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Cite this: *Food Funct.*, 2024, **15**, 10414

Development of a food frequency questionnaire for the estimation of dietary (poly)phenol intake†

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Background: (Poly)phenol intake has been associated with reduced risk of non-communicable diseases in epidemiological studies. However, there are currently no dietary assessment tools specifically developed to estimate (poly)phenol intake in the UK population. **Objectives:** This study aimed to develop a novel food frequency questionnaire (FFQ) to capture the dietary (poly)phenol intake in the UK and assess its relative validity with 7 day diet diaries (7DDs) and plasma and urine (poly)phenol metabolites. **Methods:** The KCL (poly)phenol FFQ (KP-FFQ) was developed based on the existing EPIC (European Prospective Investigation into Diet and Cancer)-Norfolk FFQ, which has been validated for energy and nutrient intake estimation in the UK population. Participants aged 18–29 years ($n = 255$) completed both the KP-FFQ and the EPIC-Norfolk FFQ. In a subgroup ($n = 60$), 7DD, spot urine, and fasting plasma samples were collected. An in-house (poly)phenol database was used to estimate (poly)phenol intake from FFQs and 7DDs. Plasma and urinary (poly)phenol metabolite levels were analysed using a validated ultra-high-performance liquid chromatography-triple quadrupole mass spectrometry method. The agreements between (poly)phenol intake estimated using the KP-FFQ, EPIC-Norfolk FFQ and 7DDs, as well as plasma and urinary biomarkers, were evaluated by intraclass correlation coefficients (ICC), weighted kappa, quartile cross-classification, and Spearman's correlations, and the associations were investigated using linear regression models adjusting for energy intake and multiple testing (false discovery rate (FDR) < 0.05). **Results:** The mean (standard deviation, SD) of total (poly)phenol intake estimated from KP-FFQs was 1366.5 (1151.7) mg d⁻¹. Fair agreements were observed between ten (poly)phenol groups estimated from KP-FFQs and 7DDs (kappa: 0.41–0.73), including total (poly)phenol intake (kappa = 0.45), while the agreements for the rest of the 17 classes and subclasses were poor (kappa: 0.07–0.39). Strong positive associations with KP-FFQ were found in ten (poly)phenols estimated from 7DDs, including dihydroflavonols, theaflavins, thearubigins, flavones, isoflavonoids, ellagitannins, hydroxyphenylacetic acids, total stilbenes, resveratrol, and tyrosols with stdBeta ranged from 0.61 (95% confidence interval CI: 0.42 to 0.81) to 0.95 (95% CI: 0.86 to 1.03) (all FDR adjusted $p < 0.05$). KP-FFQs estimated (poly)phenol intake exhibited positive associations with 76 urinary metabolites (stdBeta: 0.28 (95% CI: 0.07–0.49) to 0.81 (0.62–1.00)) and 19 plasma metabolites (stdBeta: 0.40 (0.17–0.62)–0.83 (0.64–1.02)) (all FDR $p < 0.05$). The agreement between KP-FFQs and the EPIC-Norfolk FFQs was moderate (ICC 0.51–0.69) for all (poly)phenol subclasses after adjusting for energy intake. Compared with the EPIC-Norfolk FFQs estimated (poly)phenol intake, stronger and more agreements and associations were found in KP-FFQs estimated (poly)phenol with 7DDs and biomarkers. **Conclusion:** (Poly)phenol intake estimated from KP-FFQ exhibited fair agreements and moderate to strong associations with 7DDs and biomarkers, indicating the novel questionnaire may be a promising tool to assess dietary (poly)phenol intake.

Received 24th July 2024,
Accepted 11th September 2024

DOI: 10.1039/d4fo03546a

rsc.li/food-function

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† Electronic supplementary information (ESI) available. See DOI: <https://doi.org/10.1039/d4fo03546a>

1. Introduction

(Poly)phenols are broadly distributed in the plant kingdom.¹ Flavonoids, phenolic acids, lignans, and stilbenes are among the major groups of (poly)phenols, with more than 8000 phenolic structures identified so far.² As abundant secondary metabolites of plants, (poly)phenols are ubiquitous in the human diet, especially in fruits, vegetables, wholegrains,



legumes, nuts, seeds, cocoa and soy products, coffee, and tea. The evidence from randomized clinical trials and observational studies on their health benefits is continuously accumulating, for instance, in cardiovascular health^{3,4} and age-related cognitive protection.⁵ However, the lack of accurate assessment data may hinder the establishment of valid relationships between (poly)phenol and health at the population level.^{4,6,7}

It is challenging to measure the dietary intake of (poly)phenols due to their complex nature, which includes a wide range of structures from single aromatic-ring monomer molecules to intricate condensed polymer tannins found in food.^{8,9} Different (poly)phenols can accumulate in certain plants, resulting in distinct profiles of (poly)phenols in foods. Some (poly)phenols, for instance, quercetin, are widely found in many types of plant foods, including fruits, vegetables, cereals, tea, and wine, whereas some (poly)phenols are specifically abundant in certain foods, for instance, flavanones in citrus fruit and isoflavones in soya.¹⁰ The complexity and variability of (poly)phenol abundance in the human diet require a comprehensive and targeted food list included in assessment tools.¹¹

Currently, dietary (poly)phenol intake information from large cohort studies is mainly collected through food frequency questionnaires (FFQs), due to the low burden on both participants and researchers.^{12–15} Comparison studies between FFQs and 7 day diet diaries (7DDs) indicated that the agreements were poor for the groups that contribute small percentages to total (poly)phenol intake, such as anthocyanins, chalcones, flavones, and hydroxyphenylacetic acids, which might be due to the difficulty of capturing such food sources with these tools.⁶ Since these tools were designed to estimate habitual nutrient intake, they do not necessarily capture well all the (poly)phenol food sources, so tools with a more comprehensive and detailed list of food items or food groups to accurately evaluate all the (poly)phenol subclasses are needed. In addition, only a few of them have been validated for (poly)phenol intake,¹⁶ which restricts the understanding of dietary assessment tools' performance in evaluating (poly)phenol

intake.^{17,18} Therefore, a dietary assessment tool that has been specifically developed and validated for estimating habitual (poly)phenol intake would be valuable to advancing research on the exposure and health impact of (poly)phenol consumption in the population.

No objective 'gold standard' reference biomarker tool has been established for evaluating (poly)phenol exposure and only a few biomarkers have been partially validated for individual (poly)phenols, including flavan-3-ols^{19,20} and isoflavones.²¹ However, total urinary (poly)phenols, and in particular 24 h urine, have been proposed to reflect total (poly)phenol intake,²² and urine measurements have been used as the reference tool to strengthen the relative validation of 7DDs.²³ Thus, quantitative targeted metabolomics including a comprehensive panel of (poly)phenol metabolites hold the potential to serve as a surrogate marker for (poly)phenol intake.^{24–29}

The primary objectives of this work were to (1) develop a FFQ to capture habitual (poly)phenol intake in the free-living UK population (KCL (poly)phenol FFQ or KP-FFQ), (2) conduct a relative validity study with 7DDs, and (3) test agreements between (poly)phenol intake estimated from the KP-FFQ with objective (poly)phenol metabolites from 24 h urine and plasma samples (Fig. 1). The secondary objectives of this study were to compare the novel KP-FFQ with an established FFQ (the EPIC (European Prospective Investigation into Diet and Cancer)-Norfolk FFQ) in the estimation of (poly)phenol and nutrient intake (Fig. 1).

2. Methods

2.1 Tool development

The KP-FFQ is adapted from the EPIC-Norfolk FFQ. This FFQ is a well-established tool in the UK, having been validated against nutrient intakes in the EPIC Norfolk study.³⁰ The EPIC-Norfolk FFQ was designed to estimate habitual nutrient intake in the UK EPIC-Norfolk study.³¹ It consists of 130 food items typically consumed in the UK with nine frequency

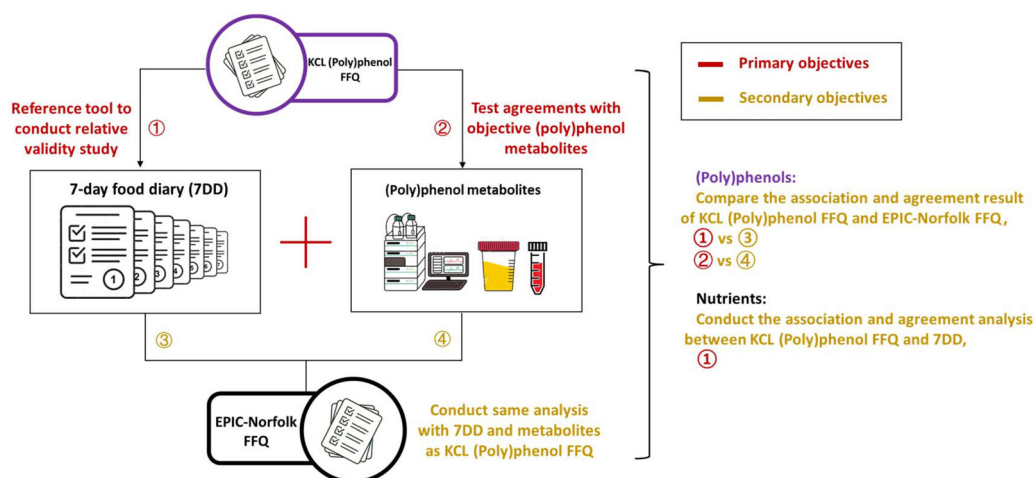


Fig. 1 Primary and secondary research objectives regarding the development and relative validation of the KCL (poly)phenol FFQ.



options from “Never or less than once a month” to “more than six times per day”. Through extensive experience using the EPIC-Norfolk FFQ in our laboratory as part of clinical trials^{32–34} and cross-sectional studies,^{6,35,36} we have identified certain challenges in accurately estimating (poly)phenol intake during its application. This recognition has led us to develop a specialized (poly)phenol-focused version, adapted from the original EPIC-Norfolk FFQ.

The KP-FFQ aims to distinguish between food items with different (poly)phenol content and composition, by either (1) disaggregation of distinct food items listed in the EPIC-Norfolk FFQ as one entry (*e.g.*, single food entry for strawberries, raspberries, and kiwi fruit); (2) differentiation of food groups with different colours (*e.g.*, red, white, and yellow onion); (3) differentiation of parts of the same food source (*e.g.*, peel and pulp); or (4) addition of additional food sources not listed in the EPIC-Norfolk FFQ, *e.g.*, blueberries, based on their (poly)phenol content. In all, food items that met the criteria of ‘providing more than 1 mg of total (poly)phenols per serving’³⁷ or considered to be rich in (poly)phenols were included in the KP-FFQ through expert agreement (ARM, RG, and YX). Food items identified as the main contributors to (poly)phenol intake among children and adults in the UK were also included.³⁸ Food groups (*e.g.*, meat and fish) and food items (*e.g.*, Horlicks, honey) not rich in (poly)phenols from the original EPIC-Norfolk FFQ were also included in the KP-FFQ to retain the structure for estimating overall nutrient intake.

The KP-FFQ required participants to report their average intake of the food items over the last year, ranging from a frequency of ‘never or less than once per month’ to ‘6 + per day’ which were numerically coded as ‘1’ to ‘9’. Missing data were recorded as ‘-9’ throughout the data entry process. Data on the frequency of intake were manually entered into Microsoft Excel sheet as numeric codes. The average daily intake was

derived by multiplying the frequency of intake with default portion sizes. The procedure used for the development of the KP-FFQ is shown in Fig. 2.

The final KP-FFQ included 442 food items. The food items under each food group in the EPIC-Norfolk FFQ and KP-FFQ are compared in ESI Table 1.†

2.2 (Poly)phenol database/estimation of (poly)phenol intake

The (poly)phenol content of all food items was established based on publicly available databases, including Phenol-Explorer,³⁹ the United States Department of Agriculture (USDA) database, and an in-house (poly)phenol database³⁵ that includes common foods and beverages in the UK diet calculated from recipes. Each potential (poly)phenol-rich food item was verified against these databases prior to being included in the KP-FFQ.

