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NUTRIENT VALIDATION IN WOMEN'S LIFESTYLE VALIDATION STUDY

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Nutrient Validation In Women's Lifestyle Validation Study

Abstract

Nutritional factors have been intensively studied as important determinants of many diseases. Food frequency questionnaires (FFQ), dietary records, 24-hour dietary recalls, nutrient biomarkers are important dietary assessment methods, and are subject to various sources of measurement error. Given the limitations of these methods, much effort has been devoted to refining them and evaluating their ability to measure diet. This dissertation focused on evaluating the performance of a semi-quantitative FFQ (SFFQ), multiple web-based automated-self-administered 24-hour recalls (ASA24), 7-day dietary records (7DDR) and biochemical indicators in assessing nutrient intakes among women. Intraclass correlation coefficient, Spearman correlation coefficient, and validity coefficient calculated by method of triads were used to evaluate the reproducibility and validity of each dietary method.

The first paper evaluated the performance of a 152-item SFFQ comparing intakes of nutrients estimated by SFFQ with those measured by the average of two 7DDR, and of four ASA24s kept over a one-year period. The study SFFQ performed consistently well when compared with multiple diet records, and that modifications to the questionnaire over time have adequately taken into account the changes in the food supply and eating patterns that have occurred since 1980. Multiple ASA24s can provide similar estimates of validity as dietary records if day-to-day variation is taken into account.

The second paper explored the validity of long-term intakes of energy, protein, sodium and

potassium assessed by SFFQ and ASA24s using recovery biomarkers and 7DDR as standards. The study SFFQ and averaged ASA24's are reasonably valid measurements for energy-adjusted protein, sodium and potassium compared to multiple recovery biomarkers or dietary records. Recovery biomarkers should not be considered to be without error, including systematic within-person error.

Finally, the third paper further evaluated the validity of nutrient assessed by SFFQ and ASA24 compared with intake by the 7DDR and plasma levels of fatty acids, carotenoids, retinol, tocopherols and folate. Again, the study SFFQ provides reasonably valid measurements for specific fatty acid, most carotenoids, alpha-tocopherol and folate compared to concentration biomarkers or dietary records. Compared to SFFQ, almost all nutrients estimated by averaged ASA24s had relatively low correlations with biomarkers, 7DDRs and estimated 'true' underlying intakes.

Key words: nutrient validation; women's lifestyle validation study; semi-quantitative food frequency questionnaire; 7-day dietary records; automated-self-administered 24-hour recalls; recovery biomarkers; concentration biomarkers; intraclass correlation coefficient; spearman correlation coefficient; validity coefficient; method of triads.

TABLE OF CONTENTS

Paper I 1
Abstract 2
I. Introduction 3
II. Materials and Methods 4
A. Study Population 4
B. The semi-quantitative food frequency questionnaire (SFFQ) 6
C. 7 Day Dietary Records (7DDR) 7
D. Web-based, self-administered 24-hour dietary recall (ASA24) 8
E. Statistical Analyses 9
III. Results 11
IV. Discussion 15
V. References 18
VI. Figure and Tables 21
Paper II 31
Abstract 32
I. Introduction 34
II. Materials and Methods 35
A. Study Population 35
B. The semi-quantitative food frequency questionnaire (SFFQ) 38
C. 7 Day Dietary Records (7DDR) 39
D. Web-based, self-administered 24-hour dietary recall (ASA24) 40
E. Doubly-labeled water (DLW) 40
F. 24-hour Urine 41
G. Statistical Analysis 42
III. Results 45
IV. Discussion 49
V. References 53
VI. Figures and Tables 58
Paper III 66

Abstract 67
I. Introduction 69
II. Materials and Methods 70
A. Study Population 70
B. The semi-quantitative food frequency questionnaire (SFFQ) 72
C. 7-Day Weighed Dietary Records (7DDRs) 73
D. Web-based, self-administered 24-hour dietary recall (ASA24) 74
E. Fasting blood collection 74
F. Statistical Analysis 76
III. Results 79
IV. Discussion 82
V. References 86
VI. Figures and Tables 92
Supplementary Tables 101

FIGURES WITH CAPTIONS

Figure 1. Timeline of the dietary assessment activity in project 1 21
Figure 2. Timeline of the dietary assessment activity in project 2 58
Figure 3. Spearman correlation coefficients with biomarkers and validity coefficients for protein, sodium, potassium and energy intakes 65
Figure 4. Timeline of the dietary assessment activity in project 3 92
Figure 5. Spearman correlation coefficients and validity coefficients for specific fatty acids 97
Figure 6. Spearman correlation coefficients and validity coefficients for specific carotenoids, retinol, tocopherols and folate 99

TABLES WITH CAPTIONS

Table 1.1. Subject characteristics among WLVS participants in project 1 22
Table 1.2. Mean absolute daily nutrient intakes estimated by SFFQs, 7DDRs, and ASA24s 23
Table 1.3. Rank intraclass correlations of daily nutrient intake from SFFQ, 7DDR, and ASA24 25
Table 1.4. Spearman correlation coefficient for comparison of SFFQ2 with 7DDRs, and ASA24s 27
Table 1.5. Summary of Spearman correlation coefficient in overall and subgroups of participants 30
Table 2.1. Subject characteristics among WLVS participants in project 2 59
Table 2.2. Energy, protein, sodium and potassium intake distributions by different dietary methods 60
Table 2.3. Spearman correlation coefficients for protein, sodium, potassium and energy estimated by different methods 61
Table 2.4. Validity coefficients for energy, protein, sodium, potassium estimated by different methods.. 63
Table 3.1. Subject characteristics among WLVS participants in project 3. 93
Table 3.2. Distribution of specific fatty acids, carotenoids, retinol, tocopherols and folate 94
Table 3.3. Reproducibility of nutrient intakes estimated by SFFQ, 7DDR and ASA24 and biomarkers ... 95

