.34	67
Gooseberries	NA	NA	NA	4.7	0.80	NA	1632	206	82	2.6	0.58	223
Grapes	NA	NA	NA	6.7	0.80	NA	1001	63	31	1.7	0.44	79
Grapefruit	NA	NA	NA	12.4	0.50	NA	1526	132	88	1.0	0.52	88
Lemons, whole	1310	580	110	NA	0.30	190	1151	484	95	0.6	2.06	119
Melons, cantaloupe	NA	NA	NA	NA	NA	NA	2711	91	122	2.1	0.42	152
Mulberries	NA	NA	NA	28.0	0.40	NA	1575	317	146	15.0	0.49	308
Nectarines	NA	NA	NA	NA	NA	NA	1620	48	73	2.3	0.69	210
Oranges	NA	NA	NA	4.4	1.00	NA	1366	302	75	0.8	0.34	106
Passionfruit (Granadilla)	NA	NA	NA	NA	NA	NA	1286	44	107	5.9	0.32	251
Peaches	NA	NA	NA	14.0	NA	NA	1707	54	81	2.2	0.61	180
Pears	NA	NA	NA	8.4	1.00	NA	731	55	43	1.0	0.50	68
Pineapple	NA	NA	NA	14.4	0.80	NA	849	96	89	2.1	0.73	59
Plums	NA	NA	NA	11.0	1.00	NA	1229	47	55	1.3	0.45	125
Pomegranate	NA	NA	NA	NA	NA	NA	1361	16	16	1.6	0.37	42
Quinces	NA	NA	NA	5.8	0.80	NA	1216	68	49	4.3	0.80	105
Raspberries	NA	NA	NA	6.2	0.80	NA	1060	175	154	4.8	0.63	204
Rhubarb	NA	NA	NA	35.6	0.90	NA	4507	1346	188	3.4	0.33	219
Strawberries	2050	310	NA	13.5	0.50	200	1691	177	144	4.6	0.53	265
Tangerines	NA	NA	NA	4.4	0.60	NA	1266	113	97	0.8	0.23	81
Watermelon	NA	NA	NA	10.9	0.90	NA	1310	82	117	2.8	0.49	129
Dried fruits												
Apricots	NA	NA	NA	NA	NA	NA	1681	80	46	3.8	0.50	103
Currants	NA	NA	NA	NA	NA	NA	1104	106	51	4.0	0.58	155
Dates	NA	NA	NA	4.9	0.30	NA	855	65	61	1.2	0.36	78
Figs	NA	NA	NA	NA	NA	NA	972	232	97	2.9	0.41	96
Peaches	NA	NA	NA	NA	NA	NA	1460	41	62	6.0	0.53	174
Prunes	NA	NA	NA	5.5	0.40	NA	1060	62	59	1.3	0.41	100
Raisins	NA	NA	NA	NA	NA	NA	937	46	37	2.7	0.37	105
Sultanas	NA	NA	NA	NA	NA	NA	877	62	41	2.1	0.43	135
Nuts												
Almonds	NA	NA	NA	4.2	1.20	NA	768	262	290	4.5	1.17	500
Barcelona	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Brazil	NA	NA	NA	4.2	1.50	NA	683	166	390	2.5	1.81	751
Chestnut	NA	NA	NA	6.3	0.70	NA	1089	70	81	2.6	0.72	151
Hazelnut	NA	NA	NA	NA	NA	NA	718	120	172	5.0	1.82	306
Coconut (solid)	NA	NA	NA	NA	1.10	NA	672	26	60	4.6	0.82	213
Peanut	NA	NA	NA	2.4	1.00	NA	754	98	180	4.9	1.22	402
Walnut	NA	NA	NA	3.7	NA	NA	460	102	165	3.0	1.65	361

[1]USDA National Nutrient Database for Standard Reference, Release 16, May 2004.
[2]All mineral concentrations are quoted in mg 100 g^{-1} DW.