The total (poly)phenol content of foods/beverages was calculated using the default average portion sizes, which reflected the average portion sizes of the UK population. The portion sizes were selected based on available UK data (EPIC-Norfolk portion sizes, <https://www.epic-norfolk.org.uk>), Nutritics (<https://www.nutritics.com/en/>), and product information in three major supermarkets in the UK: Sainsbury's™ (<https://food-to-order.sainsburys.co.uk/category/allfood>), Tesco™ (<https://www.tesco.com/>) and Morrisons™ (<https://groceries.morrisons.com/navigation>). (Poly)phenol intake of each food item (mg d^{-1}) was calculated by multiplying the total (poly)phenol content (mg per 100 g) with the default portion size (g) and the intake frequency factor as below:

$$P_{\text{total}} = \sum P_i = \sum (C_i \times Q_i \div 100)$$

$$Q_i = D_i \times F$$

Of which: P_{total} : total (poly)phenol intake (mg d^{-1}); P_i : the (poly)phenol intake of food item (i) in FFQ, mg d^{-1} ; C_i : (poly)

Workflow of KCL (Poly)phenol FFQ

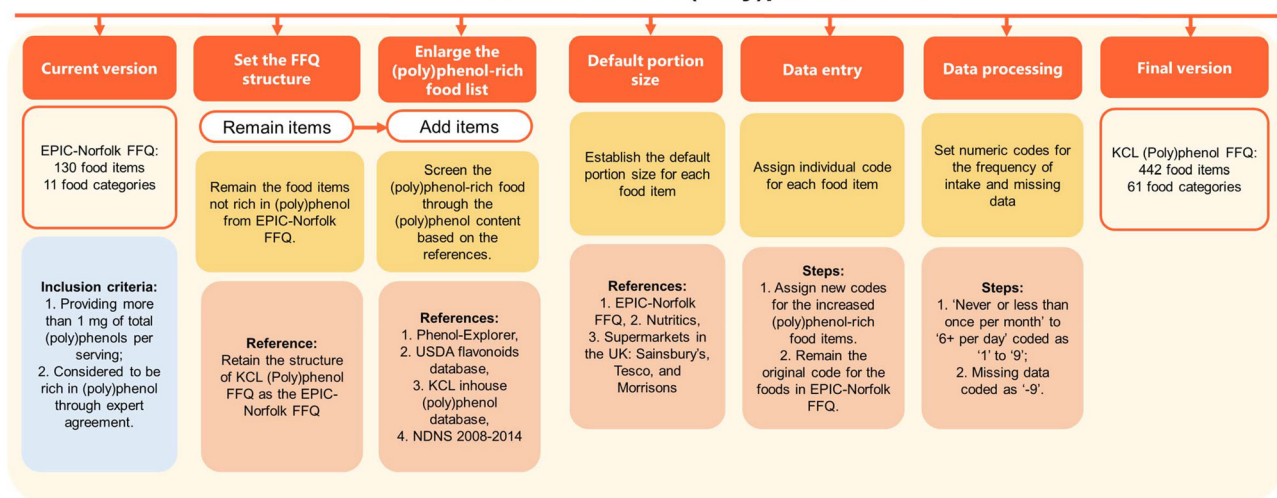


Fig. 2 Summary of processes followed in KCL (poly)phenol FFQ development. EPIC: European Prospective Investigation into Diet and Cancer, FFQ: food frequency questionnaire.



phenol content of the food item (i), mg per 100 g, Q_i : intake quantity of food item (i) (g d^{-1}); D_i : default portion size of food item (i); F : frequency factor (Never or less than once/month: 0; 1–3 per month: 0.07; once a week: 0.14; 2–4 per week: 0.43, 5–6 per week: 0.79, once a day: 1; 2–3 per day: 2.5; 4–5 per day: 4.5; 6 + per day: 6)

2.3 Pilot study

The KP-FFQ was piloted in 20 participants that were recruited from the student population at King's College London. Participants provided written informed consent and were provided with an envelope consisting of the EPIC-Norfolk FFQ, KP-FFQ and a blank 7DD with an allocated randomised code to ensure that no identifiable information was collected by the General Data Protection Regulation 2016 (Research Ethics Minimal Risk Registration number MRA-20/21-24013). Participants were given one week to complete the questionnaires. They were directed to complete the KP-FFQ on the first day and the EPIC-Norfolk FFQ on the seventh day during the 7 day period of recording the 7DD. They were also required to record the time taken to complete each FFQ. The envelopes were returned by the participants to an allocated box in the department office anonymously.

2.4 Evaluation study

Participants enrolled in the KCL (poly)phenol FFQ pilot study, a randomized controlled trial and an observational study conducted from 2021 to 2024 at the Metabolic Research Unit of the Department of Nutritional Sciences, King's College London, UK completed the KP-FFQ along with other baseline

dietary measurements. The three studies applied the same dietary assessment protocols and tools, including EPIC-Norfolk FFQ, KP-FFQ, and 7DD, to obtain the dietary intake of each participant. Healthy participants aged 18–29 years old ($n = 262$) were included to evaluate the KP-FFQ. The studies were approved by the Research Ethics Committee of King's College London (Ethics numbers: KCL (poly)phenol FFQ pilot study: MRA-20/21-24013; FOODMOOD study: HR/DP-21/22-28880; CRANMOOD study HR/DP-21/22-26721; validation of a new FFQ to estimate (poly)phenol intake in CRANMOOD study: LRS/DP-21/22-32445) and conducted following the Declaration of Helsinki. The participants were excluded if no available dietary assessment data from any of the EPIC-Norfolk/KP-FFQs or 7DDs were collected ($n = 27$). Finally, 235 participants with both FFQs and 7DDs were included in the analysis.

A detailed flow chart of this process is exhibited in Fig. 3.

2.5 Study procedures

At the baseline visit, the weight, height, and body composition data of each participant were collected using standard anthropometric methods. Their demographic characteristic, smoking status, alcohol consumption, and physical activity level were collected using case report forms and the International Physical Activity Questionnaire Long Form (IPAQ-LF) to classify participants into high, moderate, or low levels of activity.⁴⁰ Participants were asked to complete three dietary assessment tools, including KP-FFQ, EPIC-Norfolk FFQ, and 7DDs (usually dispensed 1 week prior to the baseline visit). The KP-FFQ was collected to capture their habitual (poly)phenol intake. The

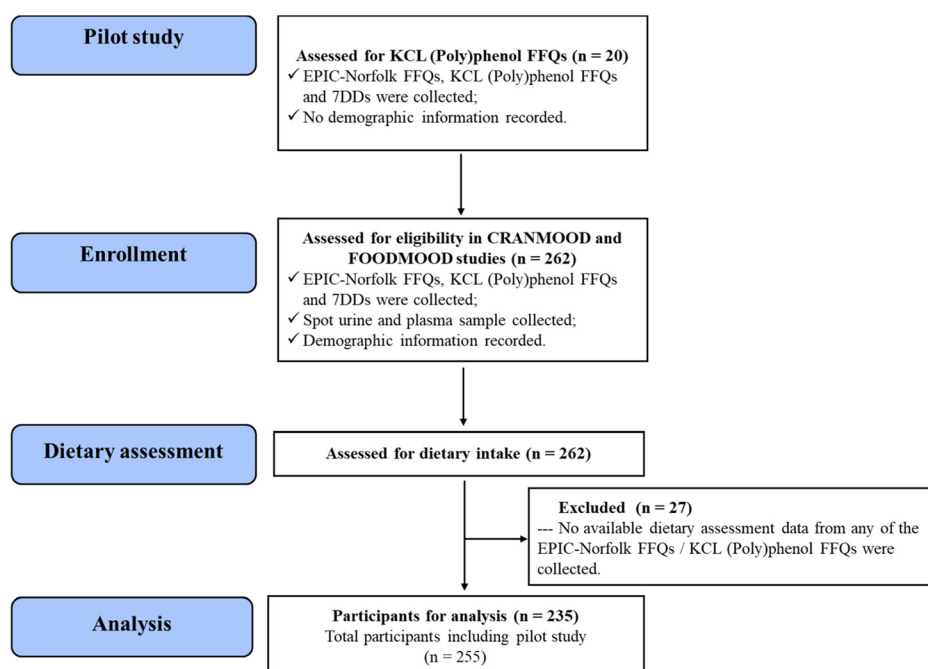


Fig. 3 Flowchart of the study. 7DDs: 7 day diet diaries, FFQs: Food Frequency Questionnaires, EPIC: European Prospective Investigation into Diet and Cancer.



EPIC-Norfolk FFQ (version 6, CAMB/PQ/6/1205)³⁰ was also collected at the baseline, to test the agreements with our newly developed FFQ for nutrients and (poly)phenol intake data. 7DDs were given to participants to record their daily consumption for seven consecutive days at baseline,³⁰ one week before the first visit. There are six-time slots in each day included in each diary, including before breakfast, breakfast, mid-morning, lunch, tea, evening meal, and later evening. Participants were required to report their daily type and amount of food and beverage consumption in detail. The instructions with standard photos of portion sizes or grams were described at the beginning pages of the paper-based diaries booklet.⁶ Furthermore, spot urine and fasting (at least 12 h) plasma samples were also collected at baseline.

2.6 Estimation of (poly)phenol intake

The EPIC-Norfolk FFQs were coded with the Microsoft Access software (Access 2019, Microsoft, USA) and transformed into daily food and nutrient intake levels by the FFQ EPIC Tool for Analysis (FETA) software.⁴¹ Nutrient composition was obtained from McCance and Widdowson's "The Composition of Foods (5th edition)" and ESI.⁴² The 7DDs were coded into standard food codes and portions by trained coders using the Nutritics software (Nutritics Research Edition v 5.76, Nutritics, Dublin, Ireland). A standard protocol was followed by all coders to minimize the coding error and improve the quality and consistency of the data. The KP-FFQs were coded following a standard protocol using Excel (Microsoft, USA) software and transformed into daily food and nutrient intake levels by Excel (Microsoft, USA) and R (version 4.1.2)⁴³ (melt() function with the "reshape2" package, version 1.44).

An in-house database was established based on the online open-access Phenol-Explorer database,³⁹ the USDA database, and several published papers^{44–66} to estimate the (poly)phenol content of each food item. Information regarding this database has been previously described.³⁵ (poly)phenol intake (mg d⁻¹) was calculated using the estimated food intake (g d⁻¹) multiplied by the corresponding (poly)phenol intake from the in-house database (mg per 100 g) and divided by 100. The classes and subclasses of (poly)phenols were extracted by adding all individual compounds within the group.

2.7 Sample collection, processing and (poly)phenol metabolite analyses

Baseline spot urine ($n = 41$) and plasma samples ($n = 36$) were collected as part of the CRANMOOD cohort. The spot urine and plasma samples were collected, stored, processed, and analysed following a validated method described in detail previously^{67,68} with ultra-high-performance liquid chromatography-triple quadrupole mass spectrometry (UHPLC-Q-q-Q MS) (TSQ Vantage, Thermo Fisher Scientific, Waltham, MA, USA). Metabolites were identified and quantified by authentic standards,²¹ and a total of 110 urinary and 89 plasma metabolites were selected in the statistical analysis. The TraceFinder software (TraceFinder 5.0, Thermo Fisher Scientific, Waltham, MA, USA) was employed for data analysis and calculation. The

urinary creatinine levels were analysed by Affinity Biomarker Labs (London, UK) using the Jaffe method, and the concentrations of the metabolites (nM) were adjusted by the creatinine levels (mg L⁻¹) into mmol g⁻¹ creatinine.

2.8 Statistical analysis

The statistical analysis in this research was performed using R (version 4.1.2).⁴³ (Poly)phenol intake was reported as mean (standard deviation, SD) and median (interquartile range, IQR). The significant levels were adjusted for multiple comparisons using the false discovery rate (FDR) method, and $p < 0.05$ was used as the significant level.

The relative validation of KP-FFQs used 7DDs, and biomarkers from spot urine and plasma samples were employed to strengthen the validity. The association and agreement between the KP-FFQs against 7DDs/biomarkers were investigated. The association was tested by linear regression with lm. beta R package (version 1.7.2) and adjusted for energy intake. To investigate the agreements in ranking participants into quartiles, weighted Kappas were calculated with the "psych" package in R. A weighted kappa value over 0.75 was considered an excellent agreement, 0.40–0.75 was considered a fair to good agreement, and lower than 0.40 was considered as a poor agreement.⁶⁹ The 95% confidence intervals (CI) were calculated for kappa. The percentages of participants grouped into the same, adjacent or opposite quartiles were also calculated to show agreements between the two methods. For comparison, the association and agreement between the EPIC-Norfolk FFQs against 7DDs/biomarkers were also explored with the above methods. Weighted kappa and cross-classification were used to assess the relative validity of KP-FFQs against 7DDs on nutrient intake.

Agreements between (poly)phenol and nutrient intake estimated from the KP-FFQs and EPIC-Norfolk FFQs were presented as two-way mixed effects intraclass correlation coefficients (ICC). The consistency (ICC-C) and agreement (ICC-A) models in ICC were calculated with the "irr" package in R. The consistency model ignores the systematic difference between the two FFQs, while the agreement model compares the absolute values of the estimated intake. The ICC values lower than 0.5 were considered poor agreement, and between 0.50–0.75 were considered moderate agreement, 0.75–0.90 good agreement, and above 0.90 excellent agreement.⁷⁰ In addition, the weighted Kappas were also employed, and Spearman's analysis was also adopted to test the correlation between these two tools. (Poly)phenol intake was further adjusted for energy intake by the residual method and calculated for the above values. The 95% CI was extracted for ICC, kappa, and Spearman's rho values.

