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Compilation of a provisional UK database for the phylloquinone (vitamin K_1) content of foods†

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This paper reports the compilation of a food composition database for phylloquinone (vitamin K_1) derived from the direct analysis of foods, recipe calculation and the assignment of values based on food similarities. All the basic and other food items used in these calculations had been analysed by HPLC and about 170 of the items had been obtained and assayed in the UK. Recipe calculations took account of the cooking method and changes in water and fat content. Currently, approximately 1501 food items with Royal Society of Chemistry/Ministry of Agriculture, Fisheries and Food food codes have been allocated a vitamin K_1 value, and a further 282 new recipe codes are included in the database. Representative values from each food group are reported together with an indication of the potential variation. Detailed examples of some recipe calculations are included, and also the impact of changing the type of fat in recipes. Vitamin K_1 is associated with, and most abundant in, photosynthetic tissues of plants. Accordingly, the highest concentrations (3000–6000 μg/kg) are found in dark-green leafy vegetables and herbs, such as kale, parsley, spinach and green cabbage. Intermediate concentrations (1000–2000 μg/kg) are found in plants with paler leaves such as white cabbage and lettuce or in green, non-leafy vegetables such as broccoli and brussel sprouts. Fats and oils contain variable amounts of vitamin K_1 with the highest concentrations (300–1300 μg/kg) in soyabean, rapeseed and olive oils and the margarines based on them. Other foods such as dairy products, meat dishes and cereal-based foods (bread, biscuits, cakes, desserts etc.), although not in themselves particularly rich in vitamin K_1 (< 200 μg/kg), may contribute significantly to intakes when consumption of green vegetables is poor. Within the scope of this present study, it has not been possible to address issues such as inter-sample variability, losses during storage or the bioavailability from different foods and further work on these aspects is needed.

Vitamin K: Food composition: Nutrient database: Phylloquinone

Vitamin K occurs in nature as a series of molecular forms that have a common 2-methyl-1,4-naphthoquinone ring, but differ in the length and degree of saturation of their isoprenoid side chain at the 3-position. They are traditionally classified into two groups according to whether they are synthesized by plants or bacteria. In plants the only major form is phylloquinone (vitamin K_1 (K_1)) with a phytyl side-chain, whereas bacteria synthesize a family of menaquinones (vitamins K_2) with fully or partially unsaturated prenyl side-chains. Evidence has been obtained recently that menaquinone-4 may be formed in animal tissues by the dealkylation and prenylation of K_1 (Thijssen & Drittij-Reijnders, 1994).

Members of both series are able to function as a cofactor for the post-translational conversion of protein-bound glutamate residues to γ-carboxyglutamate (Gla) residues in a diverse group of proteins synthesized in a wide variety of tissues (Vermeer, 1990; Ferland, 1998; Newman & Shearer, 1998). The resultant Gla proteins include proteins that function in haemostasis as procoagulants (factors II, VII,

Abbreviations: Gla, γ-carboxyglutamate; K_1, vitamin K_1; K_1 (I-H_2), 2′, 3′ dihydro-phylloquinone; MK, menaquinone; RSC/MAFF, Royal Society of Chemistry/Ministry of Agriculture, Fisheries and Food.

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† The full database with the Royal Society of Chemistry/Ministry of Agriculture, Fisheries and Food food codes is available as hard copy or on floppy disc. Please send a 9 cm disc to Dr Bolton-Smith.

IX and X), or anticoagulants (proteins C and S), and proteins whose function is presently unclear (osteocalcin and matrix Gla protein). While the synthesis of some of these proteins is unique to one tissue or organ (e.g. the liver for factors II, VII IX, X, protein C and the bone for osteocalcin), others such as protein S (liver, endothelium and bone) and matrix Gla protein (most tissues) have a more widespread distribution. From measurements of urinary Gla excretion after dietary restriction, Ferland *et al*. (1993) predicted that there are other vitamin K-dependent proteins yet to be isolated and characterized. One such recently discovered Gla protein is that encoded by a growth-arrest-specific gene, gas6 (Manfioletti *et al*. 1993). The presence of a number of Gla residues confers the vitamin K-dependent proteins with Ca binding properties. In the coagulation proteins the Gla residues facilitate the Ca-mediated binding to phospholipids that are essential for their biological activity, while in osteocalcin they are responsible for its ability to bind tightly to the Ca ions of the hydroxyapatite lattice.

For many years it was held to be axiomatic that overt deficiency states of vitamin K caused by dietary lack are rare and this is certainly true for its coagulation function where the only population group at risk from bleeding are infants in the first 6 months or so of life (Shearer, 1995). In recent years, however, there has been a growing body of evidence that vitamin K may have an important role in bone health and that there may be a specific link between suboptimal vitamin K status and osteoporosis (for review see Binkley & Suttie, 1995; Kohlmeier *et al*. 1996; Shearer *et al*. 1996; Vermeer *et al*. 1996; Shearer, 1997). It is generally assumed that any regulatory role of vitamin K on the skeleton is mediated through osteocalcin or matrix-Gla protein but this may not necessarily be the case as shown by recent work that suggests that menaquinone-4 or even its side-chain may inhibit bone resorption by other mechanisms (Hara *et al*. 1995; Kameda *et al*. 1996). The possible link between vitamin K and osteoporosis highlights the possibility that sub-optimal vitamin K nutrition, and even outright deficiency, may occur in the general population (Sokoll *et al*. 1997) and/or ‘at risk’ sub-groups such as post-menopausal women and the elderly (Booth *et al*. 1995*b*). In the UK, at present, guideline intakes of 1 μg/kg per d for adults and 2 μg/kg per d for infants are suggested, but no dietary reference values have been set (Bolton-Smith & Shearer, 1997) due to the lack of information on intakes and markers of vitamin K status (Department of Health, 1991).