TABLE III
Quotients of mineral concentrations in the dry matter of vegetables, fruits and nuts available in the UK (1980s/1930s) and in the USA (2004/1930s) calculated from the data presented in Tables I and II

	UK quotient 1980s/1930s								US quotient 2004/1930s					
	K	Ca	Mg	Fe	Cu	Na	Cl	P	K	Ca	Mg	Fe	Cu	P
Vegetables														
Potato	0.74	0.74	0.81	0.49	0.63	1.23	0.97	1.06	0.94	1.13	0.86	0.35	0.72	1.18
Butter bean	1.03	1.03	1.19	1.03	0.64	0.68	1.04	1.04	0.83	1.10	0.97	0.94	1.19	1.11
Haricot bean	1.04	1.04	1.02	1.05	0.98	1.03	1.13	1.04	1.04	0.42	2.55	0.67	1.55	0.76
Lentil	1.29	1.67	1.32	1.30	1.50	0.69	1.25	1.45						
Runner bean	0.77	0.96	0.80	1.57	0.21	0.01	0.89	1.27						
Pea	0.85	1.18	0.88	1.34	0.22	0.98	0.89	1.06	0.82	0.62	0.87	0.55	0.93	0.90
Aubergine (Eggplant)	0.82	0.90	1.07	0.73	0.11	0.74	0.21	1.23				0.21	0.83	
Cabbage	0.92	1.25	0.60	0.78	0.12	0.28	1.12	1.28	1.05	0.70	1.36	0.73	0.26	0.91
Carrot	0.79	0.54	0.26	0.55	0.25	0.27	0.50	0.74	1.30	0.70	0.60	0.14	0.38	0.91
Celery	1.53	1.05	0.78	1.26	0.13	0.58	0.95	0.88	1.45	0.37	0.62	0.28	0.05	0.82
Chicory	0.62	0.76	0.32	0.39	0.24	0.09	1.21	0.86						
Cucumber	0.99	0.79	0.88	0.99	0.11	0.23	0.69	2.03	0.69	0.45		0.22	0.36	0.67
Lettuce	1.04	1.07	0.61	0.94	0.07	0.97	1.17	0.91	0.56	0.56	0.77	0.04	0.31	0.71
Mushroom	0.78	2.38	0.78	0.67	1.28	0.63	0.93	0.67				0.63	0.68	
Mustard and cress	0.52	1.21	1.29	0.34	0.14	1.59	0.70	0.80		0.53	6.96	0.32		0.66
Onion	0.77	0.53	0.35	0.67	0.41	0.19	0.81	0.66	0.83	0.49	0.67	0.13	0.28	0.91
Parsnip	1.89	0.63	0.87	0.88	0.48	0.51	1.02	0.91	0.99	0.63	1.18	0.25	0.84	0.94
Pumpkin	0.45	0.79	1.30	1.11	0.29		1.07	1.04	1.64	0.50	0.55		3.78	2.18
Radish	1.45	0.63	0.64	0.47	0.11	0.27	2.87	1.07				0.16	0.36	
Spring onion	1.95	0.49	1.85	2.59	0.77	0.92	1.48	2.08						
Swede (Rutabaga)	1.23	0.92	0.82	0.36	0.28	0.28	1.00	2.07	1.71	0.93	1.59	0.18	0.48	2.08