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Paper I

Nutrient Validation Utilizing Self-Reported Dietary Assessment Methods In Women's Lifestyle Validation Study

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Abstract

This study evaluated the validity of a 152-item semi-quantitative food frequency questionnaire (SFFQ) by comparing intakes of energy and 44 nutrients without supplements estimated by SFFQ with those measured by the average of two 7-day dietary records (7DDR), and of four automated-self-administered 24-hour recalls (ASA24) kept over a one-year period by 632 women between 2010 to 2012. SFFQ and 7DDR were more reproducible than the ASA24. The mean rank intraclass correlation coefficient of intakes assessed by SFFQ was 0.68, 7DDRs 0.59 and ASA24s 0.25. Absolute intakes estimated by one SFFQ, two 7DDRs, and four ASA24s, were similar. Compared to the 7DDRs, the SFFQ tended to underestimate sodium intake, but overestimated intakes of energy, macronutrients and several nutrients in fruits and vegetables, such as carotenoids. Spearman correlation coefficients between energy-adjusted intakes from 7DDRs and the SFFQ2 completed at the end of the study's diet data collection ranged from 0.36 for lauric acid to 0.77 for alcohol (mean r=0.53). Correlations of the SFFQ2 were weaker when ASA24s were used as the comparison method (mean r =0.43). After adjustment for within-person variation in the comparison method, the correlations of the SFFQ2 were similar with 7DDRs (mean r =0.63) and ASA24s (mean r =0.62). For 22 nutrients that were also included dietary supplements, the correlations between SFFQ2 and 7DDR were generally higher after including supplements in those nutrients (mean r=0.71 vs 0.63 from diet alone). These data indicate that this SFFQ provides reasonably valid estimates for intakes of a wide variety of dietary variables, and that multiple ASA24 can provide similar estimates of validity as 7DDRs if day-to-day variation is taken into account.

I. Introduction

Nutritional factors have been intensively studied as important determinants of many diseases.[1] Food frequency questionnaires (FFQs) have commonly used to assess long-term or usual intake in large prospective studies. FFQs are based on an individual's recall of usual intake over an extended period of time, and thus are subject to measurement error. Dietary records, or interviewer-aided 24-hour dietary recalls, assessed at multiple time points, are most commonly used to evaluate the validity of FFQs. However, these methods are generally expensive to collect and process, and are likely to be unrepresentative of usual intake if only a few days are assessed.[1]

Given the limitations of these dietary assessment methods, much effort has been devoted to refining them and evaluating their ability to measure diet. The semi-quantitative FFQ (SFFQ) developed by our group has been shown to be a reasonably reproducible and valid measure of nutrient intakes among men and women in many populations.[1-4] After modifications to incorporate changes in the food supply and eating patterns, our current version of this SFFQ includes 152 food items. We have also developed a web-based version of this questionnaire (WebFFQ), which is enhanced by the use of branched questions and is utilized in the ongoing Nurses' Health Study 3.[5,6] Due to changes in the food supply and increases in meals eaten away from home, and reflective changes in the SFFQ since it was last evaluated in 1986[1], we conducted a detailed validation study of both the paper and web versions of the current SFFQ.

The automated-self-administered 24-hour recall (ASA24), developed by the National Cancer Institute (NCI),[7] is self-administered over the internet with minimal cost. The ASA24 could potentially serve as a lower cost method to evaluate the validity of other dietary methods. However, the performance of ASA24 as a comparison method has not been systematically evaluated. Thus, the present study aimed to evaluate the reproducibility and validity of nutrient

intakes measured by the paper and web SFFQ compared to 7-day dietary records (7DDRs) and the ASA24s among participants in the Nurses' Health Study and Nurses' Health Study II.

II. Materials and Methods

A. Study Population

The current study was based on the Women's Lifestyle Validation Study (WLVS), one of the three studies comprising The Multi-Cohort Eating and Activity Study for Understanding Reporting Error (MEASURE), which was designed to investigate the measurement error structure associated with self-reported dietary and physical activity assessments.[8] WLVS was conducted within the existing Nurses' Health Study (NHS) [9] and Nurses' Health Study II (NHS II).[10] The NHS began in 1976 when 121,701 U.S. female registered nurses aged 30-55 years were enrolled; a SFFQ was first administered in 1980. The NHS II cohort enrolled 116,671 female nurses aged 24-44 years in the United States in 1989; the first SFFQ was administered in 1991. Participants in both cohorts completed mailed questionnaires on their medical history and lifestyle factors at enrollment, and follow-up questionnaires every two or four years to update their information on disease or potential risk factors. Diet has been assessed by SFFQ every four years. This study was approved by the Human Subjects Committees of the Harvard T.H. Chan School of Public Health and Brigham and Women's Hospital.

In 2010, we randomly selected a subset of NHS and NHS II participants aged 45-80 years, from all geographical regions of the US, who had completed the 2006/2007 cohort SFFQ, had previously provided blood samples, had access to broadband internet, and were not planning to change their diet nor their physical activity levels. Women with medical history of coronary heart disease, stroke, cancer, or major neurological disease were excluded. The sample selection

was stratified by age, and African-Americans were oversampled. A total of 5,509 female nurses were invited via email, by regular US mail method or both. Of these, 796 consented to participate, and received information on the timeline and activities. Study instructions were provided on the study website. Participants received support from study staff throughout their participation via the internet, telephone, and mail. Study participants were offered $600 for completion of the study due to the many complex activities.