Some food composition data for K_1 have been reported from the USA (Booth *et al*. 1993, 1995*a*), from Austria (Jakob & Elmadfa, 1996) and from Finland (Koivu *et al*. 1997, 1998; Piironen *et al*. 1997). Germany and Denmark have already incorporated K_1 values into their national food tables (Deharveng *et al*. 1999). For the UK, Shearer and colleagues have previously reported the K_1 composition of milks (Haroon *et al*. 1982) and some selected food items (Shearer *et al*. 1996).

If research on the role of vitamin K nutrition in health and disease is to be carried forward in the UK, a reliable food composition database is required (ideally for phylloquinone and menaquinones). This present paper reports the first database for the K_1 content of foods that are specific to the UK situation. Preliminary reports of K_1 intakes and food sources in Scotland have been presented using the present database (Price *et al*. 1996; Fenton *et al*. 1997; Bolton-Smith *et al*. 1998, 1999).

Methods

Direct analysis of phylloquinone in foods

Approximately 100 basic food items had been analysed previously in the laboratory of M. J. S. (Shearer *et al*. 1996) and a further seventy items were analysed specifically for the compilation of the current database. These included re-analysis of items whose oil composition we suspected to have changed (particularly margarines).

The extraction of K_1 from foods and the chromatographic clean-up procedures using sequential solid phase extraction (Sep-Pak silica cartridges, Waters plc, Milford, MA, USA) and semi-preparative adsorption HPLC were carried out as previously described (Shearer, 1986). The extraction solvent chosen for most food items was acetone. The final separation, detection and quantification of K_1 was carried out by reversed-phase HPLC, originally using u.v. detection (Shearer *et al*. 1980; Shearer, 1986) and latterly using electrochemical detection in the redox mode (Shearer, 1991; McCarthy *et al*. 1997). In general, foods high in K_1 such as green vegetables were analysed using u.v. detection. The development of electrochemical detection methods gave a more sensitive and selective assay of K_1 in foods with low K_1 concentrations and/or complex matrices (e.g. fish and meat produce etc.) that was not previously possible with u.v. detection. All values for vegetables and most fruits were obtained using u.v. detection with either phylloquinone-2,3-epoxide or 2-chloro-phylloquinone as internal standards (Shearer *et al*. 1980; Shearer, 1986). Values for fish, meat and poultry, snack foods and beverages were obtained using electrochemical detection, normally with menaquinone-6 as an internal standard (Shearer, 1991; McCarthy *et al*. 1997). For some analyses using electrochemical detection, radio-labelled [1′, 2′-$^3H_2$]phylloquinone or [1′, 2′-$^3H_2$]phylloquinone-2,3-epoxide were employed as internal standards. Several identical food items (but different samples) of cereals, oils, fats, margarines and dairy produce were analysed on different occasions by both u.v. and electrochemical detection. The CV of both assays were less than 10 % and averaged 5 %.

Other sources of direct analyses of phylloquinone in foods

Reliable and routine assays for K_1 in biological tissues have only been available since the introduction of physicochemical assays based on HPLC. Thus, in compiling this database we have only included values obtained by HPLC. Apart from our own measurements, we used the values from two other sources; those of Langenberg *et al*. (1986), who analysed mainly vegetables and considered the effects of freezing and cooking, and those from several publications from the US Department of Agriculture Human Nutrition Research Center on Aging (Booth *et al*. 1993, 1994, 1995*a*) where work is being done on an equivalent USA food database.

A comparison of values from the USA and the UK have shown the same range of values for ‘identical’ foods, such as

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vegetable oils, milk and green vegetables. However, analysis of food in different states (raw *v.* a variety of cooking methods) makes direct comparison difficult. Some commercial products, for example white bread (4·2 μg/kg (MJ Shearer, unpublished results) *v.* 19 μg/kg (Booth *et al.* 1995*a*) in the UK and USA respectively) may differ due to different fat types and proportions used in their manufacture. Where either direct analytical or calculated values (based on analysis of raw ingredients and recipes or manufacturer information) were available for specific brands in the UK, these values have been used in preference to (even directly analysed) values for similar foods in the USA (e.g. pork sausages: from the UK analysed raw value the fried value was calculated as 1·2 μg/kg, whilst the USA analysed value of ‘pancooked’ pork sausage was 34 μg/kg (Booth *et al.* 1995*a*); condensed tomato soup: from UK recipe calculation and manufacturers’ information an average value of 60 μg/kg was calculated, compared with an analysed value of 15 μg/kg by Booth *et al.* (1995*a*) in the USA).