Tomato	0.84	0.51	0.61	1.11	0.09	3.11	1.04	1.08	0.90	0.76	0.67	0.24	0.77	0.79
Turnip	0.89	0.62	0.83	0.45	0.19	0.20	0.42	1.13	0.85	0.72	0.56	0.40	1.16	0.92
Watercress	0.87	0.91	1.05	1.61	0.09	0.97	1.29	1.19				0.04	1.31	
Endive	1.00	1.00	0.96	1.01	0.11	0.99	1.00	1.01				0.19		
Horseradish	1.00	1.01	1.01	0.99	1.52	1.02	1.01	1.00						
Fruits														
Apples, eating	1.03	1.12	1.08	0.34	0.18	1.29	0.08	1.45	0.91	0.49		0.35	0.39	1.16
Apples, cooking	0.84	1.30	1.22	0.41	0.23	1.08	0.52	0.51						
Apricots	0.88	0.91	0.93	1.40	0.52		0.98					0.66		
Avocado pear	0.77	0.49	0.58	0.52	0.63	0.25	0.68	0.86				0.08		
Bananas	1.35	1.05	0.96	0.86	0.67	1.00	1.17	1.17				0.37	0.35	
Blackberries	0.93	0.78	0.94	0.99	1.22	0.67	1.20	1.57				0.83	1.39	
Cherries, eating	0.82	0.88	1.12	0.58	1.02	0.39	1.34					0.29	0.24	
Cranberries	0.79	0.81	0.83	0.63	0.35	1.10	0.97					1.02	0.59	
Currants, black	0.99	0.99	0.99	1.03	1.03	1.11	1.02	1.00					0.28	
Currants, red	1.02	1.01	1.02	0.98	1.00	0.89	1.00	1.02						
Currants, white	1.00	0.98	1.02	0.98	1.05	1.33	1.03	1.00						
Custard Apple	0.50	1.74	1.41	1.12	0.93	0.50	0.68							
Damsons	1.00	1.03	1.00	0.99	0.89	0.74	0.97							
Figs	0.75	1.11	0.75	0.72	0.97	1.95	0.98	0.47				0.48	0.56	
Gooseberries, unripe	1.02	1.01	1.01	0.95	0.47	1.06	1.09	1.02				0.54	0.72	
Gooseberries, ripe	1.65	1.70	1.72	1.73	0.67	1.44	1.68	1.64						
Grapes	0.82	1.23	1.48	0.97	1.32	1.37	0.11	1.04				0.25	0.55	
Grapefruit	0.72	1.14	0.74	0.32	0.26	1.95	1.95	1.09				0.08	1.03	
Greengages	1.23	1.23	1.27	1.31	1.11	0.93	1.23							
Lemons, whole	1.00	0.86	1.12	1.52	1.05	0.91	1.07	0.94	0.88	0.83	0.87		6.88	0.63
Loganberries	1.01	1.00	1.00	1.03	1.04	1.25	1.02	0.99						
Medlars	1.02	1.00	1.05	1.03	0.95	0.98	0.98	1.00						
Melons, cantaloupe	0.54	0.84	0.44	0.30	0.02	0.48	0.81	0.34						
Mulberries	1.01	1.01	0.99	1.02	1.00	0.95	1.11	1.00				0.54	1.22	