3. Results

3.1 Characteristics of the study population

No demographic information was collected as part of the pilot study ($n = 20$). As for the demographic characteristics of the CRANMOOD and FOODMOOD studies, 235 participants were



Table 1 Demographic characteristics of the study population

Characteristics	Mean (SD)/ <i>n</i> (%)	Missingness (%)
Age (years)	22.6 (2.7)	0
Ethnicity		2.0
White	53 (22.6)	
Black	13 (5.5)	
Asian	155 (66.0)	
Mixed	9 (3.8)	
Sex		0
Male	36 (15.3)	
Female	199 (84.7)	
Physical activity level		0.9
Low	11 (4.7)	
Moderate	104 (44.3)	
High	118 (50.2)	
Smoking status		0.4
Never	208 (88.5)	
Current & ex-smoker	26 (11.1)	
Alcohol consumption		0.4
Not drinking	104 (44.3)	
≤5 unit per week	109 (46.4)	
>5 unit per week	21 (8.9)	
BMI		0.4
<25 kg m ⁻²	202 (86.0)	
≥25 kg m ⁻²	32 (13.6)	
Education level		0.9
Bachelor	87 (37.0)	
Master	130 (55.3)	
PhD	16 (6.8)	
Dietary assessment tools		
EPIC-Norfolk FFQs	225 (95.7)	4.3
KCL (poly)phenol FFQs	201 (85.5)	14.5
7DDs	65 (27.7)	72.3

7DDs: 7 day diet diaries, EPIC: European Prospective Investigation into Diet and Cancer, FFQs: food frequency questionnaires; SD: standard deviation.

included with 36 males and 199 females (Table 1). The average age of subjects was 22.6 (SD 2.7) years, and nearly half of the population were postgraduate students (55.3). The majority of subjects were from the Asian ethnic group (66.0%), with a BMI lower than 25 kg m⁻² (86.0%) and high or moderate physical activity levels (50.2% and 44.3%, respectively). Most of them reported non-smoking (88.5%) and alcohol consumption lower than five units per week (46.4%) or non-alcohol consumption (44.3%). There were 225 EPIC-Norfolk FFQs and 201 KP-FFQs collected. The collection of 7DDs was not incorporated into the design of the FOODMOOD study, therefore only 65 participants from the CRANMOOD cohort provided 7DDs.

3.2 The completion time of the KCL (poly)phenol FFQ from the pilot study

The pilot study (*n* = 20) found that the average time to complete EPIC-Norfolk FFQs and KP-FFQs was 13.9 ± 6.8 and 36.9 ± 21.4 min, respectively. No adjustments to the questionnaire were made as a result of the pilot study.

3.3 (Poly)phenol intake estimated from the KCL (poly)phenol FFQ

The total (poly)phenol intake from KP-FFQ stratified by baseline characteristics is listed in Table 2. The mean (SD) and

median (IQR) of total (poly)phenol intake estimated from the KP-FFQ was 1366.5 (1151.7) and 1073.2 (1022.1) mg d⁻¹, respectively in Table 3. Regarding the contribution of different (poly)phenol subclasses (Table 3), flavonoids were the main type of dietary (poly)phenols (47.1%), followed by phenolic acids (45.5%). As for food sources (Table 4), non-alcoholic beverages were the main food sources of total (poly)phenols (42.0%), followed by fruits (9.7%) (apple 5.8%, blueberries 2.4%, strawberries 1.5%), and Chocolate (5.3%) (drinking chocolate powder 3.8%, dark chocolate 1.5%). As for the individual foods, coffee contributed the most to the total (poly)phenol intake (29.4%), followed by tea (12.6%). Multiple subtypes of tea and coffee in the KP-FFQ contributed to the total (poly)phenol intake, including black tea (10.1%) (English breakfast tea (4.4%), Assam (3.2%), Earl grey (2.5%)), green tea (2.5%) in tea category and decaffeinated (10.5%), infusion (9.4%), espresso (6.2%), filtered (3.3%) in coffee category. The average coffee intake was 1.1 ± 0.4 cup per d (203.6 ± 78.6 g d⁻¹) from KP-FFQ, whereas the average tea intake was 0.8 ± 0.2 cup per d (147.0 ± 28.6) g d⁻¹ (standardized as 190 g per cup for both coffee and tea according to default portion size in the EPIC-Norfolk FFQ). The KP-FFQ lists 442 items, and 312 of them were not listed in the EPIC-Norfolk FFQ. The food/beverages unique to the KP-FFQ contributed 72.7% of the total (poly)phenol intake estimated from the KP-FFQs (74.6% of flavonoids, 67.7% phenolic acids, 87.5% stilbenes, 72.2% lignans, and 93.3% of the other (poly)phenols intake).

3.4 Relative validity with 7DDs estimated (poly)phenol and nutrients intake

3.4.1 (Poly)phenol intake. Fig. 4 shows the associations between (poly)phenol intake from 7DDs and KP-FFQs adjusted by energy intake. KP-FFQs estimated intake, including total (poly)phenol, dihydroflavonols, flavan-3-ol monomers, total phenolic acids, hydroxycinnamic acids, hydroxyphenylacetic acids, total stilbenes, resveratrol, and total lignan intake were all positively associated with the total (poly)phenol from 7DDs with standard beta ranging from 0.52 (95% CI: 0.30 to 0.74) for total stilbenes to 0.32 (95% CI: 0.08 to 0.56) for flavan-3-ol monomers (all FDR adjusted *p* < 0.05). Strong associations were found in ten (poly)phenols estimated from 7DDs, including dihydroflavonols, theaflavins, thearubigins, flavones, isoflavonoids, ellagitannins, hydroxyphenylacetic acids, total stilbenes, resveratrol, and tyrosols with stdBeta ranged from 0.61 (95% CI: 0.42 to 0.81) between thearubigins from 7DDs and flavan-3-ol monomers from KP-FFQs) to 0.95 (95% CI: 0.86 to 1.03 between dihydroflavonols from 7DDs and KP-FFQs) (all FDR adjusted *p* < 0.01).

The agreements between KP-FFQs and 7DDs in ranking participants in quartiles of (poly)phenol levels are shown in Table 5. Fair agreement was found in ten (poly)phenol groups, including total (poly)phenol intake (kappa: 0.45, 95% CI: 0.25–0.66), chalcones (kappa: 0.41, 95% CI: 0.18–0.65), isoflavonoids (kappa: 0.48, 95% CI: 0.29–0.67), total phenolic acids (kappa: 0.73, 95% CI: 0.64–0.83), hydroxycinnamic acids (kappa: 0.73, 95% CI: 0.64–0.83), hydroxyphenylacetic acids (kappa: 0.52, 95% CI: 0.31–0.73), resveratrol (kappa: 0.47, 95%



Table 2 Total (poly)phenol intake from KCL (poly)phenol FFQs stratified by baseline characteristics (mg d⁻¹)

Characteristics		N	Mean (SD)	Median (IQR)
Sex	Men	31	1340.3 (1555.2)	944.8 (922.1)
	Women	170	1398.9 (1103.6)	1085.8 (1073.2)
Age group	18–22	97	1532.8 (1429.0)	1121.3 (1117.6)
	23–29	104	1256.5 (878.2)	1065.3 (978.4)
Ethnicity	White	43	1594.2 (1384.7)	1139.0 (1092.3)
	Black	11	1062.8 (672.0)	842.6 (788.7)
	Asian	138	1339.3 (1140.8)	1057.4 (1063.9)
	Mixed	9	1587.6 (1127.0)	1274.4 (659.9)
BMI	<25 kg m ⁻²	174	1333.8 (1043.8)	1071.3 (1054.7)
	≥25 kg m ⁻²	27	1750.8 (1810.1)	1137.2 (1041.6)
IPAQ	Low	10	938.2 (723.5)	745.2 (575.5)
	Moderate	91	1146.3 (828.5)	997.3 (873.9)
	High	100	1656.6 (1413.8)	1266.1 (1129.1)
Smoking	Never	180	1359.3 (1208.3)	1048.0 (1025.1)
	Current & Ex-smoker	21	1651.5 (918.5)	1347.7 (1335.1)
Alcohol consumption	Not drinking	96	1328.4 (1211.9)	981.3 (1104.2)
	≤5 unit per week	87	1401.8 (1193.5)	1096.2 (898.1)
	>5 unit per week	18	1659.6 (928.7)	1457.2 (1478.8)
Education level	Bachelor	79	1531.6 (1562.4)	1042.2 (1031.5)
	Master	106	1248.3 (767.2)	1088.2 (1050.8)
	PhD	16	1627.4 (1179.7)	1213.5 (899.3)

FFQs: food frequency questionnaires, BMI: body mass index, IPAQ: International Physical Activity Questionnaire, IQR: interquartile range, SD: standard deviation. N: number of participants in each group.

Table 3 (Poly)phenol intake classes and subclasses estimated from KCL (poly)phenol FFQs (mg d⁻¹)

(Poly)phenols (mg d ⁻¹)	Mean (SD)	Median (IQR)	%
Total (poly)phenols	1366.5 (1151.7)	1073.2 (1022.1)	100.0
Total flavonoids	643.0 (517.6)	496.3 (464.6)	47.1
Anthocyanins	53.4 (71.5)	37.5 (41.3)	3.9
Chalcones	0.0 (0.0)	0.0 (0.0)	0.0
Dihydroflavonols	0.2 (0.4)	0.0 (0.3)	0.0
Dihydrochalcones	2.9 (3.7)	1.9 (3.3)	0.2
Total flavan-3-ols	445.0 (400.0)	315.6 (408.2)	32.6
Flavan-3-ol monomers	91.4 (104.6)	56.5 (87.7)	6.7
Theaflavins	9.8 (20.2)	1.7 (10.1)	0.7
Thearubigins	64.3 (132.3)	11.2 (66.9)	4.7
Proanthocyanidins	279.6 (228.3)	220.3 (231.3)	20.5
Flavanones	60.1 (77.0)	39.1 (57.5)	4.4
Flavones	10.0 (11.3)	6.8 (7.3)	0.7
Flavonols	59.9 (49.2)	50.2 (38.0)	4.4
Isoflavonoids	11.6 (22.6)	4.7 (11.8)	0.8
Total phenolic acids	622.4 (631.2)	418.5 (632.3)	45.5
Hydroxybenzoic acids	58.8 (69.0)	40.4 (43.2)	4.3
Ellagitannins	2.4 (9.8)	0.0 (1.3)	0.2
Hydroxycinnamic acids	563.5 (602.3)	378.0 (575.9)	41.2
Hydroxyphenylacetic acids	0.0 (0.1)	0.0 (0.0)	0.0
Total stilbenes	0.3 (0.4)	0.2 (0.4)	0.0
Resveratrol	0.2 (0.2)	0.2 (0.2)	0.0
Total lignans	4.8 (7.6)	2.4 (3.1)	0.4
Other (poly)phenols	96.0 (221.4)	40.8 (60.8)	7.0
Tyrosols	6.4 (6.1)	5.2 (7.4)	0.5
Alkylmethoxyphenols	2.6 (3.2)	1.8 (2.8)	0.2
Alkylphenols	16.0 (24.5)	8.8 (14.8)	1.2

FFQs: food frequency questionnaires, IQR: inter-quartile range, SD: standard deviation. %: percentage of contribution to the total (poly)phenol intake.

CI: 0.26–0.67), alkylmethoxyphenols (kappa: 0.45, 95% CI: 0.24–0.66), and tyrosols (kappa: 0.49, 95% CI: 0.29–0.70). The agreements of the rest (poly)phenols were poor.

3.4.2 Nutrient intake. Poor agreements were observed between nutrient intake from KP-FFQs and 7DDs in Table 6, except for fibre (kappa: 0.40, 95% CI: 0.19–0.61), which showed moderate reliability. A similar agreement result was shown with energy intake adjustment (ESI Table 2†).