Assigning vitamin K_1 values to foods in the absence of direct analysis

Recipe-based calculation. Values for K_1 contents were calculated for a total of 605 recipes. These were derived from four sources: (1) Recipes (*n* 295) from the *McCance and Widdowson’s The Composition of Foods* reference texts (Tan *et al*. 1985; Holland *et al*. 1988, 1989, 1991*a*,*b*, 1992*a*,*b*, 1993; Chan *et al*. 1994, 1995). These were entered onto Microdiet program (version 9.03, University of Salford, Manchester, UK) under a temporary code, with the codes chosen being for the raw ingredients unless the recipe stated otherwise. The Microdiet program automatically adjusted for any weight loss or gain during the cooking process and the final calculated K_1 value for each recipe was entered under the existing Royal Society of Chemistry/Ministry of Agriculture, Fisheries and Food (RSC/MAFF) food code. (2) Recipes (*n* 199) that were reported in the weighed food diaries of subjects taking part in a dietary intake study and which were, in most cases, specific to that individual. The recipe was allocated a new code and labelled. A note was made of any additional water or stock and changes in weight were incorporated into the calculation. (3) In certain cases (*n* 28), where an RSC/MAFF food code existed but no recipe was given, recipes were used from a variety of common cookery books. Where no figures have been available for weight loss or gain during cooking, an estimated change in weight was deduced by comparing with a similar recipe in the referenced RSC/MAFF texts. (4) Recipes from common cookery books were again used to estimate K_1 values for foods reported in subjects’ diaries, but without a given recipe (*n* 83).

A general problem arose during this work, due to the periodic publishing of further supplements to *McCance and Widdowson’s The Composition of Foods* (Holland *et al*. 1991*c*). This could have entailed the compilation of the database several times over without necessarily resulting in improvements, since some foods in the supplements to the 4th edition (Holland *et al*. 1988, 1989, 1991*a*) which were used, were not available in the 5th edition or its supplements. Thus, codes are still reported here for the 4th edition supplements on cereals and cereal products (Holland *et al*. 1988), milk and milk products (Holland *et al*. 1989) and vegetables, herbs and spices (Holland *et al*. 1991*a*). Whilst the *Immigrant Foods* supplement to the 4th edition (Tan *et al*. 1985) was used in the original compilation of the database, these have been updated to the 5th edition codes, with careful consideration of equivalence.

Other considerations had to be taken into account for calculating the K_1 content from recipes: whole pasteurized milk (code 190) was used in composite dishes unless specified otherwise. Blended margarine (code 309) was used in recipes where the type of margarine was unspecified. Blended vegetable oil (code 333) was chosen when the type of oil was not stated. White plain flour was chosen for those recipes where the type was not specified, and spices, dried herbs, herbal teas and seasonings were assumed to contribute negligible amounts of K_1 due to the small quantities used. A few example calculations were performed to illustrate the effect on K_1 composition of varying the type of fat in a given food or recipe.

Information from food manufacturers. Food manufacturers were predictably unwilling to impart detailed (confidential) product information other than that normally provided on the label, which was insufficient for our purposes. However, with repeated approaches, some did provide data on the types of oil generally used, and the proportions of fruits and vegetables etc. in many items. Other firms have provided lists of ingredients but without the proportions and therefore an educated guess has given us an approximate value. Two companies, with both the interest and the resources, performed their own calculations on a range of products using the analysed values that we supplied for the raw ingredients. These values then provided a basis for assessing other ‘like-foods’, and in the case of a few margarines we were also able to compare the manufacturer’s calculated value with direct analysis.

Other methods employed. The other principal method of assigning a K_1 value to a food item which had not been analysed was the similarity between foods, taking into account food type, fat and water content, edible portion (where appropriate) and degree of green pigmentation. For example, all white fish of similar fat content were assigned the analysed value for cod. Other values for white fish were adjusted for relative increase or decrease in fat content.

In the majority of these cases we have a high degree of confidence in the resulting value range, since most foods fell into either the ‘negligible’ (< 20 μg/kg) or ‘very high’ categories (>1500 μg/kg). Calculations of the effect of processing on items such as dried fruit and textured vegetable protein are more likely to contain errors, since only water and/or fat loss or gain, and sometimes, comparison between ‘like foods’, could reasonably be taken into account. Again, however, the broad range of K_1 values is likely to be correct. A brief summary of the methods employed, for each food group, follows.

Cereals and cereal products: The K_1 values for different types of bread and rolls were based on the available analysis of white and wholemeal breads taking into account the differences in fat and water content. Yeasts do not contain any K_1. Eight varieties of retail biscuits, seven from the same manufacturer, were analysed directly and had values

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ranging from 12 to 53 μg/kg; the differences were presumed to mainly reflect the type and blend of oils rather than the cereal composition. For most biscuits, the values were probably underestimates of 'phylloquinone' content because of the presence of 2′,3′ dihydro-phylloquinone (K_1 (I-H_2)) in the hydrogenated vegetable oils used in their manufacture. This production of K_1(I-H_2) from K_1 during the hydrogenation process of vegetable oils was recently reported (Davidson *et al*. 1995). Inspection of the chromatograms from the analysis of biscuits showed variable amounts of K_1(I-H_2) that, in some, exceeded the K_1 content. In US children an estimated 30 % of total vitamin K was consumed in this dihydro form of phylloquinone (Booth *et al*. 1996*b*). The relative bioactivity of K_1(I-H_2) is currently unknown. Where different codes in *McCance and Widdowson's The Composition of Foods* (Holland *et al*. 1991*c*) are allocated to home-made and retail products (e.g. oatcakes and gingernuts) and the retail biscuit had not been analysed, the retail K_1 value was assumed to be the same as the value for the home-made variety (by recipe calculation) since there was insufficient information available from the manufacturers. Kellogg's breakfast cereals were calculated by Kellogg (Manchester, UK) using the K_1 values for the basic ingredients which we supplied to them. The K_1 content of other brands of breakfast cereals were calculated by comparing their grain type and fat content with that of the known breakfast cereals. Values for cooked rice and pasta were estimated from the analysed dry values by taking into account the changes in water content.