The study included anthropometric measurements, doubly-labeled water (DLW) assessment of energy expenditure, fasting blood draws, dietary and physical activity questionnaires, saliva collections, activity monitor recordings, and multiple 24-hour and first morning urine collections. To represent the one-year period typically used as the time frame for dietary questionnaires, we spread the dietary and biomarker measurements over a period of approximately one-year and altered the order of measurements by dividing the participants into four groups. This analysis focused on the self-reported dietary measurements, and other assessments will be evaluated in separate reports. The current analysis consisted of two paper SFFQ's administered one year apart and an online WebFFQ; during that year we collected four ASA24s and two 7DDRs. The two paper SFFQs were collected at baseline and at end of each participant's diet data collection year, the WebFFQ was collected two weeks before or after completion of the second paper SFFQ and in random order, one ASA24 was collected in each season and the two 7DDRs were collected approximately 6 months apart to capture seasonal variability as shown in Figure 1. Three ASA24s were collected on weekdays and one on a weekend to capture differences in eating habits. By design, the 7DDRs and ASA24s in the same phase were collected several weeks apart to avoid artificially high correlations.[11] The study was conducted from June, 2010 to March, 2012.

Among 796 consented participants, 795 completed SFFQ1, 774 completed at least one 7DDR, 692 completed at least one ASA24, 759 completed SFFQ2, and 747 completed the WebFFQ. 19 women in WLVS had participated in previous validations in either 1980 (N=13) or 1986 (N=19). Of the 692 participants completing ASA24s, 93 (13%) competed one ASA24, 136 (20%) completed two, 219 (32%) competed three, and 244 (35%) completed four ASA24s. The distribution of ASA24 over the four phases was 512, 496, 497 and 493, respectively. Lower completion rates for the ASA24 were likely related, at least in part, to difficulties with the internet interface, which has been addressed in more recent versions of ASA24. In the quality control of SFFQ nutrient measurements, we excluded those participants with total daily energy intake <600 kcal or >3500 kcal or with more than 70 blanks. Overall, 771 SFFQ1 (97%), 742 SFFQ2 (98%), and 721 WebFFQ (97%) were included. These same restrictions for kilocalories and blank food items have been applied in analyses of diet and disease in the NHS and NHSII studies. Our primary analysis included 632 participants with complete data for SFFQ1, SFFQ2, WebFFQ, at least one 7DDR and one ASA24. African-Americans comprised 7% of participants. Information on year of birth, height, weight, ethnicity and smoking status was collected at enrollment. Weight information was also collected every three months during follow up.

B. The semi-quantitative food frequency questionnaire (SFFQ)

The 152-food-item semi-quantitative SFFQ is an expanded version of a previously validated questionnaire (www.channing.harvard.edu/nhs/questionnaires/pdfs/NHSII/2007.pdf).[1,4] The questionnaire was designed to classify individuals according to levels of average intake of selected foods and nutrients over the past year. Respondents were asked how often, on average, they had consumed the specified amount of each type of food or beverage during the past year; nine possible frequency categories ranged from never/almost never to 6 or more times per day.

Open-ended questions were used for usual brand and type of margarine, cooking oil, cold breakfast cereal, and multivitamins. We also collected detailed information regarding the type of fat used at the table and in food preparation. Participants were also asked to report up to 3 foods that they consume more than once per week that were not included in the SFFQ. Nutrient intakes were calculated from the questionnaire by multiplying a weight proportional to the frequency of use (where once per day is equal to one) by the nutrient composition for the portion size specified for each food or vitamin supplement. Nutrients were then summed across all foods and supplements to obtain a total nutrient intake for each individual; we also calculated nutrient intake from food without supplement intake.

Over 200 nutrients and dietary constituents are derived from the SFFQ using an extensive and regularly updated food composition database maintained at Harvard T.H. Chan School of Public Health. The food composition data base used to calculate nutrient values is based primarily on US Department of Agriculture publications and is continually supplemented by other published sources and personal communications from laboratories and manufacturers (https://regepi.bwh.harvard.edu/health/nutrition/repeatUser.html). The web-based version of this questionnaire was similar to the standard paper questionnaire and was enhanced by the use of branched questions to collect details of items such as breakfast cereals and margarines.

C. 7 Day Dietary Records (7DDR)

WLVS research dietitians developed the 7DDRs with detailed instructions and a DVD instructional video. Each participant received an Escali food scale and ruler, the instructional DVD, and instructions via telephone by our research dietitians explaining how to keep detailed 7DDRs. The detailed instructions were also included in the 7DDR booklet. The Harvard Automated Research Information System (a computerized reminder system) was used to track

participant study activities, sent reminder emails to participants on day 2, day 3 or 4 and day 6 of the diet recording week and to encourage participants to complete their diet record, to ask study research dietitians for clarifications on how to complete their diet records, if needed, and to review the DVD. Participants reported the gram weights for food intake and provided recipes of all home prepared foods including the number of recipe servings, and the portion of the recipes they each consumed. Participants also reported gram weights of foods before and after consumption, as relevant, so actual intake could be computed. Additionally, participants collected labels of store brand products and returned them with their records for analysis. A research dietitian contacted each participant during the recording week to discuss any questions and review procedures. Issues found in their first 7DDR provided after coding by the Nutrition Coordinating Center (NCC) at the University of Minnesota[12] were specifically addressed during their second 7DDR instructional session with the research dietitian. The NCC used Nutrition Data System for Research (NDSR) 2011 to analyze the 7DDRs.[13,14] Nutrient intakes were calculated using the NCC food composition data base that is derived primarily from USDA sources; over 150 nutrient and dietary constituents were derived.