3.5 Validity against objective (poly)phenol metabolites from urine and plasma

Fig. 5 shows the association between (poly)phenol intakes estimated from KP-FFQs and objective (poly)phenol metabolites from urine (Fig. 5a) and plasma (Fig. 5b). (Poly)phenol intake from KP-FFQs showed positive associations with 76 subgroup and individual urinary metabolites with standard beta ranging from 0.28 (95% CI: 0.07–0.49) between urinary 3-methoxybenzoic acid-4-sulfate and total flavonoids intake to 0.81 (95% CI: 0.62–1.00) between urinary (–)-epicatechin-3'-sulfate and ellagitannins intake (all FDR adjusted *p* values < 0.05) (Fig. 5a). As for plasma metabolites, KP-FFQs estimated intake showed positive associations with 19 subgroup and individual metabolites with standard beta ranging from 0.40 (95% CI: 0.17–0.62, between 3-hydroxybenzoic acid and flavones intake) to 0.83 (95% CI: 0.64–1.02, between (–)-epicatechin and total flavan-3-ols from plasma and ellagitannins intake estimated from FFQs) (all FDR adjusted *p* values < 0.05) (Fig. 5b).

The agreements between dietary assessment and metabolites in ranking participants in quartiles are shown in Table 7. Poor agreements were seen for all groups of (poly)phenols, including total (poly)phenols, total flavonoids, total lignans, total stilbenes, and total other (poly)phenols between



Table 4 Food sources of the (poly)phenol from KCL (poly)phenol FFQs

(Poly)phenols (mg d ⁻¹)	(Poly)phenol FFQs estimated (poly)phenol food sources (% to total)
Total (poly)phenols	Coffee (decaffeinated 10.5%, infusion 9.4%, espresso 6.2%, filtered 3.3%) (29.4%), black tea (English breakfast tea 4.4%, Assam 3.2%, Earl grey 2.5%) (10.1%), apple (red & green) (5.8%), chocolate (drinking chocolate powder 3.8%, dark chocolate 1.5%) (5.3%), cloves (3.0%), green tea (2.5%), blueberries (2.4%), white rice (2.0%), strawberries (1.5%), orange juice (1.5%)
Total flavonoids	Black tea (English breakfast tea 8.4%, Assam 6.1%, Earl grey 4.7%) (19.2%), apple (red & green) (11.0%), chocolate (drinking chocolate powder 7.9%, dark chocolate 3.0%) (10.9%), green tea (4.5%), citrus (orange 2.2%, mandarins 1.5%) (3.7%), blueberries (3.4%), hazelnut milk (2.9%), orange juice (2.8%), strawberries (2.6%), tomatoes (2.1%), spinach (2.0%), grapes (1.6%)
Anthocyanins	Blueberries (13.2%), cherries (12.4%), Cabbage (purple & red) (20.5%), aubergine (8.5%), grapes (7.2%), strawberries (7.2%), blackberries (3.9%), blackcurrants (3.0%), black beans (3.0%)
Chalcones	Broad beans (70.2%), Ale, beer (23.0%), regular, beer (6.8%)
Dihydroflavonols	Wine (red 84.6%, white 7.6%, rose 4.2%), moussaka (3.5%)
Dihydrochalcones	Apple (red, flesh and skin 41.0%, flesh only 16.9%, green flesh and skin 14.5%, flesh only 9.5%), apple juice (concentrate 9.6%, pure juice 7.9%), apple chutney (0.5%), pomegranate juice (0.1%)
Total flavan-3-ols	Black tea (English breakfast tea 11.9%, Assam 8.7%, Earl grey 6.7%) (27.3%), apple (red & green) (14.8%), drinking chocolate powder (11.4%), green tea (6.3%), chocolate (4.3%), Hazelnut milk (4.2%), blueberries (3.1%), strawberries (2.8%), almond (2.1%), hazelnuts (2.1%)
Flavan-3-ol monomers	Black tea (English breakfast tea 18.5%, Assam 13.6%, Earl grey 10.4%) (42.5%), green tea (27.7%), apple (red & green) (6.2%), chocolate (drinking chocolate powder 4.4%, dark chocolate 1.2%) (5.6%), broad beans (2.5%), mint tea (1.6%), grape (1.1%)
Theaflavins	Black tea (English breakfast tea 43.5%, Assam 32.0%, Earl grey 24.6%) (100%)
Thearubigins	Black tea (English breakfast tea 43.2%, Assam 31.8%, Earl grey 24.4%) (99.4%), green tea (0.6%)
Proanthocyanidins	Chocolate (drinking chocolate powder 16.8%, dark chocolate 6.5%) (23.3%), apple (red & green) (21.6%), hazelnut milk (6.7%), black tea (English breakfast tea 3.4%, Assam 1.0%, Earl grey 0.8%) (5.2%), blueberries (5.0%), strawberries (4.3%), grapes (green & black) (4.2%), hazelnuts (3.3%), almond (3.2%)
Flavanones	Citrus (orange 23.9%, mandarins 15.2%, lemons 11.1%, limes 3.3%, grapefruit 2.7%) (56.2%), citrus juice (orange juice 28.7%, grapefruits juice 1.6%, blood orange juice 1.5%, lemon juice 0.7%) (32.5%), mints (8.1%), tomatoes (cherry 1.1%, raw 0.4%, ketchup 0.1%) (1.6%)
Flavones	Parsley (11.0%), mint (8.7%), tortilla (wholemeal flour & wheat flour) (9.8%), orange juice (9.0%), bagel (plain & Wholemeal) (9.5%), citrus (mandarins 3.3%, lemon 2.4%, blood orange 0.8%) (6.5%), pizza (cheese and tomatoes) (4.6%), croissant (butter, chocolate, almond) (4.3%), spinach (1.8%), celery (1.6%)
Flavonols	Tomatoes (raw 22.0%, ketchup 6.6%, soup 4.6%, cherry 1.3%) (34.5%), spinach (21.7%), onion (red 3.2%, yellow 2.1%, white 0.7%) (6.0%), black tea (English breakfast tea 1.6%, Assam 1.2%, Earl grey 0.9%) (3.7%), broccoli (3.1%), vegetable soup (2.0%), lettuce (red & green) (2.1%), green tea (1.9%), blueberries (1.6%)
Isoflavonoids	Soy milk (32.2%), tofu (16.1%), edamame bean (15.8%), beansprouts (8.9%), soy based (Greek style & low-fat yoghurt) (11.2%), tempeh (6.6%), soya mince (4.2%)
Total phenolic acids	Coffee (infusion 20.5%, decaffeinated, espresso based 16.3%, espresso 13.5%, filtered 7.2%, decaffeinated, instant or ground 6.5%, decaffeinated, filtered 3.5%) (67.5%), white & brown rice (5.1%), black tea (English breakfast tea 1.0%, Assam 0.8%, Earl grey 0.6%) (2.4%), chestnut (2.1%), blueberries (1.8%)
Hydroxybenzoic acids	Chestnuts (22.6%), black tea (English breakfast tea 9.4%, Assam 6.9%, Earl grey 5.3%) (21.6%), green tea (6.5%), strawberries (6.7%), garlic (6.4%), white rice (3.8%), pomegranate juice (concentrate 3.5% and pure juice 1.4%) (4.9%), clove (2.1%)
Ellagitannins	Pomegranate juice (72.1%), low fat yoghurt, raspberry (18.6%), full fat yoghurt, raspberry (5.5%), yoghurt drinks, raspberry (3.9%)
Hydroxycinnamic acids	Coffee (infusion 22.6%, decaffeinated, espresso based 18.0%, espresso 14.9%, filtered 8.0%, decaffeinated, instant or ground 7.2%, decaffeinated, filtered 3.9%) (74.6%), white & brown rice (5.3%), apple (red & green) (2.0%), blueberries (1.9%)
Hydroxyphenylacetic acids	Regular beer (49.3%), wine (red 20.4%, white 10.5%) (30.9%), olive oil (extra virgin 9.6%, virgin 4.2%, refined 3.0%) (16.8%)
Total stilbenes	Wine (red 35.5%, white 5.2%, rose 2.4%) (43.1%), citrus (mandarins 16.8%, lemon 12.4%) (29.2%), strawberries (9.1%), grape (black 4.5%, green 4.1%) (8.6%), low fat (mixed berries 1.0%, peach 0.5%, mango 0.4%) (1.9%), moussaka (1.5%)
Resveratrol	Citrus (mandarins 24.7%, lemon 18.1%) (42.8%), wine (red 12.0%, rose 3.2%, white 3.1%) (18.3%), strawberries (13.5%), grape (black 6.4%, green 6.1%) (12.5%), low fat (mixed berries 1.5%, peach 0.7%, mango 0.6%) (2.8%), redcurrant (1.6%)
Total lignans	Flaxseed (33.8%), sesame seeds (13.3%), bread, seeded (10.1%), potatoes (boiled 9.0%, roast 4.1%, crisps 1.7%) (14.8%), broccoli (5.0%)
Other (poly)phenols	Cloves (41.4%), turmeric (20.6%), olive oil (extra virgin 3.1%, virgin 1.4%, refined 1.0%) (5.5%), break cereal (5.0%), star anise (4.8%), bread (wholemeal 1.9%, rye 1.6%, pitta 0.8%) (4.3%), coffee (decaffeinated 1.4%, infusion 1.1%, espresso 0.7%, filtered 0.4%) (3.6%), spaghetti (wholemeal & white) (3.2%), curry powder (2.5%)
Tyrosols	Olive oil (extra virgin 47.2%, virgin 20.5%, refined 14.8%) (82.5%), pesto (green & red) (6.8%), red wine (2.4%), pizza, pesto (2.2%)
Alkylmethoxyphenols	Coffee (decaffeinated, espresso based 27.0%, infusion 17.2%, espresso 11.3%, filtered 6.1%, decaffeinated, instant or ground 10.7%, decaffeinated, filtered 5.9%) (78.2%), rapeseed oil (18.3%)
Alkylphenols	Breakfast cereal (30.0%), bread (wholemeal 11.6%, rye 9.3%, pitta 4.9%) (25.8%), spaghetti (wholemeal & white) (19.2%), tortilla (wholemeal & wheat flour) (8.9%), bagel (wholemeal & plain) (5.7%), pizza (pesto 0.8%, cheese and tomato 0.6%, vegetarian 0.2%) (1.6%)



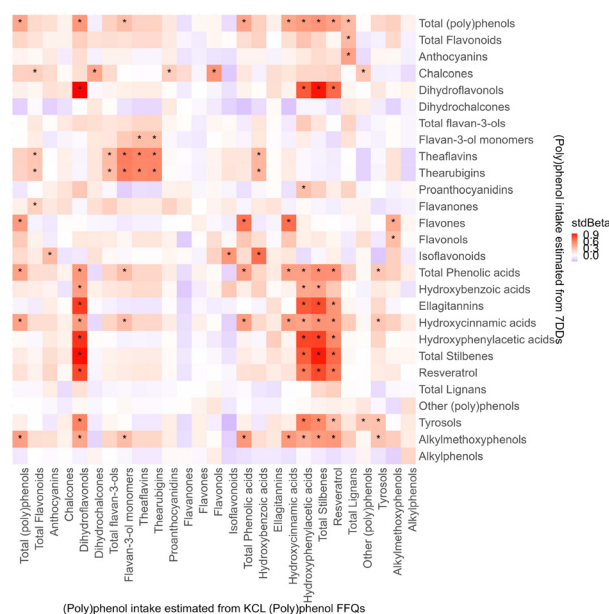


Fig. 4 Association between estimated (poly)phenol intakes from 7DDs and intakes from KCL (poly)phenol FFQs adjusted for energy intake. 7DDs: 7 day diet diaries, FFQs: food frequency questionnaires. The heatmap was plotted according to the standardized regression coefficients (stdBeta). The colour scale indicates the effect (stdBeta) of association between 7DDs and KCL (poly)phenol FFQs estimated (poly)phenol intake level. Red and blue illustrate positive and negative effects, and colour intensity represents the degree of effect. The asterisks showed significance (*: $\text{fdr-adjusted } p < 0.05$). Energy intakes were adjusted for using the residual method.

biomarkers in urine and plasma samples and KP-FFQ ($\kappa < 0.40$). Regarding the agreement between total (poly)phenol metabolite levels and the total estimated (poly)phenol intake, metabolites from plasma ($\kappa = 0.29$) showed a slightly better agreement than urinary metabolites ($\kappa = 0.16$) with KP-FFQ, although all these agreements were poor.

3.6 Comparison between KCL (poly)phenol FFQs and EPIC-Norfolk FFQs

3.6.1 (Poly)phenol intake. EPIC-Norfolk FFQs estimated (poly)phenol and food sources are listed in ESI Table 3.† The median (IQR) of total (poly)phenol was 741.2 (844.2) mg d^{-1} , and phenolic acids were the major (poly)phenols (53.6%), followed by flavonoids (44.6%). Regarding the individual food source contribution, coffee contributed the most (41.0% , 0.8 ± 1.0 cup per d ($145.3 \pm 187.1 \text{ g d}^{-1}$)), followed by tea (23.6% , 0.7 ± 1.1 cup per d ($129.8 \pm 200.0 \text{ g d}^{-1}$)) (default portion size: 190 g per cup for coffee and tea).