Nectarines	1.13	3.15	1.41	1.57	1.80	0.20	1.88	1.64						
Oranges	0.77	1.14	0.78	0.31	0.72	1.71	0.94	0.89				0.17	0.34	
Passion Fruit (Granadilla)	0.61	0.76	0.80	1.23	0.71	1.26								
Peaches	0.77	1.80	1.42	1.29	1.80	0.45	1.48					0.16		
Pears	1.31	1.58	1.17	1.03	0.46	1.32	2.06	1.20				0.12	0.50	
Pineapple	0.76	1.73	1.11	0.55	1.63	1.48	1.19	1.48				0.14	0.91	
Plums, Victoria Dessert	1.15	1.14	1.11	1.30	1.19	1.30	1.11					0.12	0.45	
Plums, cooking	1.14	0.87	0.94	1.24	1.04	0.96	1.46							
Quinces	0.99	1.01	1.00	0.95	1.03	0.95	1.05	1.00				0.74	1.00	
Raspberries	0.98	0.79	1.13	0.75	0.59	1.54	1.28	1.39				0.78	0.79	
Rhubarb	0.69	0.91	0.97	0.77	0.55	1.40	1.01	0.82				0.10	0.37	
Strawberries	1.06	0.77	0.91	0.60	0.61	4.08	1.09	1.11	0.82	0.57		0.34	1.06	1.33
Tangerines	1.02	1.00	0.97	1.13	0.11	0.88	0.84	1.01				0.18	0.38	
Watermelon	0.36	0.38	0.45	0.93	0.65	0.08	0.77				0.26	0.55		
Dried fruits														
Apricots	1.00	1.00	1.00	1.00	1.56	0.99	1.03	1.02						
Currants	0.94	0.90	0.77	0.67	1.60	0.66	0.95	1.62						
Dates	0.93	0.67	0.71	0.80	1.52	1.95	1.28	0.94				0.25	1.20	
Figs	0.96	0.88	0.86	1.02	1.19	0.72	1.03	0.97						
Peaches	1.00	1.01	1.00	1.01	1.07	1.01	1.08	1.04						
Prunes	0.98	1.01	1.00	0.99	1.02	1.00	1.45	0.98				0.24	1.02	
Raisins	1.07	0.69	0.76	2.19	1.50	1.05	0.94	2.08						
Sultanas	1.19	1.18	0.85	1.18	1.18	0.35	0.99	0.87						
Nuts														
Almonds	0.91	0.97	1.04	0.71	10.44	2.44	9.39	1.24				1.08	0.98	
Barcelona	1.01	1.00	0.99	1.03	1.02	1.06	1.00	1.00						
Brazil	0.82	0.91	0.94	0.83	1.51	1.54	0.88	0.94				0.60	1.20	
Chestnut	1.01	1.00	0.99	1.04	0.95	0.99	1.00	1.00				0.42	1.03	
Hazelnut	1.32	1.98	1.78	1.86	3.22	3.14	1.89	0.82						
Coconut (solid)	0.90	1.03	0.83	1.06	1.16	1.10	1.02	1.05					0.75	
Peanut	1.00	1.00	1.19	1.27	3.63	0.36	1.07	1.20				2.04	1.22	
Walnut	0.52	1.22	0.96	0.96	3.45	2.40	0.82	0.59				0.82		