D. Web-based, self-administered 24-hour dietary recall (ASA24)

Participants logged into the Beta version of ASA24 website to complete their 24hr recall entries using accounts created by the NCI. Participants were reminded on assigned days by a computerized reminder system to complete the web-based recall to ensure their readiness and availability to do so that day. The USDA's Food and Nutrient Database for Dietary Studies (FNDDS 4.1) was used to derive the nutrient data.[15] A Daily Total Nutrients Analysis File which contains 61 nutrients and dietary constituents from all foods in a given day for each recall was

downloaded from the research website. Since the Beta version of ASA24 did not include supplement intake, we could only use ASA24 nutrients from food in our analyses.

E. Statistical Analyses

The performance of SFFQ2, which assesses intake over the same period as the 7DDR and ASA24 collection, was the primary focus of the current analyses. The validity of SFFQ1, which referred to the year before collection of the 7DDRs and ASA24s, and the WebFFQ were also evaluated. Intakes of total energy and 44 common nutrients available from the SFFQs, 7DDRs and ASA24s were used to evaluate the validity of SFFQs. Because nutritional supplements represent an important contribution to the intake of many micronutrients, we also evaluated 22 nutrients including supplements that were available from both the SFFQs and 7DDRs.

We first calculated means and standard deviations for absolute total daily nutrient intake from SFFQ1, SFFQ2, WebFFQ, averaged 7DDRs (mean = 14 days) and averaged ASA24s (mean = 2.9 days). Energy-adjusted intakes are of greatest importance because individuals primarily alter their intakes of specific nutrients by changing the composition of their diet, keeping total energy constant.[1,16,17] Therefore, we utilized two energy adjustment methods: residual method, which uses the residuals from the regression of the nutrients on total energy with reference energy level equals 1800 kcal and the energy density method, which divides the nutrient by total energy intake. Since most nutrient distributions were skewed toward higher values, we log transformed all variables after setting zeros to a fixed non-zero value (0.0001 unit/day), and performed residual method on the log scale .To further reduce the influence of the extreme nutrient intakes, analyses on correlations were based on the ranks of the log-transformed nutrient and energy-adjusted nutrient values.

To assess the reproducibility of repeated SFFQ, 7DDR, and ASA24, rank intraclass correlation coefficients (ICC) were calculated for each nutrient before and after adjustment for total energy intake.

Because the primary issue in epidemiologic studies is the ability of a method to rank subjects by intake of food or nutrients, we used rank correlation coefficients with true intake to assess validity of the SFFQ. In this analysis, the averaged 7DDR or ASA24 intakes, repeated at an interval of at least several months over a one-year period, were assumed to be unbiased estimates of the true underlying nutrient intakes, and were considered the 'gold standard'. Spearman correlation coefficients (r_s) and their 95% confidence intervals (95% CI) between nutrient intakes reported on each SFFQ and the corresponding intakes reported on the 7DDRs or ASA24s were also calculated. Random within-person variation in the 7DDRs or ASA24s can attenuate correlations between these measures and the SFFQs. Therefore, we calculated de-attenuated Spearman correlation coefficients to reduce the effect of random within person error in the reference methods. This method first calculates the within- and between- person components of variation in nutrient intakes of the reference methods,[18,19] and corrects correlation coefficients for the within-person variation by $r_c = r_0\sqrt{(1 + \gamma / k)}$, where r_0 represent the observed correlation coefficient, r_c represents the corrected correlation coefficient, and γ equals the ratio of estimated within-person variation and between-person variation, k is the number of repeated observations of the reference methods.[18,19]

Because the number of repeats of the reference method (k) differed across individuals, we used a previously described extension [20] of the methods by Rosner and Glynn [21] and Perisic and Rosner [22] to correct for random within-person measurement error in the presence of unbalanced data. In each strata of k, Spearman correlation coefficients and de-attenuated correlation coefficients[19]

corrected for random within-person variation in the probit score of nutrient ranks (nutrient ranks were calculated separately for each ASA24, and each 7DDR) were calculated. After Fisher's Z transformation[19], we obtained a weighted average of the Z transformed estimates and the 95% CIs. Next, we used inverse Fisher's Z and arcsin transformation to derive the Spearman correlation coefficients with or without de-attenuation.

The above analyses were conducted among the overall group of participants, and subgroups divided by age 45-60 years (N=309) and 61-80 years (N=323) and by BMI<25 kg/m2 (N=298) and>=25 kg/m2 (N=334) at WLVS enrollment. In addition, we evaluated the validity of SFFQs and ASAs in obese (BMI>=30 kg/m2) participants (N=127). We also calculated the de-attenuated correlation coefficients among a subgroup of participants with all four repeats of ASA24 (N=226). Regression calibration coefficients for each nutrient, which can be used for the correction of the relative risk estimates in regression models[23], were also derived from models predicting nutrient intakes based on7DDRs or ASA24s using intakes from each SFFQ.

III. Results

At baseline, participants had a mean age of 61 years and a mean BMI of 26.5 kg/m^2; participants were predominately white (90 %), and 2 % were current smokers. The subgroups had similar characteristics to the overall participants (Table 1.1).