Milk, milk products and eggs: Several assays were performed on full-fat milk that had been pasteurized and homogenized and the mean of the various values was calculated. Channel Island, skimmed, evaporated and condensed milk were also analysed separately. For cream, K_1 was analysed in fresh double cream and therefore the differences in fat content were examined and used to determine the values for all other varieties of cream. The figures for fresh cream compared with ultra-heat treated or the frozen varieties were assumed to be the same since the fat content did not alter greatly and K_1 is not destroyed on heating. Seven different varieties of cheeses were analysed. Where no analysis was available, a K_1 value was estimated by comparing the fat and water content with those cheeses it most closely resembled in appearance and composition. The K_1 values for the different types of yoghurts were all estimated from Greek yoghurt, which was analysed, and estimates were based purely on the fat content and not the fruit content which would vary considerably. The value for whole egg was calculated from the analysed values for raw yolk (30 μg/kg) and white (undetectable) taking the proportion of yolk to white as 40 : 60.

Vegetables, beans, herbs and spices: Values for vegetables were obtained by direct analysis and where necessary estimating the changes in the K_1 content during either freezing or the cooking. Direct analyses made by Langenberg *et al*. (1986) showed that cooking vegetables in water did not result in any losses in K_1 content other than those that could be explained by the uptake of water. One exception was spinach, which lost water during cooking resulting in a relative gain in K_1 content per unit weight. The relative effects of cooking and freezing on K_1 contents of different vegetables were noted and applied to the figures for the raw vegetables obtained by analysis. Where certain foods were cooked in oil or fat, the uptake of fat was recorded and the K_1 values for that particular fat was added to the K_1 value for the raw vegetable, with adjustment for water changes as appropriate. For some foods (e.g. chips and roast potatoes), the change in the carbohydrate proportion was considered the best indicator of the change in hydration on cooking. In these cases, where more than one method of calculation could be considered a valid approach, it is important to note that the differences in the calculated K_1 composition for a food was, for most practical purposes, negligible (<10 μg/kg). Another factor that was occasionally considered when determining the levels of K_1 in certain fruit and vegetables was the colour. Analyses of the inner, middle, and outer leaves of cabbage indicated progressively more K_1 in the darker leaves (Shearer *et al*. 1980). Those foods with a negligible fat content and lack of colour, e.g. water chestnuts and bean sprouts, were assumed to have a value of zero as an interim measure, pending direct analysis. When values for some vegetables were unknown, this was taken as a guideline for finding a suitable 'like-vegetable'. Values for a small number of the more commonly used fresh herbs were available from direct analysis by ourselves (parsley) or from Booth *et al.* (1993) (chives, coriander and mint). Values for these same herbs in the dried state were calculated on the basis of water content of the fresh herb; other dried green herbs were assigned the mean value of those analysed. It was not possible to realistically estimate values for some ground spices and these have been temporarily assigned a value of zero.

Fruits, nuts and seeds: All the fruits were analysed raw. Only the edible portion was analysed in the majority of cases but this included the skin where appropriate e.g. pears, apples. Where K_1 values were required for whole fruits, e.g. melon, oranges, then the calculation involved taking into account the figure for the edible portion as listed in *McCance and Widdowson's The Composition of Foods* (Holland *et al*. 1991*c*). The values for dried fruits were estimated by assessing the difference in water content, although that of 'sun-dried' fruits might be expected to be far lower due to the u.v. light exposure. The only type of nuts which had been analysed were peanuts, pecans and pistachios. K_1 values for all other varieties were estimated from these figures on the basis of fat content.

Fish and fish products: The only white fish analysed was cod; therefore, the values for all other white fish were estimated using cod as a reference value. K_1 values for oily fish were based on analysed values for salmon, sardines and mackerel. Tinned tuna and pilchards in brine were also analysed, and the K_1 values for those fish canned in oil was estimated by taking the mean value from the following oils: maize oil, soyabean oil, olive oil and rapeseed oil. The difference in fat content between the raw and tinned versions was noted and calculated accordingly. The vitamin K_1 values for shellfish were based on the analysed value for prawns.

Meat and meat products: Values for the different cuts of beef were estimated from our analyses of either raw minced beef or beef steak (rump) which had 22 μg and 8 μg K_1/kg respectively. The K_1 content of pork chop meat was low

(0·3 μg/kg) and undetectable in bacon; therefore gammon and ham were also assumed to contain only trace amounts. Those meats products with high fat content e.g. salami, luncheon meat etc. were compared with pork sausages with regard to fat content.

Fats and oils: Butter was assigned the mean analysed value of five different varieties marketed in the UK. The figure for olive oil was calculated by taking a mean of extra virgin oils (849 μg/kg) and the cheaper oils from multiple pressings (300 μg/kg). The figure of 450 μg K_1/kg allocated to a blended margarine (code 309) was calculated by taking a mean of six different, well-known household brands, namely a soya-based margarine (Sainsbury's own brand) analysed in the 1980s, and Echo, Blue Band, Stork SB, Krona Gold and Summer County, all analysed in 1995. This figure was incorporated into recipes where the type of margarine used was not known. Soft animal and vegetable margarines are grouped together under one code (code 312) on Microdiet. A figure for this group was calculated by taking the mean of Stork SB (analysed value 360 μg/kg), Krona Gold (analysed value 120 μg/kg) and I Can't Believe It's Not Butter (280 μg/kg, value calculated by Van den Bergh Foods Ltd, Crawley, Sussex, UK). These values were available from direct analysis and calculations provided by Van den Bergh Foods Ltd based on the analysed values of the individual oils used. Margarines also contained variable (unquantified) amounts of K_1(I-H_2). Blended vegetable oil was assumed to contain approximately 900 g rapeseed oil/kg and 100 g soyabean oil/kg (values supplied by the Ministry of Agriculture, Fisheries and Food) and the K_1 value was calculated on that basis.