There was moderate reliability between EPIC-Norfolk and KP-FFQs estimated total (poly)phenol in absolute values (ICC-A: 0.54, 95% CI: 0.33–0.68). As for classes and subclass intakes, strong agreement was found between flavonols (ICC-A: 0.77, 95%CI: 0.63, 0.84), and moderate agreements were found between total flavonoids, dihydrochalcones, flavan-3-ols, flavan-3-ol monomers, theaflavins, thearubigins, isoflavonoids, total phenolic acids, hydroxycinnamic acids, and hydroxyphenylacetic acids with ICC-A ranging from 0.55 (iso-

Table 5 Agreements between KCL (poly)phenol FFQs and 7DDs in the estimated (poly)phenol intake

(Poly)phenols	Kappa	(95% CI)	Same quartile (%)	Adjacent quartile (%)	Correctly classified ^a (%)	Opposite quartile (%)
Total (poly)phenols	0.45	(0.25, 0.66)	33.33	51.67	85.00	5.00
Total flavonoids	0.08	(−0.19, 0.35)	36.67	30.00	66.67	13.33
Anthocyanins	0.16	(−0.08, 0.40)	21.67	48.33	70.00	8.33
Chalcones	0.41	(0.18, 0.65)	46.67	33.33	80.00	6.67
Dihydroflavonols	0.33	(0.06, 0.60)	48.33	30.00	78.33	10.00
Dihydrochalcones	0.31	(0.10, 0.52)	28.33	43.33	71.66	3.33
Total flavan-3-ols	0.09	(−0.15, 0.34)	25.00	38.33	63.33	8.33
Flavan-3-ol monomers	0.24	(0.01, 0.47)	25.00	45.00	70.00	5.00
Theaflavins	0.27	(0.00, 0.54)	41.67	33.33	75.00	10.00
Thearubigins	0.24	(−0.03, 0.51)	41.67	36.67	78.34	13.33
Proanthocyanidins	0.09	(−0.16, 0.34)	23.33	43.33	66.66	10.00
Flavanones	0.19	(−0.06, 0.43)	30.00	36.67	66.67	6.67
Flavones	0.11	(−0.15, 0.36)	30.00	38.33	68.33	11.67
Flavonols	0.31	(0.07, 0.54)	30.00	46.67	76.67	6.67
Isoflavonoids	0.48	(0.29, 0.67)	43.33	35.00	78.33	1.67
Total phenolic acids	0.73	(0.64, 0.83)	43.33	53.33	96.66	0.00
Hydroxybenzoic acids	0.16	(−0.08, 0.40)	28.33	36.67	65.00	6.67
Ellagitannins	0.41	(0.17, 0.66)	43.33	43.33	86.66	10.00
Hydroxycinnamic acids	0.73	(0.64, 0.83)	43.33	53.33	96.66	0.00
Hydroxyphenylacetic acids	0.52	(0.31, 0.73)	51.67	30.00	81.67	3.33
Total stilbenes	0.39	(0.17, 0.60)	38.33	36.67	75.00	3.33
Resveratrol	0.47	(0.26, 0.67)	50.00	25.00	75.00	1.67
Total lignans	0.37	(0.19, 0.56)	26.67	48.33	75.00	1.67
Other (poly)phenols	0.37	(0.16, 0.59)	35.00	40.00	75.00	3.33
Tyrosols	0.49	(0.29, 0.70)	38.33	48.33	86.66	5.00
Alkylmethoxyphenols	0.45	(0.24, 0.66)	38.33	45.00	83.33	5.00
Alkylphenols	0.37	(0.16, 0.59)	33.33	45.00	78.33	5.00

^a Correctly classified (%): correctly classified the (poly)phenols into the same or adjacent quartiles (%). FFQs: food frequency questionnaires, 7DDs: 7 day diet diaries, kappa: weighted kappa coefficient (linear weights). CI: confidence interval.



Table 6 Agreements between KCL (poly)phenol FFQs estimated nutrient intake and 7DDs

Nutrients	Kappa	(95% CI)	Same quartile (%)	Adjacent quartile (%)	Correctly classified ^a (%)	Opposite quartile (%)
Energy (kcal)	−0.01	(−0.27, 0.24)	20.00	41.67	61.67	11.67
Fibre (g d ^{−1})	0.40	(0.19, 0.61)	36.67	40.00	76.67	3.33
Calcium (mg d ^{−1})	0.13	(−0.08, 0.35)	20.00	40.00	60.00	3.33
Iron (mg d ^{−1})	0.27	(0.03, 0.5)	30.00	43.33	73.33	6.67
Potassium (mg d ^{−1})	0.19	(−0.04, 0.42)	23.33	48.33	71.66	8.33
Retinol (μg d ^{−1})	0.16	(−0.06, 0.38)	20.00	45.00	65.00	5.00
Carotene (μg d ^{−1})	0.28	(0.03, 0.53)	41.67	31.67	73.34	8.33
Vitamin C (mg d ^{−1})	0.23	(0.01, 0.44)	21.67	48.33	70.00	5.00
Fat (g d ^{−1})	0.08	(−0.15, 0.31)	23.33	36.67	60.00	6.67
Cholesterol (mg d ^{−1})	−0.12	(−0.37, 0.13)	20.00	38.33	58.33	15.00
MUFA (g d ^{−1})	0.11	(−0.15, 0.36)	26.67	40.00	66.67	10.00
PUFA (g d ^{−1})	0.09	(−0.14, 0.33)	18.33	50.00	68.33	10.00
SFA (g d ^{−1})	0.12	(−0.10, 0.34)	20.00	41.67	61.67	5.00
Protein (g d ^{−1})	−0.01	(−0.28, 0.25)	33.33	26.67	60.00	13.33
Total carbohydrate (g d ^{−1})	0.07	(−0.16, 0.29)	16.67	41.67	58.34	5.00
Sugars (g d ^{−1})	0.16	(−0.06, 0.38)	21.67	40.00	61.67	3.33
Fructose (g d ^{−1})	−0.01	(−0.27, 0.24)	26.67	30.00	56.67	10.00
Galactose (g d ^{−1})	0.24	(0.00, 0.48)	26.67	48.33	75.00	8.33
Glucose (g d ^{−1})	0.05	(−0.20, 0.30)	23.33	40.00	63.33	10.00
Starch (g d ^{−1})	0.29	(0.05, 0.54)	30.00	48.33	78.33	8.33
Sucrose (g d ^{−1})	0.16	(−0.06, 0.38)	25.00	38.33	63.33	5.00
Lactose (g d ^{−1})	0.29	(0.06, 0.53)	31.67	43.33	75.00	6.67
Maltose (g d ^{−1})	0.32	(0.09, 0.55)	35.00	38.33	73.33	5.00

^a Correctly classified (%): correctly classified the (poly)phenols into the same or adjacent quartiles (%). FFQs: food frequency questionnaires, 7DDs: 7 day diet diaries, kappa: weighted kappa coefficient (linear weights). CI: confidence interval.

flavonoids 95% CI: 0.41–0.66) to 0.69 (hydroxyphenylacetic acids 0.56–0.78) (Table 8). In the ability to rank participants according to (poly)phenol intake levels, the reliabilities between flavonols exhibited high reliability (ICC-C: 0.79, 95% CI: 0.73–0.84). Moderate reliability was exhibited in total (poly)phenols, total flavonoids, dihydrochalcones, flavan-3-ols, flavan-3-ol monomers, theaflavins, thearubigins, proanthocyanidins, isoflavonoids, total phenolic acids, hydroxycinnamic acids and hydroxyphenylacetic acids with ICC-C ranging from 0.53 (proanthocyanidins, 95% CI: 0.39–0.64) to 0.71 (hydroxyphenylacetic acids, 0.62–0.78). When sorting participants into quartiles by intake, a poor agreement between the FFQs was exhibited, including anthocyanins, chalcones, flavanones, flavones, ellagitannins, other (poly)phenol, tyrosols, and alkylphenols with kappa ranging from 0.04 (ellagitannins, −0.09, 0.17) to 0.39 (anthocyanins and chalcones, 0.27–0.51), whereas the agreements between the total (poly)phenol (kappa: 0.54, 0.44–0.64) and all other classes and subclasses were fair (kappa: from 0.42 (resveratrol, 0.30–0.54) to 0.65 (hydroxycinnamic acids, 0.57–0.73)) (Table 8). After adjusting for energy intake, all the agreements were increased to moderate for ICC-A and ICC-C among each group of (poly)phenols (ESI Table 4†).

3.6.2 Agreements with 7DDs in comparison with KCL (poly)phenol FFQ. Regarding EPIC-Norfolk FFQs estimated (poly)phenols in ESI Fig. 1,† only other (poly)phenols and alkylphenols were found significantly positively associated with alkylphenols from 7DDs (stdBeta: 0.63, 95% CI: (0.43, 0.83) and 0.69 (0.50, 0.88), respectively, FDR adjusted $p < 0.01$). Compared with the agreements between KP-FFQ and 7DDs,

the kappa results were lower between (poly)phenols estimated by EPIC-Norfolk FFQs and 7DDs. In ESI Table 5,† poor agreements were seen for most groups, except for dihydroflavonols, dihydrochalcones, total phenolic acids, hydroxycinnamic acids, and alkylmethoxyphenols, which had moderate agreements (kappa: 0.41–0.48). The agreements were especially low between total flavonoids, anthocyanins, total flavan-3-ols, flavanones, flavones, total other (poly)phenols, and alkylphenols, with kappa ranging from −0.16 (−0.41–0.09) to −0.05 (−0.31–0.20).

3.6.3 Agreements with biomarkers in comparison with KCL (poly)phenol FFQ. EPIC-Norfolk FFQs estimated (poly)phenol intake showed positive associations with 100 individual and subgroup of urinary metabolites with standard beta ranging from 0.27 (95% CI: 0.09–0.45) between 3'-hydroxycinnamic acid and proanthocyanidins intake to 0.65 (95% CI: 0.41–0.88) between 3-(3'-methoxyphenyl)propanoic acid-4'-sulfate and alkylphenols intake (all FDR adjusted p values < 0.05) (ESI Fig. 2a†). ESI Fig. 2b† shows the association between intakes from EPIC-Norfolk FFQs and (poly)phenol metabolites from the plasma. Only 5 positive associations were found, including hydroxyphenylacetic acids from EPIC-Norfolk FFQs and (−)-epicatechin and total flavan-3-ols from plasma (0.60 (95% CI: 0.33–0.87), FDR adjusted p values < 0.05). Similar associations were also found between 3-(4'-methoxyphenyl)propanoic acid-3'-sulfate from plasma and EPIC-Norfolk FFQs estimated flavones, alkylphenols, and other (poly)phenols intake (all FDR adjusted p values < 0.05).