The following results were mainly based on total energy and 44 common nutrients measured across SFFQ, 7DDR and ASA24, as well as 22 common nutrients with supplements measured in both the SFFQ and 7DDR. In addition, the results for several specific fatty acids, and carotenoids without appreciable intakes from supplements and total folate intake (11 nutrients without

supplements, and 5 nutrients with supplements) are shown in the supplementary materials (Supplementary Table 1.1a-c).

The distribution of mean daily nutrient intakes is described in Table 1.2. The absolute mean daily intakes estimated by each SFFQ, by two 7DDRs, and by four ASA24s were comparable among the 632 women included in this study. Most nutrients measured by the average of ASA24s and SFFQ had wider distributions compared to the average of two 7DDRs. Sodium intake was underestimated by SFFQ2 compared with 7DDR and ASA24. Intakes of energy, total protein, fat and carbohydrate assessed by SFFQ2 were comparable to those assessed by ASA24s, but tended to be slightly overestimated compared to the 7DDRs. Nutrients with supplements were overestimated by SFFQ2 compared to 7DDRs. SFFQ2 also tended to overestimate several nutrients contained in fruits and vegetables, such as carotenoids. This overestimation of intake by the SFFQ2 was primarily due to extreme values for a few individuals. The wide distribution of ASA24 was mainly due to the large day-to-day variations of food intakes.

As shown in Table 1.3, we observed a high degree of reproducibility for nutrient intakes by two paper SFFQs spaced one year apart, and by two 7DDRs separated by a six-months interval. The reproducibility of unadjusted nutrient intakes measured by two paper SFFQs was lowest for iron with supplements (ICC=0.50) and highest for alcohol intake (ICC=0.91). For unadjusted nutrients measured by two 7DDRs, ICCs ranged from 0.24 (lycopene) to 0.88 (caffeine). The reproducibility of the ASA24 over one year was much lower compared to the SFFQs and 7DDRs, with ICC = 0.10 for N-3 (DHA+EPA) fatty acids without supplements and 0.62 for caffeine intake. The mean ICCs of the unadjusted nutrient intakes assessed by SFFQs, 7DDRs and ASA24s were 0.68, 0.59 and 0.25, respectively. Lower ICCs for most nutrients measured by ASA24 were due to large within-person variation in nutrient intake over a one-year interval. The

average reproducibility of nutrient intakes was slightly lower after adjustment for total energy intake for all dietary methods.

When using 7DDRs as the reference method, the Spearman correlation coefficients between the unadjusted nutrient intakes from SFFQ2, which assesses intake over the same period as the 7DDR collection, and those from the two 7DDRs ranged from 0.28 for total energy, polyunsaturated fat and sodium to 0.86 for alcohol (Table 1.4). Results were similar when Pearson correlation coefficients were used (Supplementary Table 1.2). The two energy-adjusted methods gave similar results and energy-adjustment improved the correlation between the SFFQ and 7DDRs for many macronutrients, but not all nutrients; the following results were focused on energy-adjusted results using residual-method. Energy-adjusted correlations ranged from 0.36 for lauric acid to 0.77 for alcohol, mean r =0.53 (mean r=0.57 after additionally including nutrients with supplements). After correcting for random within-person error in the 7DDRs, the mean Spearman correlation coefficient increased to 0.63 (mean r=0.66 after additionally including nutrients with supplements), the lowest and highest correlations were 0.46 for lauric acid to 0.84 for alcohol intake (Table 1.4 & Table 1.5). When comparing the 22 common nutrients with supplements, the correlations between SFFQ2 and 7DDR were generally higher after including supplements in those nutrients, mean de-attenuated r=0.71 (mean r=0.63 if supplements were not included) (Table 5). Similar patterns of de-attenuated correlations with 7DDR and SFFQ2 were observed for the WebFFQ (mean r=0.61) and SFFQ1 (mean r=0.60) (Supplementary Table 1.3 & 1.4).

When the ASA24s were used as the comparison method, the unadjusted Spearman correlation coefficients for nutrients assessed by SFFQ2 compared four ASA24s were lowest for lycopene without supplements (r=0.23) and highest for alcohol (r=0.75). Energy-adjustment increased

correlations compared to unadjusted values for many nutrients. Correlation coefficients increased substantially after de-attenuation (mean r: 0.62 vs. 0.43). The confidence intervals of the correlations of SFFQ2 with ASA24 were somewhat wider compared to the correlations with 7DDR after de-attenuation even though the study sample size was large. For example, the de-attenuated Spearman correlations for cholesterol intake were 0.65 (95% CI=0.58-0.71) with 7DDR, and 0.68 (95% CI=0.55-0.79) with ASA24 (Table 1.4). Because the degree of correction depends on the ratio of within- to between-person variation in the ASA24, the effect of de-attenuation for nutrients with very high within-person variation (as reflected by a low ICC) was large and the confidence intervals of the corrected correlation were also generally wide. For example, the reproducibility of polyunsaturated fat (ICC=0.12) and N-3 (DHA+EPA) fatty acids without supplements (ICC=0.08) assessed by ASA24 was relatively low, therefore the de-attenuated correlations increased greatly, and the corresponding confidence intervals were wide (Table 1.4). Slightly lower magnitudes of correlations with ASA24 were observed with SFFQ1 (mean de-attenuated r= 0.57) and with the WebFFQ (mean de-attenuated r= 0.60) (Supplementary Table 1.3 & 1.4).