TABLE IV

Statistical analysis to test if the concentrations of Ca, Cl, Cu, Fe, K, Mg, Na and P have declined in UK produce between the 1930s and the 1980s

	Mineral	n	Mean quotient (1980s/1930s)	Variance	S.D.[1]	S.E.[2]	Lower 95% C.I.[3]	Upper 95% C.I.	Null hypothesis: "$\log_e$ of 1980s/1930s quotient equals zero" Test statistic (t)	d.f.[4]	*P*
ALL SAMPLES	Ca	80	–0.022	0.121	0.348	0.039	–0.100	0.055	–0.58	79	0.567
	Cl	69	–0.013	0.365	0.604	0.073	–0.158	0.132	–0.18	68	0.856
	Cu	79	–0.516	1.171	1.082	0.122	–0.759	–0.274	–4.24	78	< 0.001 declined
	Fe	80	–0.125	0.209	0.457	0.051	–0.227	–0.024	–2.45	79	0.016 declined
	K	80	–0.078	0.084	0.290	0.032	–0.143	–0.014	–2.42	79	0.018 declined
	Mg	80	–0.084	0.117	0.342	0.038	–0.161	–0.008	–2.21	79	0.030 declined
	Na	78	–0.253	0.775	0.881	0.100	–0.451	–0.054	–2.54	77	0.013 declined
	P	80	0.047	0.096	0.310	0.035	–0.022	0.116	1.36	79	0.177
VEGETABLES[a]	Ca	26	–0.126	0.138	0.371	0.073	–0.276	0.024	–1.72	25	0.097
	Cl	26	–0.067	0.213	0.461	0.090	–0.253	0.120	–0.74	25	0.469
	Cu	26	–1.319	0.904	0.951	0.187	–1.703	–0.935	–7.07	25	< 0.00 declined
	Fe	26	–0.175	0.260	0.510	0.100	–0.381	0.031	–1.75	25	0.092
	K	26	–0.056	0.119	0.345	0.068	–0.196	0.083	–0.83	25	0.415
	Mg	26	–0.212	0.199	0.447	0.088	–0.392	–0.031	–2.42	25	0.023 declined
	Na	25	–0.701	1.171	1.082	0.217	–1.148	–0.254	–3.24	24	0.004 declined
	P	26	0.081	0.091	0.302	0.059	–0.041	0.203	1.37	25	0.182
FRUITS[a]	Ca	38	0.040	0.130	0.360	0.058	–0.079	0.158	0.68	37	0.503
	Cl	27	–0.091	0.517	0.719	0.138	–0.376	0.194	–0.66	26	0.516
	Cu	37	–0.411	0.755	0.869	0.143	–0.701	–0.122	–2.88	36	0.007 declined
	Fe	38	–0.164	0.216	0.465	0.075	–0.317	–0.011	–2.17	37	0.036 declined
	K	38	–0.107	0.082	0.286	0.046	–0.201	–0.013	–2.31	37	0.026 declined
	Mg	38	–0.014	0.080	0.283	0.046	–0.107	0.079	–0.31	37	0.760
	Na	37	–0.097	0.499	0.706	0.116	–0.332	0.139	–0.83	36	0.410
	P	38	0.026	0.111	0.333	0.054	–0.083	0.136	0.48	37	0.633
DRY FRUITS[a]	Ca	8	–0.103	0.039	0.197	0.070	–0.268	0.063	–1.47	7	0.185
	Cl	8	0.080	0.023	0.152	0.054	–0.047	0.207	1.48	7	0.181
	Cu	8	0.270	0.034	0.185	0.065	0.115	0.425	4.13	7	0.004 increased
	Fe	8	0.043	0.119	0.346	0.122	–0.246	0.332	0.35	7	0.737
	K	8	0.005	0.007	0.083	0.029	–0.064	0.074	0.18	7	0.865
	Mg	8	–0.149	0.019	0.138	0.049	–0.264	–0.034	–3.06	7	0.018 declined
	Na	8	–0.134	0.241	0.491	0.174	–0.544	0.277	–0.77	7	0.466
	P	8	0.128	0.095	0.308	0.109	–0.129	0.386	1.18	7	0.277
NUTS[a]	Ca	8	0.099	0.063	0.251	0.089	–0.111	0.309	1.11	7	0.302
	Cl	8	0.330	0.659	0.812	0.287	–0.349	1.008	1.15	7	0.289
	Cu	8	0.822	0.694	0.833	0.295	0.126	1.518	2.79	7	0.027 increased
	Fe	8	0.052	0.083	0.288	0.102	–0.189	0.293	0.51	7	0.625
	K	8	–0.097	0.072	0.268	0.095	–0.321	0.127	–1.03	7	0.338
	Mg	8	0.060	0.054	0.233	0.082	–0.135	0.254	0.73	7	0.491
	Na	8	0.308	0.484	0.695	0.246	–0.274	0.889	1.25	7	0.251
	P	8	–0.043	0.057	0.238	0.084	–0.242	0.156	–0.51	7	0.624

[a]Pooled values for various appropriate crops listed in Tables I–III.
[1]S.D. = standard deviation.
[2]S.E. = standard error of mean.
[3]C.I. = confidence interval.
[4]d.f. = degrees of freedom.