Regarding the agreements in differentiating participants into quartiles in both specimens, there were 10 out of



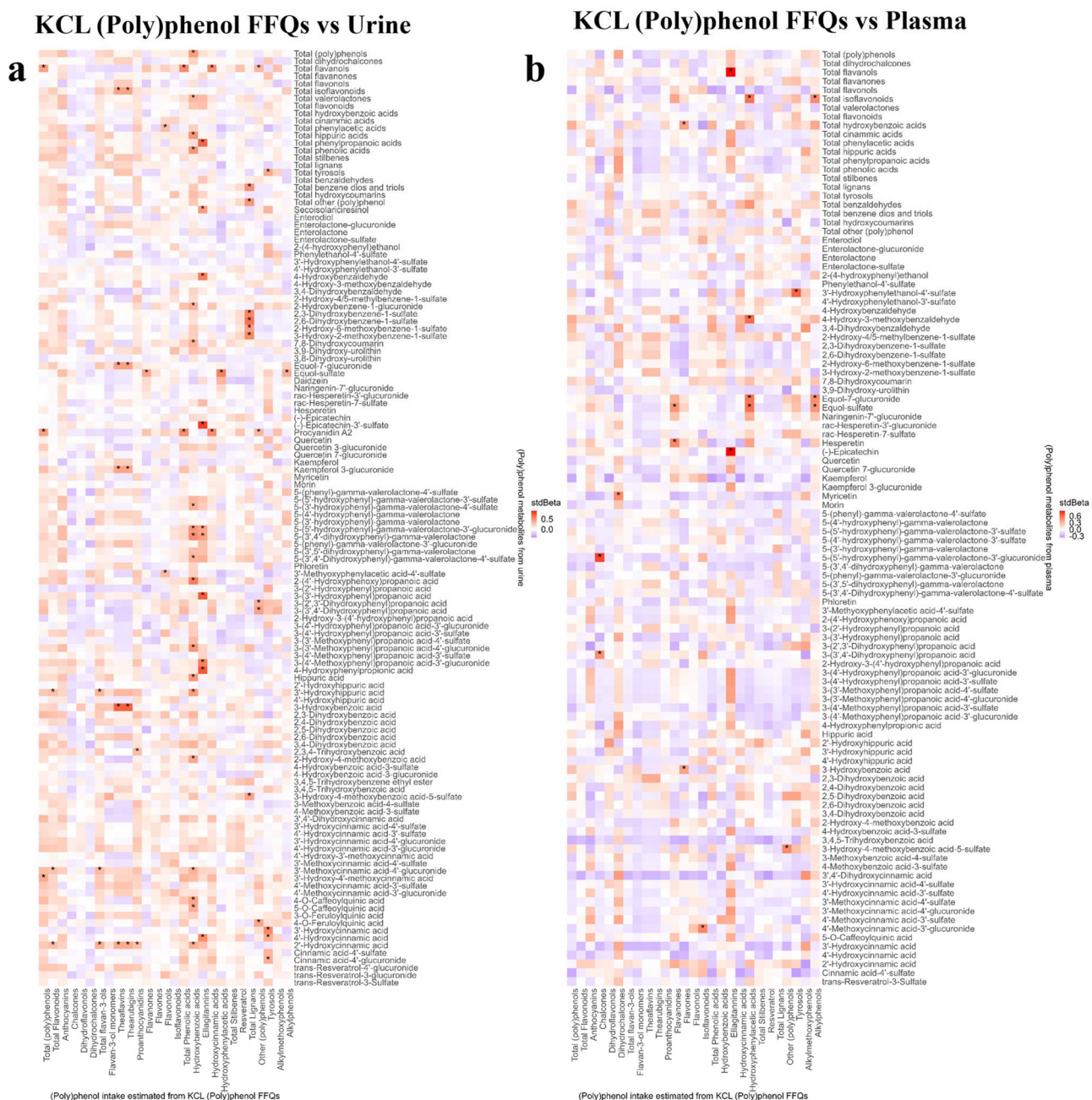


Fig. 5 Association between (poly)phenol intakes from KCL (poly)phenol FFQs and (poly)phenol metabolites from spot urine (a) and plasma sample (b) adjusted for energy intake. FFQs: food frequency questionnaires. The heatmap was plotted according to the standardized regression coefficients (stdBeta). The colour scale indicates the effect (stdBeta) of association between (poly)phenol metabolites and KCL (poly)phenol FFQs estimated (poly)phenol intake level. Red and blue illustrate positive and negative effects, and colour intensity represents the degree of effect. The asterisks showed significance (*: *fdr*-adjusted $p < 0.05$).

15 groups of (poly)phenol intake estimated from EPIC-Norfolk FFQ, which exhibited a lower correctly classified percentage (ranking subjects into the same or adjacent quartile) than the KP-FFQ estimated (poly)phenol intake groups (ESI Table 6†).

3.6.4 Nutrient intake. The agreements between the EPIC-Norfolk and KP-FFQs estimated nutrient intake are listed in Table 9, with a majority showing moderate reliability of nutrients (17 and 19 out of 23 nutrients for ICC-A and ICC-C,

respectively). Strong reliability was shown in the agreement of glucose intake (ICC-C: 0.76, 95% CI: 0.51–0.82). When sorting participants into quartiles by intake, poor agreement was seen in calcium, retinol, galactose, lactose, and maltose with kappa from 0.32 (maltose, 95% CI: 0.20–0.45) to 0.39 (retinol 0.27–0.51) between the EPIC-Norfolk and KP-FFQs. Fair agreements were exhibited for the rest of nutrients (kappa: 0.48, 95% CI: 0.37–0.59 to 0.63, 0.55–0.71) (Table 9).

Table 7 Agreements between KCL (poly)phenol FFQs estimated (poly)phenol intake and biomarkers

Metabolite levels	Groups	Kappa	(95% CI)	Same quartile (%)	Adjacent quartile (%)	Correctly classified ^a (%)	Opposite quartile (%)
Urine sample	Total (poly)phenols	0.16	(−0.14, 0.46)	26.83	46.34	73.17	12.20
	Total flavonoids	0.06	(−0.25, 0.37)	29.27	39.02	68.29	14.63
	Total dihydrochalcones	0.12	(−0.20, 0.44)	31.71	36.59	68.29	12.20
	Total flavan-3-ols	−0.21	(−0.51, 0.10)	19.51	41.46	60.98	21.95
	Total flavanones	−0.02	(−0.33, 0.30)	31.71	29.27	60.98	14.63
	Total flavonols	0.04	(−0.26, 0.34)	19.51	46.34	65.85	12.20
	Total isoflavonoids	−0.05	(−0.38, 0.27)	34.15	26.83	60.98	17.07
	Total phenolic acids	0.23	(−0.04, 0.51)	29.27	41.46	70.73	7.32
	Total hydroxybenzoic acids	−0.03	(−0.34, 0.27)	26.83	34.15	60.98	14.63
	Total cinnamic acids	0.06	(−0.26, 0.38)	26.83	46.34	73.17	17.07
	Total phenylacetic acids	−0.11	(−0.42, 0.20)	31.71	29.27	60.98	19.51
	Total stilbenes	0.08	(−0.24, 0.40)	34.15	34.15	68.29	14.63
	Total lignans	0.04	(−0.24, 0.32)	24.39	31.71	56.10	7.32
	Total other (poly)phenol	−0.17	(−0.46, 0.12)	19.51	36.59	56.10	17.07
	Total tyrosols	−0.25	(−0.54, 0.05)	19.51	34.15	53.66	19.51
Plasma sample	Total (poly)phenols	0.29	(0.01, 0.57)	27.78	41.67	69.44	2.78
	Total flavonoids	−0.13	(−0.44, 0.18)	19.44	36.11	55.56	13.89
	Total dihydrochalcones	−0.09	(−0.37, 0.19)	5.56	58.33	63.89	13.89
	Total flavan-3-ols	0.22	(−0.10, 0.54)	30.56	41.67	72.22	8.33
	Total flavanones	−0.16	(−0.50, 0.19)	25.00	36.11	61.11	19.44
	Total flavonols	0.07	(−0.26, 0.39)	30.56	33.33	63.89	11.11
	Total isoflavonoids	−0.13	(−0.46, 0.19)	19.44	36.11	55.56	13.89
	Total phenolic acids	0.22	(−0.10, 0.54)	38.89	30.56	69.44	8.33
	Total hydroxybenzoic acids	−0.18	(−0.51, 0.15)	30.56	22.22	52.78	16.67
	Total cinnamic acids	0.04	(−0.29, 0.38)	22.22	47.22	69.44	13.89
	Total phenylacetic acids	−0.09	(−0.4, 0.22)	22.22	36.11	58.33	13.89
	Total stilbenes	0.24	(−0.07, 0.56)	27.78	47.22	75.00	8.33
	Total lignans	−0.07	(−0.38, 0.25)	11.11	52.78	63.89	13.89
	Total other (poly)phenol	−0.02	(−0.36, 0.32)	13.89	52.78	66.67	13.89
	Total tyrosols	−0.16	(−0.47, 0.16)	27.78	27.78	55.56	16.67

^a Correctly classified (%): correctly classified the (poly)phenols into the same or adjacent quartiles (%). FFQs: food frequency questionnaires, kappa: weighted kappa coefficient (linear weights). CI: confidence interval.

4. Discussion

This is the first study that has developed and validated a FFQ to capture (poly)phenol intake in a UK population. A comprehensive and (poly)phenol-focused food list was established based on the EPIC-Norfolk FFQ and was expanded to 442 items. Non-(poly)phenol items were included to retain the structure of the EPIC-Norfolk FFQ, thus allowing for the estimation of overall nutrient intake as well. The KP-FFQ was evaluated against 7DDs as the dietary referent assessment, and objective (poly)phenol biomarkers from urine and plasma samples. Moderate agreements were shown in 10 groups of (poly)phenols estimated from KP-FFQs and 7DDs, and strong associations were found in 10 (poly)phenols estimated from 7DDs. A moderate agreement was found in fibre between KP-FFQs and 7DDs. As for biomarkers, KP-FFQs estimated (poly)phenol intake exhibited positive associations with 78 urinary metabolites and 19 plasma metabolites. Total (poly)phenol, total phenolic acids, and cinnamic acids from the urine sample and flavan-3-ols and stilbenes from the plasma sample exhibited more than 70% correct classification in the same or adjacent quartiles. Although moderate agreements were shown in all (poly)phenol intake levels from KP-FFQs and the EPIC-Norfolk FFQs after adjusting for energy intake, stronger and more agreements and associations were found in

KP-FFQs estimated (poly)phenol with 7DDs and biomarkers than the (poly)phenol intake from EPIC-Norfolk FFQs.

In this study, the total (poly)phenol intake levels estimated from KP-FFQs were higher than those derived from EPIC-Norfolk FFQs (1366.5 mg d^{−1} vs. 962.1 mg d^{−1}). The results from KP-FFQs were similar to the intakes reported by the EPIC-calibration study using 24 h recalls (around 1600 and 1750 mg d^{−1} for women and men, respectively),¹⁵ despite the differences between the databases in the EPIC and our study,¹⁵ for instance, the (poly)phenols were in the form of glycosides in the EPIC study rather than aglycone equivalents in the current study. Data from the UK NDNS (National Diet and Nutrition Survey) (2008 to 2014) shows that total (poly)phenol intake was around 600 mg d^{−1} from 4 day food diaries in a similar age group (19–34 years old) as in our study,³⁸ which was lower than the intake from the KP-FFQ but similar to the intake from EPIC-Norfolk FFQ in our study. However, the result reported from the NDNS might be lower than the actual intake since it only used Phenol-Explorer as the data source and did not include lignans and other (poly)phenols in the estimation of total (poly)phenol intake.¹⁵ In the EPIC-Norfolk FFQ, phenolic acids were identified as the primary contributors (53.6%) to total (poly)phenol intake rather than flavonoid intake (44.6%) in accordance with our prior research estimated by EPIC-Norfolk FFQ.⁶ However, flavonoids represent the





Table 8 Agreements between (poly)phenol intake estimated from EPIC-Norfolk and KCL (poly)phenol FFQ