The Spearman correlation coefficients showed similar patterns for the subgroup of participants with all four ASA24 assessments (Supplementary Table 1.5), and among different age groups (Supplementary Tables 1.6-1.7). For participants with BMI<25 kg/m2, the mean adjusted and de-attenuated correlation coefficient (mean r=0.65 vs 7DDR, and 0.62 vs ASA24) was slightly higher compared to the subgroup with BMI>=25 kg/m^2 (mean r=0.61 vs 7DDR, and 0.61 vs ASA24) or subgroup with BMI>=30 kg/m^2 (mean r=0.59 vs 7DDR, and 0.57 vs ASA24), regardless of which comparison method was used (Supplementary Tables 1.8-1.10). Supplementary Table 1.11 showed similar regression coefficients derived from models when

each SFFQ nutrient intake was used to predict intake estimated by 7DDR or ASA24, respectively.

IV. Discussion

We evaluated the performance of both paper and web versions of our semi-quantitative SFFQ by comparing nutrient intakes from each SFFQ with those measured by two 7DDRs, and by four ASA24s kept over a one-year period among women aged 45-80 years participating in prospective cohort studies. The correlations were generally stronger when nutrient intakes were adjusted for total energy intake and when supplements were included into nutrient values. As a comparison method, after adjustment for within-person variation, the ASA24s performed similarly to the 7DDRs. In subgroup analysis, the validity of our SFFQs was found to be similar among different age groups but slightly lower among obese than in normal weight women. Similar patterns of correlations were observed when comparing SFFQ1, SFFQ2 and WebFFQ to the 7DDRs or ASA24s, although, as expected, the correlations were slightly lower for SFFQ1 because this referred to the year before collection of the 7DDRs and ASA24s.

Many validation studies have been conducted within large cohorts, comparing intakes from the study's SFFQ with intakes from dietary records or 24-hour recalls.[1,3,4,11,24-28] In general, correlations between nutrients calculated from SFFQs and from multiple food records or diet recalls ranged from 0.40 to 0.70 [1], which suggests considerable error but still sufficient information to detect important hypothesized associations with disease. Among those studies, our team has conducted two validation studies among Boston-area women in the NHS in 1980 (N=173) [3] and 1986 (N=191) [1] by comparing SFFQs with multiple 7-day weighed dietary records. The 1980 study evaluated the 61-item semi-quantitative SFFQ among 225 women of age 34-59 years. Correlation coefficients between energy-adjusted intakes from the four one-

week diet records and those from the second SFFQ ranged from 0.36 for vitamin A without supplements to 0.75 for vitamin C with supplements (mean r=0.55). The 1986 study evaluated an expanded version of the questionnaire with 116 items. After de-attenuation for week-to-week variation in diet records, the correlations between the 1986 diet records and the 1986 SFFQ reflecting the diet record year ranged from 0.48 for polyunsaturated fat to 0.79 for total vitamin A without supplements (mean r=0.54 and mean r=0.64 after further de-attenuation). The current study evaluated additional nutrients and enrolled more participants. The de-attenuated correlation coefficients between the energy-adjusted intakes assess by SFFQ2 and two 7DDRs ranged from 0.46 for lauric acid to 0.84 for alcohol (mean r=0.63 and mean r=0.66 after additionally including nutrients with supplements). Overall, those studies indicate that our SFFQ performed consistently well when compared with multiple diet records over 30 years, and that modifications to the questionnaire over time have adequately taken into account the many changes in the food supply and eating patterns that have occurred since 1980.

In addition, our study also suggest that after adjustment for within-person variation, the ASA24s used in our study performed similarly to the 7DDRs as a comparison method when using mean values, de-attenuated correlation coefficients, and regression coefficients to assess validity. Since day-to-day variability in the intake of many nutrients is large, four ASA24s every three months over a year to estimate the true long term intake may still be insufficient to measure accurately intake of some nutrients, especially if used to assess validity on an individual basis. [29] The ASA24 is designed to address the limitations in traditional 24-h dietary recalls, which rely on trained interviewers with higher cost and limited utility in large studies. In a study assessing meals for one day, the ASA24 agreed well with a measure of true intakes in terms of portion size, food group intake, energy and nutrient intakes, although the accuracy was slightly less compared

an interviewer-administered 24-hr recall.[30] Thus, the ASA24 has the potential to be utilized as a comparison methods in future validation studies if adjustment is made for day to day variation ; its potential to be used as a dietary assessment method for large population studies needs further evaluation, including the number of ASA24s needed, and the appropriate time interval between each measurement to be representative of long-term dietary intake.

To the best of our knowledge, this is the largest validation study on dietary assessment methods among women, but it is not without limitations. Study participants were registered female nurses from the NHS and NHS II cohorts, thus our result may not be generalized to the general population or to men. It should be noted that in earlier validation studies among a variety of populations, the validity of SFFQ assessments are broadly similar.[1] Furthermore, errors in dietary assessments with the SFFQ, diet records, and 24h-hr dietary recalls may be correlated if so the validity of SFFQ will be over-estimated. However, many sources of error in dietary assessment by diet records and 24-hour recalls are likely to be uncorrelated, as they depend very differently on memory and assessment of serving sizes; the similarity of their correlations with SFFQ is reassuring. Also, recalls and diet records both have their own sources of error, which would lead to underestimation of validity for the SFFQ.

In conclusion, our data indicates that the 152-item SFFQ appears is reasonably valid and consistent for measuring nutrient intakes compared to multiple dietary records or 24-hr dietary recalls among women. If within-person variation is taken into account, the ASA24 and 7DDRs provided comparable information when serving as the comparison method for assessing validity of the SFFQ, which has important implications for the design of future validation studies as the cost of the ASA24 is minor compared to 7DDRs. Further studies could utilize web-based 24-hr recalls to validate change in diet as measured by food frequency questionnaires, and determine

optimal dietary measurement methods that potentially combine both SFFQ's and 24-hr recall approaches.