cell defence mechanisms, red and white blood cell maturation, Fe transport, cholesterol and glucose metabolism, myocardial contractility, bone strength and brain development (Food Standards Agency, 2003). The Food Standards Agency Expert Group on Vitamins and Minerals concluded that Cu deficiency would be rare in the UK (Food Standards Agency, 2003), although it may occur in individuals with a genetic defect, such as Menke's syndrome. Nevertheless, there is considerable genetic variation in the ability of plant species (Broadley *et al*., 2001) and cultivars to accumulate Cu. For example, Cu concentrations in spinach (Römer and Keller, 2002) and onion (Alvarez *et al*., 2003) shoots differed by 50% between cultivars; and tubers of different potato cultivars varied over four-fold when grown under comparable conditions (Rivero *et al*., 2003). Copper concentrations in apples (Iwane, 1991) and strawberries (Hakala *et al*., 2003) differed by two-fold between cultivars, and five cranberry fruit cultivars varied up to 16-fold in their Cu concentrations (Davenport and Provost, 1994). Thus, there may be potential for genetic improvement in the Cu composition of crops. Furthermore, Cu concentrations in vegetables and fruits can be increased by fertilisation (Smilde *et al*., 1981; Sterrett *et al*., 1983; Karam *et al*., 1998; Shuman, 1998; Tüzen *et al*., 1998; Rengel *et al*., 1999; Bunzl *et al*., 2001; Tamoutsidis *et al*., 2002; Wen *et al*., 2002; Bolan *et al*., 2003). However, the combination of crop variety and Cu fertilisation must be managed appropriately to ensure a balance between adequate and excessive Cu nutrition.

Animals also require Fe to maintain the activities of many important enzymes, and for vital haem proteins such as haemoglobin, myoglobin and the cytochromes which are involved in oxygen transport and energy metabolism, respectively. Like Cu, Fe is under tight homeostatic control, although unlike Cu, Fe deficiency is common in both industrialised and developing countries (Frossard *et al*., 2000; Welch and Graham, 2004). The estimated Fe requirement for the UK population is 6 – 12 mg d^{-1} (Food Standards Agency, 2003). In the UK, some groups may not receive sufficient amounts of Fe in their diets, in particular bioavailable haem-Fe from animal sources, which is more easily absorbed by the gut

TABLE V

Statistical analysis to test if the concentrations of Ca, Cl, Cu, Fe, K, Mg, Na and P have declined in US produce between the 1930s and 2004

									Null hypothesis: "$\log_e$ of 2004/1930s quotient equals zero"		
	Mineral	n	Mean quotient (2004/1930s)	Variance	S.D.[1]	S.E.[2]	Lower 95% C.I.[3]	Upper 95% C.I.	Test statistic (t)	d.f.[4]	*P*
ALL SAMPLES	Ca	19	–0.465	0.097	0.311	0.071	–0.615	–0.315	–6.52	18	< 0.001 declined
	Cu	48	–0.396	0.599	0.774	0.112	–0.621	–0.172	–3.55	47	< 0.001 declined
	Fe	50	–1.202	0.702	0.838	0.119	–1.440	–0.964	–10.14	49	< 0.001 declined
	K	18	–0.029	0.083	0.288	0.068	–0.173	0.114	–0.43	17	0.670
	Mg	16	0.008	0.441	0.664	0.166	–0.346	0.361	0.05	15	0.964
	P	19	–0.035	0.120	0.346	0.079	–0.201	0.132	–0.44	18	0.666
VEGETABLES	Ca	16	–0.461	0.106	0.325	0.081	–0.634	–0.287	–5.67	15	< 0.001 declined
	Cu	19	–0.507	0.850	0.922	0.212	–0.951	–0.063	–2.40	18	0.028 declined
	Fe	20	–1.375	0.679	0.824	0.184	–1.761	–0.989	–7.46	19	< 0.001 declined
	K	15	–0.008	0.097	0.312	0.081	–0.181	0.165	–0.10	14	0.925
	Mg	15	0.018	0.470	0.686	0.177	–0.362	0.398	0.10	14	0.922
	P	16	–0.039	0.122	0.349	0.087	–0.225	0.147	–0.45	15	0.661
FRUITS	Ca	3	–0.487	0.077	0.277	0.160	–1.175	0.201	–3.05	2	0.093
	Cu	22	–0.440	0.526	0.725	0.155	–0.762	–0.118	–2.84	21	0.010 declined
	Fe	23	–1.261	0.629	0.793	0.165	–1.603	–0.918	–7.62	22	< 0.001 declined
	K	3	–0.138	0.003	0.051	0.029	–0.265	–0.011	–4.68	2	0.043 declined
	P	3	–0.012	0.160	0.400	0.231	–1.006	0.983	–0.05	2	0.964
DRY FRUITS	Fe	2	–1.401	0.000	0.009	0.006	–1.478	–1.324	–231.05	1	0.003 declined
	Cu	2	0.098	0.013	0.115	0.082	–0.939	1.135	1.21	1	0.441
NUTS	Fe	5	–0.159	0.364	0.603	0.270	–0.908	0.590	–0.59	4	0.588
	Cu	5	0.019	0.040	0.200	0.090	–0.230	0.268	0.21	4	0.842