(Poly)phenols (mg d ⁻¹)	ICC-A	(95% CI)	ICC-C	(95% CI)	Kappa	(95% CI)	Same quartile (%)	Adjacent quartile (%)	Correctly classified ^a (%)	Opposite quartile (%)	Spearman's Rho
Total (poly)phenols	0.54	(0.33, 0.68)	0.58	(0.45, 0.68)	0.54	(0.44, 0.64)	43.13	42.65	85.78	2.84	0.60 a
Total flavonoids	0.58	(0.29, 0.73)	0.64	(0.52, 0.72)	0.53	(0.43, 0.64)	44.55	39.81	84.36	2.84	0.56 a
Anthocyanins	0.09	(-0.14, 0.29)	0.13	(-0.14, 0.33)	0.39	(0.27, 0.51)	37.44	41.23	78.67	5.21	0.44 a
Chalcones	0.39	(0.20, 0.54)	0.39	(0.20, 0.53)	0.39	(0.27, 0.50)	27.96	55.45	83.41	6.16	0.38 a
Dihydroflavonols	0.10	(-0.15, 0.30)	0.11	(-0.16, 0.32)	0.56	(0.45, 0.67)	53.55	30.81	84.36	3.32	0.56 a
Dihydrochalcones	0.63	(0.51, 0.72)	0.64	(0.53, 0.73)	0.52	(0.41, 0.63)	48.82	35.07	83.89	3.79	0.57 a
Total flavan-3-ols	0.62	(0.44, 0.73)	0.65	(0.54, 0.73)	0.55	(0.45, 0.66)	48.34	37.44	85.78	3.32	0.56 a
Flavan-3-ol onomers	0.68	(0.58, 0.76)	0.69	(0.60, 0.77)	0.56	(0.46, 0.66)	43.60	44.08	87.68	3.32	0.63 a
Theaflavins	0.61	(0.49, 0.71)	0.62	(0.50, 0.71)	0.49	(0.38, 0.60)	41.71	41.71	83.41	3.79	0.52 a
Thearubigins	0.61	(0.49, 0.71)	0.62	(0.50, 0.71)	0.56	(0.46, 0.67)	47.39	39.34	86.73	3.32	0.57 a
Proanthocyanidins	0.42	(-0.09, 0.66)	0.53	(0.39, 0.64)	0.47	(0.36, 0.58)	44.55	35.55	80.09	3.32	0.54 a
Flavanones	0.23	(-0.01, 0.41)	0.25	(0.02, 0.43)	0.38	(0.26, 0.50)	40.28	35.55	75.83	4.27	0.41 a
Flavones	0.14	(-0.11, 0.34)	0.17	(-0.08, 0.37)	0.35	(0.24, 0.47)	34.12	41.23	75.36	4.27	0.35 a
Flavonols	0.77	(0.63, 0.84)	0.79	(0.73, 0.84)	0.59	(0.49, 0.68)	46.45	39.34	85.78	1.42	0.68 a
Isoflavonoids	0.55	(0.41, 0.66)	0.55	(0.41, 0.66)	0.55	(0.45, 0.65)	48.34	33.65	81.99	1.42	0.57 a
Total phenolic acids	0.59	(0.46, 0.68)	0.59	(0.46, 0.69)	0.64	(0.55, 0.73)	50.71	38.86	89.57	1.90	0.67 a
Hydroxybenzoic acids	0.42	(0.22, 0.57)	0.44	(0.27, 0.57)	0.49	(0.38, 0.60)	43.60	38.86	82.46	3.79	0.49 a
Ellagitannins	0.38	(0.19, 0.53)	0.38	(0.19, 0.53)	0.04	(-0.09, 0.17)	22.75	41.23	63.98	10.90	0.03 b
Hydroxycinnamic acids	0.60	(0.47, 0.69)	0.60	(0.48, 0.69)	0.65	(0.57, 0.73)	49.76	40.28	90.05	1.42	0.68 a
Hydroxyphenylacetic acids	0.69	(0.56, 0.78)	0.71	(0.62, 0.78)	0.54	(0.43, 0.64)	46.45	38.39	84.83	3.32	0.57 a
Total stilbenes	0.16	(-0.12, 0.36)	0.20	(-0.05, 0.39)	0.48	(0.37, 0.59)	41.23	42.18	83.41	4.27	0.49 a
Resveratrol	0.25	(-0.09, 0.47)	0.32	(0.10, 0.48)	0.42	(0.30, 0.54)	41.23	37.91	79.15	4.74	0.44 a
Total lignans	0.08	(-0.17, 0.28)	0.09	(-0.20, 0.30)	0.44	(0.33, 0.55)	34.60	48.34	82.94	4.74	0.49 a
Other (poly)phenols	0.03	(-0.23, 0.24)	0.03	(-0.27, 0.26)	0.27	(0.15, 0.39)	31.75	39.34	71.09	5.21	0.29 a
Tyrosols	0.04	(-0.13, 0.21)	0.09	(-0.20, 0.30)	0.30	(0.17, 0.42)	32.70	40.76	73.46	5.69	0.31 a
Alkylmethoxyphenols	0.45	(0.25, 0.59)	0.47	(0.31, 0.60)	0.54	(0.44, 0.65)	45.02	41.71	86.73	3.79	0.57 a
Alkylphenols	0.19	(-0.06, 0.38)	0.19	(-0.06, 0.38)	0.37	(0.25, 0.50)	40.76	37.91	78.67	6.64	0.35 a

^a Correctly classified (%): correctly classified the (poly)phenols into the same or adjacent quartiles (%). ICC-C: intraclass correlation coefficient-consistency model: when the systematic difference between EPIC-Norfolk FFQ and KCL (poly)phenol FFQ estimated (poly)phenol intakes were not relevant. ICC-A: intraclass correlation coefficient-agreement model: when the systematic difference between EPIC-Norfolk FFQ and KCL (poly)phenol FFQ estimated (poly)phenol intakes were relevant. EPIC: European Prospective Investigation into Diet and Cancer, FFQ: food frequency questionnaire, kappa: weighted kappa coefficient (linear weights). CI: confidence interval. a $p < 0.001$. b $p > 0.05$.



Table 9 Agreements between EPIC-Norfolk FFQs and KCL (poly)phenol FFQs estimated nutrients

Nutrients	ICC-A	(95% CI)	ICC-C	(95% CI)	Kappa	(95% CI)	Same quartile (%)	Adjacent quartile (%)	Correctly classified ^a (%)	Opposite quartile (%)	Spearman's Rho
Energy (kcal)	0.59	(0.23, 0.76)	0.66	(0.27, 0.74)	0.55	(0.45, 0.65)	44.81	40.09	84.90	2.36	0.61 a
Fibre (g d ⁻¹)	0.60	(0.21, 0.77)	0.68	(0.41, 0.75)	0.69	(0.62, 0.77)	53.77	37.74	91.51	0.94	0.72 a
Calcium (mg d ⁻¹)	0.54	(0.40, 0.65)	0.54	(0.16, 0.65)	0.38	(0.26, 0.50)	39.15	38.21	77.36	5.19	0.43 a
Iron (mg d ⁻¹)	0.52	(0.11, 0.72)	0.61	(0.29, 0.70)	0.62	(0.53, 0.71)	45.75	42.92	88.67	1.42	0.65 a
Potassium (mg d ⁻¹)	0.67	(0.50, 0.78)	0.71	(0.36, 0.78)	0.60	(0.50, 0.70)	51.42	35.38	86.80	2.36	0.64 a
Retinol (µg d ⁻¹)	0.27	(0.04, 0.44)	0.27	(-0.09, 0.44)	0.39	(0.27, 0.51)	40.57	37.26	77.83	5.19	0.44 a
Carotene (µg d ⁻¹)	0.67	(0.48, 0.77)	0.70	(0.45, 0.77)	0.69	(0.62, 0.77)	49.53	43.40	92.93	0.94	0.76 a
Vitamin C (mg d ⁻¹)	0.47	(0.07, 0.67)	0.55	(0.29, 0.66)	0.56	(0.46, 0.66)	46.23	39.15	85.38	2.36	0.62 a
Fat (g d ⁻¹)	0.55	(0.09, 0.74)	0.64	(0.21, 0.73)	0.54	(0.44, 0.64)	43.87	39.62	83.49	1.89	0.56 a
Cholesterol (mg d ⁻¹)	0.36	(-0.25, 0.65)	0.52	(0.18, 0.63)	0.48	(0.37, 0.59)	41.98	39.15	81.13	3.30	0.53 a
MUFA (g d ⁻¹)	0.58	(0.31, 0.72)	0.63	(0.22, 0.72)	0.52	(0.42, 0.62)	41.51	40.09	81.60	1.42	0.54 a
PUFA (g d ⁻¹)	0.55	(0.26, 0.70)	0.60	(0.29, 0.70)	0.49	(0.38, 0.60)	42.92	41.51	84.43	4.72	0.49 a
SFA (g d ⁻¹)	0.66	(0.53, 0.75)	0.68	(0.23, 0.76)	0.52	(0.42, 0.63)	41.98	42.45	84.43	2.83	0.60 a
Protein (g d ⁻¹)	0.67	(0.54, 0.76)	0.69	(0.34, 0.77)	0.58	(0.49, 0.68)	47.17	38.21	85.38	1.42	0.65 a
Carbohydrate (g d ⁻¹)	0.64	(0.45, 0.76)	0.68	(0.37, 0.76)	0.58	(0.49, 0.68)	48.58	36.79	85.37	1.89	0.62 a
Sugars (g d ⁻¹)	0.69	(0.52, 0.79)	0.72	(0.40, 0.79)	0.59	(0.50, 0.68)	45.28	41.04	86.32	1.42	0.64 a
Fructose (g d ⁻¹)	0.63	(0.33, 0.77)	0.69	(0.41, 0.76)	0.62	(0.53, 0.70)	45.28	42.45	87.73	0.94	0.67 a
Galactose (g d ⁻¹)	0.21	(-0.02, 0.4)	0.22	(-0.05, 0.41)	0.38	(0.26, 0.51)	42.45	34.91	77.36	5.66	0.41 a
Glucose (g d ⁻¹)	0.72	(0.53, 0.82)	0.76	(0.51, 0.82)	0.62	(0.53, 0.71)	46.70	42.45	89.15	1.89	0.69 a
Starch (g d ⁻¹)	0.59	(0.41, 0.71)	0.62	(0.36, 0.71)	0.51	(0.40, 0.62)	49.06	32.55	81.61	3.30	0.56 a
Sucrose (g d ⁻¹)	0.74	(0.67, 0.81)	0.74	(0.44, 0.80)	0.63	(0.54, 0.72)	48.58	40.09	88.67	1.42	0.68 a
Lactose (g d ⁻¹)	0.28	(-0.18, 0.54)	0.39	(0.13, 0.54)	0.36	(0.24, 0.48)	35.38	42.92	78.30	6.13	0.37 a
Maltose (g d ⁻¹)	0.15	(-0.09, 0.35)	0.18	(-0.09, 0.37)	0.32	(0.20, 0.45)	34.43	42.92	77.35	7.08	0.34 a

^a Correctly classified (%): correctly classified the (poly)phenols into the same or adjacent quartiles (%). ICC-C: intraclass correlation coefficient-consistency model: when the systematic difference between EPIC and KCL (poly)phenol FFQs estimated (poly)phenol intakes was not relevant. ICC-A: intraclass correlation coefficient-agreement model: when the systematic difference between EPIC-Norfolk and KCL (poly)phenol FFQs estimated (poly)phenol intakes were relevant. EPIC: European Prospective Investigation into Diet and Cancer, FFQs: food frequency questionnaires, kappa: weighted kappa coefficient (linear weights). CI: confidence interval. a $p < 0.001$.

highest contributor compared with phenolic acids in the total (poly)phenol intake estimated from KP-FFQ as reported in the NDNS research,³⁸ which is likely due to the different coffee and tea contribution between the two FFQs in our cohort. Here, in the KP-FFQ, coffee and tea contributed 31.0% (1.1 ± 0.4 cup per d (203.6 ± 78.6 g d⁻¹)) and 12.7% (0.8 ± 0.2 cup per d (147.0 ± 28.6 g d⁻¹)) of the total (poly)phenol intake, respectively, whereas in the EPIC-Norfolk FFQ, coffee and tea contributed 41.0% (0.8 ± 1.0 cup per d (145.3 ± 187.1 g d⁻¹)), and 23.6% (0.7 ± 1.1 cup per d (129.8 ± 200.0 g d⁻¹)) to the total (poly)phenol intake (standardized as 190 g per cup according to the default portion size in the EPIC-Norfolk FFQ). This varying ratio of tea and coffee consumption could partially elucidate the differential contributions of phenolic acids and flavonoids to the overall intake of (poly)phenols. Notably, the population we evaluated comprises young individuals (19–29 years old) with high education level (college students) and high percentage of Asian than the general UK population, and a higher proportion of coffee consumers (76.2 and 79.6% from EPIC-Norfolk and KP-FFQs, respectively) than that in the UK adults (62.0%).⁷¹ Moreover, due to the large proportion of Asian people in the targeted population, white rice, as the major food source, represents a good source of dietary (poly)phenols, contributing 2% to the total (poly)phenol intake estimated from KP-FFQ.

The estimated class and subclass of (poly)phenol intake levels were different between the two FFQs, with 23 out of 26 (poly)phenol groups being higher in the KP-FFQs, which aligns with our expectations. Compared with EPIC-Norfolk FFQs, KP-FFQs captured more food sources of (poly)phenols, for instance, blueberry, grape, aubergine, olive, herbs and spices, seeds, alternative milk such as almond, oat, soy milk, and sauces such as soy sauce. Anthocyanins, a subclass of (poly)phenols, play a role in the skin colouring of fruits such as apples.⁷² The (poly)phenol content of fruits differs depending on whether the skin is included, such as apple and pear (apple: peeled 26.52, non-peeled 55.94 mg per 100 g; pear: peeled 0.58, non-peeled 1.65 mg per 100 g fresh weight of total (poly)phenol (aglycone equivalent) from in-house database^{6,36}), while this was not distinguished in the EPIC-Norfolk FFQ. Besides, the fruits or vegetables with diverse colours can have diverse (poly)phenol content, which are also not distinguished in the EPIC-Norfolk FFQ, for instance, grapes (green: 10.30, black: 7.49 mg per 100 g fresh weight of total (poly)phenol (aglycone equivalent)^{6,36}), and onions (red: 2.60, white: 0.34, yellow: 1.73 mg per 100 g fresh weight of total (poly)phenol (aglycone equivalent)^{6,36}). Moreover, several food items with distinct (poly)phenol levels or profiles were grouped in one question in the EPIC-Norfolk FFQs, for instance, tea (black, green, and herbal tea), wine (white, rose, or red wine), “strawberries, raspberries, kiwi fruit”, “peanuts or other nuts”, and “dried lentils, beans, peas”. Participants may interpret the questions differently, while in analysis, those foods were transformed into a combination of default items.⁶ The above issues could all lead to potential underestimation of (poly)phenols in the

EPIC-Norfolk FFQs compared with the KP-FFQs. In addition, due to the expanded food list in the KP-FFQs, it has a longer completion time than the EPIC-Norfolk FFQs (36.9 ± 21.4 min vs. 13.9 ± 6.8 min). While the amount of time needed is acceptable, it may lead to over or underestimation of (poly)phenol consumption. As a result, the agreement between the two FFQs was moderate. Agreements were extremely poor for the groups contributing a small percentage of the total intake, including anthocyanins, dihydroflavonols, flavones, total other (poly)phenols, and tyrosols, which required a more detailed measurement tool⁶ such as the KP-FFQ to be fully captured.