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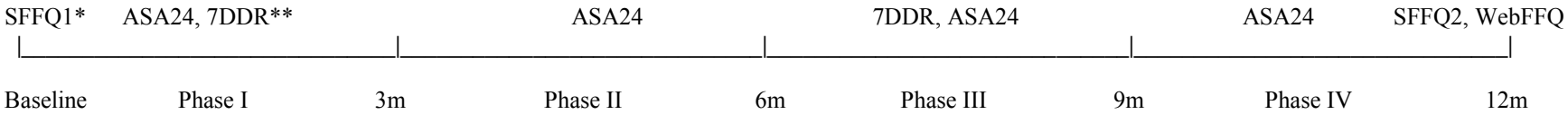
VI. Figure and Tables

Figure 1. Timeline of the dietary assessment activity in project 1.



* This figure showed the timeline for group 1 participants; group 3 went through the same data collection timeline as group 1; groups 2 and 4 went through similar data collection timeline as group 1, except Groups 2 and 4 participants completed the 7DDR in phase II and IV.
**Within the same phase, 7DDRs and ASA24s, SFFQ2 and WebFFQ were collected 2-5 weeks apart from each other and in random order to avoid artificially high correlations.

Table 1.1. Subject characteristics among WLVS participants in project 1

Variables	All participants (n=632)	Subgroups				
		Four ASA24s (n=226)	Age (45-60 years) (n=309)	Age (61-80 years) (n=323)	BMI<25 kg/m^2 (n=298)	BMI>=25 kg/m^2 (n=334)
Age, mean (sd), year	61 (10)	61 (9)	53 (5)	69 (5)	61 (10)	61 (9)
Height, mean (sd), m	1.64 (0.07)	1.64 (0.07)	1.66 (0.07)	1.63 (0.07)	1.65 (0.08)	1.63 (0.07)
Weight, mean (sd), kg	71.6 (15.5)	71.4 (15.7)	73.6 (17.2)	69.6 (13.3)	60.8 (6.7)	81.1 (14.7)
Weight change, mean (sd), kg	-0.2 (2.7)	-0.1 (2.7)	-0.2 (2.9)	-0.2 (2.4)	-0.2 (1.8)	-0.2 (3.2)
BMI, mean(sd), kg/m2	26.5 (5.4)	26.5 (5.4)	26.8 (6.0)	26.2 (4.8)	22.3 (1.8)	30.3 (4.8)
White, %	90.4	90.7	86.1	94.4	94.0	87.1
Current smokers, %	1.9	2.7	2.6	1.3	1.4	2.4
Total energy (kcal/day)						
SFFQ2	1857 (526)	1814 (490)	1864 (530)	1852 (523)	1846 (486)	1868 (559)
7DDR	1741 (336)	1738 (329)	1788 (344)	1695 (321)	1714 (302)	1765 (362)
ASA24	1821 (476)	1801 (389)	1846 (493)	1797 (459)	1792 (421)	1846 (520)
Protein (% total energy)						
SFFQ2	17.5 (3.0)	17.8 (2.9)	17.8 (3.1)	17.2 (2.8)	17.4 (2.8)	17.7 (3.1)
7DDR	16.9 (2.9)	16.9 (2.9)	16.8 (2.9)	16.9 (2.8)	16.8 (2.8)	16.9 (2.9)
ASA24	17.4 (3.8)	17.2 (3.4)	17.6 (3.9)	17.1 (3.8)	17.2 (3.5)	17.6 (4.1)
Total fat (% total energy)						
SFFQ2	33.9 (6.1)	33.8 (6.1)	34.2 (5.8)	33.5 (6.3)	33.3 (6.3)	34.4 (5.9)
7DDR	34.4 (5.7)	34.2 (5.7)	35.0 (5.9)	33.9 (5.4)	33.9 (5.5)	35.0 (5.8)
ASA24	35.3 (7.0)	34.9 (5.9)	35.4 (6.4)	35.1 (7.5)	34.9 (7.0)	35.6 (7.0)
Carbohydrates (% total energy)						
SFFQ2	47.3 (7.5)	47.1 (7.1)	46.9 (7.2)	47.6 (7.7)	47.8 (7.6)	46.8 (7.2)
7DDR	47.8 (7.6)	48.3 (7.5)	47.9 (7.5)	47.8 (7.7)	48.3 (7.6)	47.5 (7.6)
ASA24	45.5 (8.7)	46.2 (7.6)	45.6 (8.5)	45.4 (8.9)	45.7 (8.7)	45.3 (8.8)

Note: data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012.
SFFQ, The semi-quantitative food frequency questionnaire
7DDR, 7 Day Dietary Records
ASA24, Web-based, self-administered 24-hour dietary recall

Table 1.2. Mean absolute daily nutrient intakes estimated by SFFQs, 7DDRs, and ASA24s