Beverages: All alcoholic drinks were assumed to contain only a trace of K_1 (<0·1 μg/kg) as judged from the analysed values for red wine and beer (bitter, lager and Guinness). An infusion of instant coffee (5 g coffee in 120 ml water) was found to contain 1·8 μg K_1/l, and a similar infusion of decaffeinated coffee contained 0·5 μg/l. From this, the amount in an average cup (2 g in 150 ml) was calculated to be 0·6 μg/l, and the K_1 content of instant coffee powder was calculated as 43 μg/kg. Both an infusion of tea (15 g tea/l) and dried tea leaves were analysed.

Miscellaneous foods: The K_1 content of jam made from fruit with edible seeds was calculated by assuming a fruit content of 350 g/kg and choosing strawberry and raspberry as the jams most commonly consumed. Jam made with stoned fruit used plums as the source of fruit. Marmalade was assumed to have only a trace, since oranges are extremely low in K_1. Confectionery items such as pastilles, boiled sweets, peppermints, marshmallows and fruit gums that primarily consist of sugar were given a K_1 value of zero. Milk and white chocolate were estimated from the value given for plain dark chocolate by assessing the quantity of fat present. Chocolate bars were examined by estimating the proportion of chocolate, toffee, fudge etc. and attempting to reach similar nutritional values as those quoted in *McCance and Widdowson's The Composition of Foods* (Holland *et al*. 1991*c*). The brand-name snack foods Mars bar and Kit Kat were also specifically analysed. Savoury snacks such as tortilla chips, potato hoops and corn snacks were given a vitamin K_1 value based on their fat content and comparison with the analysed values for potato crisps (96 μg/kg) and maize-based cheese puffs (Wotsits, 155 μg/kg).

Results

Using the variety of techniques as described, it was possible to assign vitamin K_1 compositional values to a total of 1783 foods. K_1 values now exist for 1178 foods and 323 recipes with an RSC/MAFF food code and a further 282 new recipes have been given a code. The number of foods in each food group (excluding the recipes) are: milk, milk products and eggs 133; vegetables and vegetable dishes, 279; fruits, 158; cereals and cereal products, 134; fats and oils, 29; nuts and seeds, 24; fish and fish products, 95; meat and poultry, 132; soft and alcoholic beverages, 48; herbs and spices, 41; miscellaneous (including soups, sauces and confectionery), 105.

Table 1 compares the K_1 content of some margarines as determined by calculation from the raw ingredients by the manufacturers and by direct analysis at two time points. Table 2 illustrates the effect of margarine or oil type on the K_1 content of several foods, as calculated from recipes. Table 3 gives details of the analysed variation in the K_1

Table 1. Comparison of phylloquinone (K_1) content in fat spreads, margarines and oils by analysis and calculation (μg/kg)

	RSC/MAFF food code*	HPLC analysis (1980s)†	HPLC analysis (1995)‡	Calculated value from the composite oils (1995)§
Flora (sunflower spread)	17023	620	130	140
Blue Band (soft margarine)	17021	1180	780	690
Stork SB (soft margarine)	17020	not analysed	360	180
Delight (low-fat spread)	17026	not analysed	360	200
Olivio (olive-oil spread)	17025	not analysed	560	490
Krona Gold (hard margarine)	17022	120	120	60
Echo (hardened animal and vegetable fat for baking)	17018	290	90	90
Summer County (soft margarine)	17021	not analysed	200	130
Spry Crisp & Dry (vegetable oil)	333	not analysed	1340	1130

RSC/MFF, Royal Society of Chemistry/Ministry of Agriculture, Fisheries and Foods.
* The appropriate food code into which each fat type would be categorized.
† MJ Shearer (1980s), unpublished results.
‡ MJ Shearer (1995), unpublished results.
§ Calculation by Van Den Bergh Foods Ltd, Crawley, Sussex, UK, from analysed values of the constituent oils by MJ Shearer (unpublished results).

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Table 2. Examples of the effect of different fat sources on the phylloquinone (K_1) composition of foods, by calculation

Food name	RSC/MAFF code	Phylloquinone (μg/kg)
Chips fried in blended vegetable oil (code 333)	674	88
Chips fried in maize oil (code 322)	675	18
Chips fried in dripping (beef fat, code 317)	676	32
Home-made biscuits made with soft margarine (animal and vegetable, code 312)	100	68
Home-made biscuits made with high polyunsaturated margarine (code 314)	100	36
Cherry cake made with blended vegetable oil (code 333)	11193	130
Cherry cake made with butter (code 306)	11193	39

RSC/MAFF, Royal Society of Chemistry/Ministry of Agriculture Fisheries and Food.

content of the inner and outer leaves of green vegetables, and Table 4 provides representative K_1 compositional values for foods in each food group. The values assigned to the RSC/MAFF food code in the database are given along with the number of food samples analysed and the range of values in parentheses. Where only one food sample was used, the number of separate extractions which were carried out and the range of these results are all given in parentheses.

Discussion

This paper reports the methods that have been used to produce the first food composition database for phylloquinone (K_1) in the UK. Care has been taken to use only those analysed values that are appropriate to the UK situation, and that have been measured by accurate physico-chemical methods based on HPLC. Generally, either two or more samples have been analysed, or different samples pooled before analysis to give one mean value. Calculations have been based on the best information available and particularly rough estimates, due to the lack of any comparative data, have been emphasized. The similar range of K_1 values in basic food items to those reported in other countries (Booth *et al*. 1995*a*; Jakob & Elmadfa, 1996; Koivu *et al*. 1997, 1998; Piironen *et al*. 1997) provide further support for these data.