The food records are not limited to a predefined food list, which enables more specificity as (poly)phenol content is linked to individual food items rather than less food groups in the FFQs and captures day-to-day variabilities and less common foods.¹⁶ Regarding the relative validity against 7DDs, moderate agreements were exhibited in limited (poly)phenols, including dihydroflavonols, dihydrochalcones, total phenolic acids, hydroxycinnamic acids, and alkylmethoxyphenols, and only alkylphenols from 7DDs was significantly associated with the intake from EPIC-Norfolk FFQs. 7DDs do not have predefined food lists and allow matching individual food items with (poly)phenol content,¹⁶ which allow more accurate estimation of the (poly)phenol intake. However, 7DD only captures intake for a short period (1 week), which are prone to inter-day/seasonal variations in the diet. These difference in tools could all lead to the discrepancies between the two FFQs and 7DDs in our results. Previous validation studies between EPIC-Norfolk FFQs and 3 day food records found an ICC of 0.489 for total (poly)phenol intake⁷³ and a cross-classification test of 30.2%⁷³ to 36.6%⁷⁴ of the same quartiles for total (poly)phenols and 18.0% to 31.0% for flavonoids subclasses,⁷⁵ which is in accordance with our results (33.3% for total (poly)phenol and 21.7% to 48.3% for flavonoids subclasses). In comparison, a 0.73 kappa for total phenolic acids and hydroxycinnamic acids was estimated from KP-FFQs and 7DDs with a cross-classification test of 96.7% in the same and adjacent quartile with no opposite quartile. This high agreement may be due to the similar food source and contribution in the two measurements, with coffee, rice, tea, chestnuts, and blueberries contributing 67.5%, 5.1%, 2.4%, 2.1%, and 1.8% in KP-FFQs and 65.7%, 4.2%, 2.0%, 1.3% and 2.2% in 7DDs for total phenolic acids, and coffee, rice, apple, and blueberries contributing 74.6%, 5.3%, 2.0%, and 1.9% in KP-FFQs and 72.3%, 4.7%, 1.8%, and 2.3% in 7DDs for hydroxycinnamic acids, respectively. In addition, compared to the agreements between EPIC-Norfolk FFQs and 7DDs, more (poly)phenols with moderate agreements between KP-FFQs and 7DDs were exhibited, including total (poly)phenol and subclass of (poly)phenols from four out of five classes, for instance, chalcones, isoflavonoids, ellagitannins, hydroxyphenylacetic acids, resveratrol, tyrosols, and alkylmethoxyphenols. The 442 food items in KP-FFQs cover the most important (poly)phenol dietary sources in the UK diet through the NDNS study 2008–2014.³⁸ It integrates closely with the free-living eating habits and agrees more with the detailed food source of (poly)phenols captured



by 7DDs, which contributed to better agreements than the EPIC-Norfolk FFQs. Coherently, strong associations were exhibited in ten (poly)phenols from four classes estimated from 7DDs with (poly)phenols from KP-FFQs, including dihydroflavonols, theaflavins, thearubigins, flavones, isoflavonoids, ellagitannins, hydroxyphenylacetic acids, total stilbenes, resveratrol, and tyrosols. To note, no gold standard method has been established to measure (poly)phenol intake. The food records also require repeat measurements to capture a period of dietary estimation, for instance, conducted in different seasons to represent yearly diet intake.^{76,77} Compared with the time coverage of the past year in the FFQs, the lack of yearly representative of food records may contribute to the poor agreements of several (poly)phenols estimated from KP-FFQs.

A large panel of plasma and urine (poly)phenol metabolites were used to enhance the validity testing of FFQs against 7DDs. Regarding the association with urinary and plasma metabolites, stronger significant relationships were observed in KP-FFQs estimated intake compared to EPIC-Norfolk FFQs, which aligns with the better association result of KP-FFQs estimated intake with 7DDs. Compared with urinary metabolites, fewer positive associations were found with plasma metabolites, which may be attributed to the collection time of plasma with more than 8 hours of fasting, resulting in the removal of many metabolites from circulation. Despite the better associations between urinary metabolites with (poly)phenol intake than plasma, spot urine in our research only provided a snapshot of excreted (poly)phenols compared with 24 hour urine, which is able to capture more comprehensive information due to the longer collection time. The agreements were poor between metabolites in urine and plasma and estimated dietary (poly)phenol intake from both EPIC-Norfolk FFQ and KP-FFQ. These poor agreements may be attributed to the extensive metabolism of dietary (poly)phenols after ingestion, including phase II metabolisms into glucuronides, sulfates, and ring fissions into smaller molecules by the gut microbiota.⁷⁸ Some phenolic compounds with small molecular weight, such as phenolic acids, benzaldehydes, and benzenes, would be present in food and be generated by the gut microbiota from various types of (poly)phenol molecules. Therefore, the endogenous pathways of phenolic metabolites and inter-individual variability in (poly)phenol gut microbial metabolism may lead to poor agreements between the (poly)phenols of the same class/subclass from diet and in biosamples. Different half-lives of the various (poly)phenols and the sample collection time concerning the dietary assessment further influenced the agreements. For example, FFQs reflect habitual intakes, whereas the spot urine and fasting plasma rather reflect recent (poly)phenol intake in the past 24 hours. In addition, the restrictions in the reporting accuracy of the dietary assessment methods, the limited number of specimen samples, and diverse sources of (poly)phenol exposure, such as food additives, also contributed to the discrepancies. Further validation in larger cohorts with higher specimen sample sizes is required.

As for the nutrient intake, including energy, fibre, and macronutrients, the agreement between EPIC-Norfolk and KP-FFQs was moderate, with the ICC, kappa, and Spearman's correlation higher than 0.5. More than 80% of participants were found correctly classified into the same or adjacent quartiles in our study, from fat (83.5%) to fibre (91.5%), which is similar to the previous findings between FFQ and 3 day dietary record with a fair agreement of more than 70% correct classification from fat (70.3%) to energy (84.2%).⁷⁹ In addition, less than 2.5% of the participants were grossly misclassified (in the opposite quartile), with only 0.89% of misclassification of fibre intake. Plant-based foods are sources of dietary (poly)phenols, which are also important sources of fibre in the human diet.⁸⁰ The (poly)phenol-focused food list of the questionnaire contributed to the fair agreement in fibre intake. When comparing the nutrients estimated from KP-FFQs against 7DDs, the agreements were poor in general, and only fibre in KP-FFQs showed moderate agreement with 7DDs (kappa = 0.40). However, more than 50% of the participants were correctly classified into the same or adjacent quartiles. Less than 10% of the participants were grossly misclassified, except for energy (11.67%), protein (13.33%) and animal-related nutrient cholesterol (15.0%). The validation study of macronutrient intakes from the EPIC FFQ compared against 24 hour dietary recalls reported a higher percentage of correctly classified nutrients, including energy, protein, fat, and carbohydrate (73.1%, 74.8%, 75.3%, and 71.7%) compared to our study (61.7%, 60.0%, 60.0%, and 58.3%).⁸¹ However, the percentage of correctly classified fibre intake was lower in their research compared to ours (66.4% vs. 76.7%).⁸¹ Dietary validation studies recommended that more than half of the correct classification, less than 10% grossly misclassification, and weighted kappa values above 0.4 are desirable for nutrients of interest to minimise the false-negative associations between diet and health outcome.⁸² In the present study, overall agreements for energy and nutrients between two FFQs were considered fair and reasonable, but the agreement between the KP-FFQs and the 7DDs was less satisfactory. The discrepancy in the agreements may be due to the small sample size of the validation study against 7DDs ($n = 60$), though a sample size of at least 50 has been suggested for validity testing.¹¹ A larger sample would be warranted in future studies for the validation test against dietary records. The finding also implied that KP-FFQs might overestimate the dietary intake due to the expanded food list, which may also explain the fair agreement with EPIC-Norfolk FFQs, which are also prone to overreport.^{83,84}

The strengths and limitations should be noted when interpreting the results. To our knowledge, this is the first study to develop an FFQ to capture the dietary (poly)phenol among a young adult UK population. The (poly)phenol metabolites estimated from 24 hour urine and plasma were conducted as the reference method to strengthen the relative validity assessment power of 7DDs since no objective 'gold standard' reference measurement tool has been developed for dietary assessment. In addition, the collection of KP-FFQ was prior to the reference



tool of 7DDs following the guidance of Cade *et al.* on the order of validation, administering the test instrument before the reference instrument to avoid drawing participants' attention to their diets.¹¹ The KP-FFQ extended the (poly)phenol-rich food list and kept the (poly)phenol-free food items included in the EPIC-Norfolk FFQ, such as meat products. Since dietary (poly)phenol consumption cannot be captured fully with a finite food list, the extending of the questionnaire length is unavoidable.¹¹ It disaggregated the combined questions of plant-rich foods and included more plant food groups, which may potentially overestimate the intake of plant food and nutrients than the established EPIC-Norfolk FFQ. Since the 442 food items may result in a higher burden on participants and lead to misreport, we tested the average time to complete the EPIC-Norfolk and KP-FFQs in the pilot study with 13.9 and 36.9 minutes, respectively. Considering the much longer completion time than EPIC-Norfolk FFQs, the proper order of food groups is significant. The food groups of particular interest, (poly)phenol-rich food, should be placed at the beginning of the FFQs,¹¹ and the (poly)phenol-free food groups towards the end since the accuracy of responses may decline due to boredom or fatigue.¹¹ Other limitations are mainly related to the general characteristics of FFQs to estimate dietary components. FFQs are prone to self-reporting errors in determining frequencies over the long-term and pre-quantified food portion size.¹⁷ Clear instructions on completion and photos of portion sizes^{85–87} may help estimate the portion size.¹¹ In addition, the prolonged reference period of FFQs has been proved to overestimate healthy food intake, such as fruits and vegetables, prominent sources of (poly)phenols,^{17,88} which may also contribute to errors in the present study. Another limitation relates to the reference (poly)phenol database. Although our in-house database included the well-established Phenol-Explorer database,³² the USDA database and several published papers,^{36–58} it is important to acknowledge that the limited information on the influence of harvest conditions, food processing, storage, and cooking methods on the (poly)phenol content of foods restricted the proper interpretation of dietary (poly)phenol intake data.⁸⁹ Moreover, the lack of representative population in our study should also be noted for ethnicity, and especially the narrow age range, which primarily consists of university students with high education levels. A larger and more representative population for the further reliability test of the newly developed FFQ may be warranted.

5. Conclusion

In conclusion, a novel KP-FFQ developed to estimate dietary (poly)phenol intake in the UK population demonstrated satisfactory validation results, exhibiting moderate to strong agreements and associations with 7DDs and biofluid metabolites. KP-FFQ performs better than established EPIC-Norfolk FFQ for (poly)phenol intake when tested against biomarkers and 7DDs. However, limitations were identified in KP-FFQ for estimating nutrient intake, as it potentially overestimates plant-

based food intake due to disaggregation and inclusion of more plant food groups compared with the established FFQ. Future studies should aim to validate this tool against repeated food diaries or weighed food records collected across several seasons to assess the instrument's ability to capture seasonal dietary (poly)phenol intake more accurately and potentially improve the validity for measuring nutrients. A representative UK population group is warranted to test the further usability.

Author contributions

Conceptualisation, Y. L., A. R. M., and R. G.; data curation, Y. L., Y. X., M. L. S., N. N. Z. K., H. W., J. H. and S. L.; formal analysis and writing – original draft, Y. L.; methodology, Y. L., Y. X., A. R. M. and R. G.; writing – review & editing; supervision, A. R. M., and R. G.; project administration and funding acquisition, A. R. M.

Data availability

Due to privacy and ethical concerns, the raw data supporting the findings of this study may be subject to restrictions. Detailed information on the dietary assessment tools, (poly)phenol intake estimates, and biomarker analysis methods can be provided to interested researchers in accordance with institutional and ethical guidelines.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

We extend our highest gratitude to the participants of all the studies. YL and YX are funded by the King's-China Scholarship Council (K-CSC) joint scholarship.

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