[a]Pooled values for various appropriate crops listed in Tables II.
[1]S.D. = standard deviation.
[2]S.E. = standard error of mean.
[3]C.I. = confidence interval.
[4]d.f. = degrees of freedom.

than the tightly-bound non-haem Fe which occurs in plant-derived products. For example, the 2003 UK National Diet and Nutrition Survey estimated that 25% of women had inadequate Fe consumption, rising to > 40% in the 18–34 year-old age range (Henderson *et al.*, 2003; Marriott and Buttriss, 2003). Notably, total Fe intake does not equate to the Fe status of an individual and Fe-homeostasis is critically dependent on whether an individual is susceptible to blood loss or has a diet low in haem-Fe or vitamin C (Food Standards Agency, 2003). The main sources of dietary Fe in the UK are cereals and cereal products (44%), while meat and meat products (17%) and vegetables excluding potatoes (10%) are also significant sources. Fruits and nuts are not significant sources of Fe intake, contributing only 3% of the Fe, and 7% of the K to the average UK diet (Henderson *et al.*, 2003). Thus, any decline in Fe in fruits or nuts will have little impact on the UK diet.

To address potential Fe-deficiencies in the UK, there is mandatory fortification of white and brown flour, and many breakfast cereals are fortified with Fe on a voluntary basis (Food Standards Agency, 2003). However, there is considerable genetic variation in the ability of plants to accumulate Fe, and international endeavours to breed staple crops with increased Fe content are being adopted to address Fe-deficiency in the Developing World (Frossard *et al.*, 2000; Welch and Graham, 2004). For example, a 2.6-fold variation in Fe concentrations in field grown *Phaseolus* genotypes has been the target for a breeding programme at the Centro Internacional de Agricultura Tropical (Welch and Graham, 2004). Iron concentrations in apples (Iwane, 1991) and strawberries (Hakala *et al.*, 2003) differ by only 20–60% between cultivars; however, an eight-fold variation in Fe concentrations has been observed across 11 plum varieties (Nergiz and Yildiz, 1997), and the five cranberry fruit cultivars quoted above differed by almost 20-fold in their Fe concentrations (Davenport and Provost, 1994). Iron concentrations in vegetables and fruits can also be increased by appropriate fertilisation (Shuman, 1998; Rengel *et al.*, 1999; Frossard *et al.*, 2000).

In conclusion, the average concentrations of some minerals within a range of horticultural products, in general, has decreased since the 1930s in the UK. Parallel changes can be seen in the USA. However, this phenomenon is unlikely to affect UK diets since the proportion of these minerals derived from horticultural products is generally low (Henderson *et al.*, 2003). If necessary, micro-nutrient fertilisation strategies could be adopted to alter crop mineral composition. However, there is considerable genetic variation between horticultural crop genotypes, and crop selection or breeding could also be used to increase dietary mineral intakes in certain sections of the population in the future.

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