Infant formula milks and baby foods have not been included in this database as they are generally supplemented with K_1 well above the level of their natural constituents. Information has been published (Haroon *et al*. 1982), and some vitamin K levels are also mentioned in the *Nutrition in Infancy* briefing paper (Wharton, 1997), but otherwise the statutory labels on each item should provide this information.

The differences between the 1980s and 1995 HPLC analyses of K_1 in the margarines (Table 1) is primarily due to the change in oil composition of the margarines over this time. The differences between the 1995 HPLC analysed and calculated K_1 values were minor, and likely to reflect the errors entailed from using several different analysed values of oils to perform the calculations *v*. a single extraction and analysis step for the laboratory value. In all cases, the most modern laboratory analysed value has been used in the database. Ideally, this should be repeated whenever the formulations change if the K_1 food composition database is to be 'current'. Changes in the types of oils used in food manufacture is a general problem which precludes food composition tables remaining up-to-date, especially for the fatty acids and fat-soluble vitamins. In this respect, regular communication between the manufacturers and the researchers and analysts would be desirable, at least for some agreed 'staple' food items. The effect of fat type used in cooking (Table 2) further highlights the need to know the composition of these items if the accuracy of an assessment of K_1 intake is to be maximized.

The variation in K_1 content between inner and outer leaves of green vegetables, shown in Table 3, is just one of the factors contributing to variability; Ferland & Sadowski (1992*a*) have reported differences by stage of maturation and geographical location.

The main table of K_1 food compositional values (Table 4)

Table 3. Examples of the variation in the phylloquinone (K_1) content of green vegetables*

Food name	RSC/MAFF food code	No. of food samples†	Range of values (μg/kg)‡	Average phylloquinone content (μg/kg)
Brocoli, raw	744	3	1470,1840,2050	1790
Brussel sprout tops, raw, whole	746	3	1150,1480,1770	1530
Brussel sprout tops, outer leaves	NA	1 (2)	(4500,4610)	4560
Brussel sprout tops, middle leaves	NA	1 (2)	(3970,4100)	4040
Brussel sprouts, inner leaves (yellow)	NA	1 (2)	(3340,3480)	3410
Cabbage, winter, raw outer leaves	NA	1 (2)	(1740,2040)	1890
Cabbage, winter, raw inner leaves	751	1 (4)	(460,600)	520
Spinach, raw	813	3	2940,4150,4330	3810

RSC/MAFF, Royal Society of Chemistry/Ministry of Agriculture, Fisheries and Food; NA, not applicable,

* Analysed in the laboratory of M. J. Shearer.

† Number of separate food samples collected for analysis. When only one sample was analysed, the numbers in parentheses indicate the number of separate extractions.

‡ Range of K_1 values for the food samples. When only one sample was analysed, the numbers in parentheses indicate the range for the separate extractions.

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Table 4. The phylloquinone (K_1) composition of a representative range of foods, by food group

	RSC/MAFF food code	Assigned K_1 value (μg/kg)	No. of food samples*	Range of K_1 (μg/kg)†	Source of the assigned K_1 value‡
Vegetables and beans					
Potatoes, boiled	668	9·4	NA	NA	Calculated from raw, *n* 2, mean 9: MJS
Broccoli, boiled	745	1310	NA	NA	Calculated from raw, *n* 3, mean 1790: MJS
Brussel sprouts, boiled	747	1220	NA	NA	Calculated from raw, *n* 3, mean 1530: MJS
Spinach, boiled	814	5750	NA	NA	Calculated from raw, *n* 3, mean 3810
Parsley	846	5480	1 (2)	(5300,5660)	MJS
Cauliflower, raw	759	310	3	270–390	MJS
French/green beans	707	360	2	260–460	MJS
Lettuce, average raw	777	1290	4	1200–1400	MJS, three varieties (Webbs, Round, Iceberg)
Tomatoes, raw	827	60	1 (2)	(48,71)	MJS
Carrots, old raw	754	55	1 (2)	(53,56)	MJS
Baked beans	694	27	1 (2)	(26,29)	MJS (H.J. Heinz Co. Ltd)
Lentils, red, split dry	712	220	NA	NA	Booth *et al.* 1993, 1995*a*
Coleslaw with mayonnaise	15077	451	NA	NA	Recipe (retail mayonnaise)
Vegetable flan	15175	278	NA	NA	Recipe (blended margarine)
Cauliflower cheese	166	233	NA	NA	Recipe (whole milk)
Pakora/bhajia	15230	314	NA	NA	Recipe (fried in blended vegetable oil)
Fruit and nuts					
Apple, Cox's with skin	14017	56	1 (2)	(44,67)	MJS
Apple, stewed (no skin)	855	3	NA	NA	Booth *et al.* 1993, 1995
Kiwi fruit	908	250	NA	NA	Booth *et al.* 1993, 1995
Grapes, average (no seeds)	903	86	2	81,91	MJS (white and black grapes analysed separately)
Raisins	958	37	1 (2)	(32,44)	MJS
Oranges, fresh (no peel)	931	0·5	1 (2)	(0·4,0·5)	MJS
Peanuts, dry roasted	989	3·9	1 (2)	(3·8,4·0)	MJS
Fats and oils					
Soyabean oil	331	1310	2	1120,1500	MJS (aged and fresh samples respectively)
Rapeseed oil	327	1125	1 (2)	(1120,1130)	MJS
Olive oil	324	548	3	300,740,850	MJS (mean of standard (300 μg/kg) and two extra virgin samples)
Maize oil	322	31	1 (2)	(28,34)	MJS (Mazola)
Blended vegetable oil	333	1144	NA	NA	Calculated as 900 g rapeseed oil/kg and 100 g soyabean oil/kg
Butter	306	74	5	39–95	MJS (mean of five varieties)
Delight, low-fat spread	308	360	1 (4)	(350–370)	MJS
Dripping (beef fat)	317	245	1 (2)	(240,250)	MJS
Margarine, blended	309	430	6	120–780	Calculated, mean of six brands
Milk and milk products					
Milk, full-fat	190	6	24	3·6–9	MJS (samples taken over 12 months)
Semi-skimmed milk	186	2	NA	NA	Calculated from full-fat milk
Cheddar cheese	228	47	2	(35,59)	MJS
Full-fat soft cheese	242	47	1 (2)	(46,47)	MJS
Greek yoghurt, cows	252	7·8	1 (4)	(7·3–8·4)	MJS
Yoghurt, fruit, low-fat	12190	1	NA	NA	Calculated from Greek yoghurt
Ice cream, vanilla, non-dairy	269	8	1	(7·2,8·2)	MJS (Tesco's own brand)
Fish					
Cod, raw flesh only	563	0·1	1 (2)	(0·1,0·2)	MJS
Tuna, tinned in brine	632	2·5	3	1·1–4·5	MJS
Tuna, tinned in oil	631	64	NA	NA	Calculated from 'Tuna, tinned in brine' + mean of maize, soyabean, olive and rapeseed oils
Beverages (μg/l)					
Tea, infusion	1079	2·7	2	2·3,3·2	MJS (Tetley and PG Tips)
Coffee, infusion	1053	0·6	NA	NA	Calculated from instant granules assuming 10 g/l
Orange juice, unsweetened	1091	0·6	1 (2)	(0·5,0·6)	MJS
Beer, draught bitter	1095	Tr	1 (1)	$<0·1$	MJS (trace only from each of lager, bitter and stout)
Wine, red	1107	Tr	1 (1)	$<0·1$	MJS
Spirits, 70 % proof	1122	Tr	1 (1)	$<0·1$	MJS
Cereals					
White bread, sliced	49	4·2	1 (2)	(3·8,4·6)	MJS (Mothers' Pride)
Hovis, sliced	40	20	1 (2)	(19,21)	MJS
Cornflakes	69	0·6	1 (1)	NA	MJS

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Table 4. (*continued*)

	RSC/MAFF food code	Assigned K_1 value (μg/kg)	No. of food samples*	Range of K_1 (μg/kg)†	Source of the assigned K_1 value‡
White rice, boiled	23	0·3	NA	NA	Calculated from raw value 1·2 (American long grain)
Pasta, spaghetti, boiled	30	0·5	NA	NA	Calculated from raw value 2·1 (Sainsbury's durum wheat)
Sponge cake with fat	119	181	NA	NA	Recipe: blended margarine
Gingernut biscuit (retail)	99	16	1 (2)	(16,17)	MJS (McVities' Ginger Crunch)
Gingernut biscuits (home-made)	11173	149	NA	NA	Calculated from recipe: blended margarine
Meat and Poultry					
Mince beef, stewed	370	23·8	NA	NA	Calculated from raw value 21·8
Sausages, pork and beef, raw	19087	1·6	1 (2)	(1·5,1·8)	MJS (Walls, chipolata)
Chicken, roast, light meat	440	0·5	NA	NA	Calculated from raw value of thigh 0·5
Sausage roll, flaky	522	125	NA	NA	Calculated from recipe
Meat samosa	174	166	NA	NA	Calculated from recipe
Miscellaneous					
Bone and vegetable broth	937	93	NA	NA	Calculated from recipe
Chocolate, plain	858	23	1 (4)	(21·8–23·4)	MJS
Jam, edible seeds	849	9	NA	NA	Calculated from recipe
Mayonnaise	12277	433	NA	NA	Calculated from recipe

RSC/MAFF, Royal Society of Chemistry/Ministry of Agriculture, Fisheries and Food; NA, not applicable to calculated values, or see the source reference(s) cited; MJS, direct analysis in laboratory of M. J. Shearer; Tr, trace.

* Number of separate food samples collected for analysis. When only one sample was analysed, the numbers in parentheses indicate the number of separate extractions.

† Range of K_1 values for the food samples. When only one sample was analysed, the numbers in parentheses indicate the range for the separate extractions.

‡ See pp. 391–393.

makes it clear why the current database should be considered as a preliminary version because further extensive work is necessary to extend the number of samples analysed and to confirm the calculated values, for at least some foods, by direct analysis. These foods include some for which there is currently no feasible way of obtaining a K_1 value and others for which the 'like-food' and educated guess-work approach is clearly highly unsatisfactory (e.g. textured soya protein, mycoprotein, sun-dried fruits, salami-style sausage). Other aspects of vitamin K nutrition that have not been addressed include loss of K_1 due to u.v. light exposure (Ferland & Sadowski 1992*b*), whether due to lit supermarket display-cabinets or home storage (a single *ad hoc* experiment indicated a 50 % decrease in the K_1 content of vegetable oil in a standard transparent bottle after 6 months on a kitchen worktop (MJ Shearer and DJ Harrington, unpublished results)) and the importance of both K_1(I-H_2) and dietary menaquinones (MK) to total vitamin K intake. Probably the two most important sources of MK are cheeses (MK9, 50–200 μg/kg, from the starter bacteria) and ruminant liver (100–200 μg/kg) (Shearer *et al*. 1996), although some meats, e.g. chicken, also contain MK4, probably derived directly from their diet (C Vermeer, personal communication). Without further food analysis it is impossible to estimate levels of MK in the diet; however, it is likely to be only a small fraction of total vitamin K intake, unless an individual's K_1 intake is low and their diet is particularly high in these MK-rich foods.

Little is yet known about the bioavailability of phylloquinone or MK from different foodstuffs. However, a recent study of plasma concentration–time profiles in five human subjects (Gijsbers *et al*. 1996) showed that the intestinal absorption of K_1 from boiled spinach was very inefficient, but was improved by the addition of butter to the test meal. They estimated that <10 % of the K_1 content of spinach was absorbed, which is less than had generally been assumed. The dependence of absorption of K_1 on other food components in the meal, in this case fat, is in agreement with the known bioavailability of other hydrophobic micronutrients from plants, such as the carotenoids. The absorption of different MK is likely to depend on their relative hydrophobicity. Studies in rats have shown MK9 absorption to be poorer than K_1, whilst activity may be longer due to slower turnover (Will & Suttie, 1992; Groenen-van Dooren *et al*. 1995).

Issues such as storage and bioavailability clearly need addressing further. For example, if marginal vitamin K adequacy is occurring in any sector of the population, then strategies to preserve the natural phylloquinone content of foods may be particularly important and relatively easy to achieve (e.g. brown bottles, reduced lighting, etc.). Greater knowledge of bioavailability would facilitate the setting of dietary reference values and also enable appropriate advice to be given to maximize intestinal absorption.

In spite of these caveats, the current database is likely to be able to provide at least as accurate an assessment of K_1 intake as the existing RSC/MAFF database can for vitamin E. It is also likely to be able to provide adequate assessment of the major food groups which supply K_1 in the diet (Fenton *et al*. 1997). This is the case because the pattern of K_1 distribution in foods and food groups is largely unambiguous: green vegetables having the highest values, followed by fats and oils, and composite dishes which contain one or other, or both, of these food types.

Different K_1 values may be obtained for a single food item: for vegetables this will depend on freshness, degree of green pigmentation, plant maturation and possibly climatology (Booth *et al*. 1993), while for composite dishes the type of fats and oils used in recipes introduces enormous variation. These issues illustrate the importance of never taking food compositional data entirely at face value. It would be appropriate for others who wish to estimate the intake of K_1 to consider these aspects, and to perform their own recipe calculations if the fat source in dishes is known to be different from that indicated.

Given these provisos, and in spite of the need for further refinement and extension, the current K_1 food composition database will allow the calculation of new K_1 values for different recipes, facilitate the assessment of dietary K_1 intake to within an estimated 10–15 μg/d from a given set of foods, and provide essential guidance to clinicians and dietitians who aim to stabilize a patient's K_1 intake whilst on oral anticoagulant therapy (Lubetsky *et al.* 1999). If future evidence justifies a recommendation to increase dietary intake of K_1 for optimal health, the necessary information on the composition of UK foods is now available to facilitate informed food choice. Following the further work that is needed on the database, to both confirm and extend the values, the aim is to encourage access to the database by including the K_1 values in new supplements to, and new editions of, the UK food composition tables.

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Appendix

Example calculations (values are expressed per kg)

(1) Deep fried chipped potatoes

Potato, old raw and peeled (code 664)

Water 795 g, fat 2 g, carbohydrate 172 g, K_1 9·4 μg

Chips, fried in maize oil (code 675)

Water 565 g, fat 67 g, carbohydrate 301 g

Taking the change in carbohydrate content as an indicator of the quantity of raw potato in 1 kg of deep-fried chips, $(301/172) = 1{\cdot}75$ kg then:

the K_1 content of the potato component is $9{\cdot}4 \times 1{\cdot}75 = 16{\cdot}5$

the fat content of the potato component is $2 \times 1{\cdot}75 = 3{\cdot}5$

the quantity of absorbed fat $(67 - 3{\cdot}5) = 63{\cdot}5$ g

the K_1 content of 63·5 g of maize oil is $63{\cdot}5 \times 0{\cdot}03 = 1{\cdot}9$ μg

Vitamin K_1 content is $16{\cdot}5 + 1{\cdot}9 = 18$ μg/kg

(2) Butter beans, soaked and boiled

Butter/lima beans, dry (code 13070)

Water 116 g, fat 17 g, vitamin K 60 μg

Butter/lima beans, soaked and boiled (13071)

Water 705·1 g, fat 6 g

Fat content reduces $(6/17) \times 60 = 21{\cdot}2$ μg

Vitamin K_1 content = 21 μg/kg

(3) Whole avocado pear
Avocado, flesh only (code 865)
Edible portion 1000 g, vitamin K 140 μg
Avocado, weighed with skin and stone (code 14038)
Edible portion 710 g
Vitamin K_1 content is $(710/1000) \times 140 = 99{\cdot}4$ μg/kg

(4) Fresh single cream
Cream, fresh double (code 215)
Water 475 g, fat 480 g, vitamin K 64 μg
Cream, fresh single (code 212)
Water 737 g, fat 191 g
Adjusting for the decreased fat content: vitamin K_1 content is $(191/480) \times 64 = 25{\cdot}5$ μg/kg

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