Nutrient	SFFQ2	SFFQ1	WebFFQ	7DDRs*	ASA24s**
	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)
Total energy (kcal)	1857 (526)	1931 (528)	1770 (518)	1741 (336)	1821 (476)
Total fat (g)	70 (25)	73 (26)	67 (24)	67 (19)	73 (27)
Saturated fat (g)	22 (9)	23 (9)	21 (8)	22 (8)	25 (11)
Polyunsaturated fat (g)	15 (6)	16 (6)	14 (6)	15 (5)	16 (7)
Monounsaturated fat (g)	27 (11)	28 (11)	26 (11)	24 (7)	26 (11)
Arachadonic FA (g)	0.18 (0.09)	0.19 (0.09)	0.18 (0.09)	0.11 (0.06)	0.13 (0.08)
Lauric FA (g)	0.63 (0.59)	0.66 (0.61)	0.63 (0.59)	1.04 (0.89)	0.84 (0.89)
Linoleic FA (g)	12.5 (5.1)	13.1 (5.5)	11.8 (5.4)	13.1 (4.3)	13.8 (6.2)
Linolenic FA (g)	1.5 (0.9)	1.6 (1.0)	1.5 (1.1)	1.5 (0.7)	1.6 (1)
N-3 (DHA+EPA) FA (g)	0.25 (0.22)	0.26 (0.23)	0.24 (0.21)	0.14 (0.15)	0.18 (0.31)
With supplements	0.47 (0.38)	0.49 (0.40)	0.47 (0.38)	0.32 (0.42)	
Oleic FA (g)	25.0 (10.1)	26.0 (10.7)	24.1 (10.2)	22.8 (6.8)	24.5 (10.1)
Cholesterol (mg)	238 (114)	241 (107)	229 (116)	233 (94)	269 (142)
Protein (g)	81 (24)	84 (24)	77 (24)	73 (16)	77 (24)
Carbohydrate (g)	219 (72)	229 (71)	208 (69)	208 (50)	205 (64)
Total sugar (g)	100 (43)	103 (42)	94 (40)	92 (32)	97 (41)
Fiber (g)	23.7 (8.6)	24.8 (8.6)	22.9 (8.5)	20.5 (6.5)	17.5 (7.0)
Alcohol (g)	8.9 (12.5)	9.3 (12.4)	8.3 (11.1)	9.0 (12.0)	10.1 (15.5)
Retinol activity eqvts (mcg)	995 (406)	1029 (431)	971 (411)	813 (388)	762 (482)
With supplements	1805 (1299)	1879 (1421)	1830 (1349)	1477 (1293)	
Alpha carotene (mcg)	852 (763)	919 (871)	850 (767)	609 (487)	495 (769)
Beta carotene (mcg)	5974 (3611)	6153 (3761)	5882 (3574)	3979 (2470)	3335 (3307)
With supplements	6489 (4046)	6752 (4316)	6368 (3972)	4291 (2818)	
Lutein-zeaxanthin (mcg)	3794 (2898)	3749 (2539)	3725 (2513)	2237 (1517)	2589 (2931)
Lycopene (mcg)	5479 (3970)	5618 (4454)	5596 (4585)	4870 (3543)	5088 (5402)
Beta cryptoxanthin (mcg)	111 (95)	120 (103)	107 (95)	154 (181)	99 (137)
Vitamin B1 (mg)	1.5 (0.5)	1.6 (0.5)	1.4 (0.4)	1.5 (0.4)	1.4 (0.5)
With supplements	8.6 (19.4)	8.8 (18.0)	7.9 (15.6)	11.0 (51.3)	
Vitamin B2 (mg)	2.2 (0.8)	2.3 (0.8)	2.1 (0.7)	2.0 (0.6)	2.2 (0.7)
With supplements	9.2 (17.3)	9.7 (17.6)	8.9 (15.9)	7.2 (18.0)	
Vitamin B3 (mg)	23.5 (7.1)	24.6 (7.5)	22.6 (7.2)	20.7 (5.5)	20.9 (6.9)
With supplements	49.3 (57.4)	53.7 (61.0)	47.1 (49.9)	54.4 (114)	
Vitamin B6 (mg)	2.2 (0.8)	2.3 (0.7)	2.1 (0.7)	1.8 (0.6)	1.9 (0.8)
With supplements	13.5 (28.3)	15.2 (32.2)	14.0 (29.6)	9.6 (26.8)	
Vitamin B12 (mg)	6.3 (2.8)	6.5 (2.7)	6.1 (2.6)	4.9 (2.7)	5.4 (4.7)
With supplements	55.2 (141.7)	61.0 (145.2)	51.2 (125.0)	73.1 (307.8)	

Table 1.2 (continued): Mean absolute daily nutrient intakes estimated by SFFQs, 7DDRs, and ASA24s

Nutrient	SFFQ2	SFFQ1	WebFFQ	7DDRs*	ASA24s**
	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)
Natural folate (mcg)	319 (122)	331 (123)	310 (119)	232 (73)	233 (95)
Folic acid (mcg)	139 (115)	147 (109)	133 (92)	156 (99)	143 (111)
With supplements	424 (312)	442 (294)	412 (280)	438 (355)	
Dietary folate eqvts (mcg)	641 (263)	669 (241)	620 (218)	497 (182)	477 (212)
With supplements	1126 (647)	1170 (595)	1095 (570)	1062 (713)	
Vitamin C (mg)	118 (60)	123 (61)	116 (62)	96 (51)	94 (67)
With supplements	311 (426)	325 (375)	319 (366)	311 (453)	
Vitamin D (mg)	5.3 (3.1)	5.5 (3.1)	5.2 (3.0)	4.9 (2.7)	5.1 (3.9)
With supplements	20.4 (12.2)	21.5 (12.5)	23.6 (12.7)	31.5 (29.3)	
Vitamin E (mg)	9.9 (4.8)	10.3 (4.9)	9.5 (4.9)	9.2 (4.0)	8.2 (4.4)
With supplements	42.1 (71.7)	43.5 (67.5)	45.6 (83.8)	37.1 (54.0)	
Vitamin K (mg)	195 (142)	191 (121)	190 (125)	129 (71)	156 (183)
With supplements	202 (143)	199 (122)	197 (126)	219 (1806)	
Calcium (mg)	909 (371)	954 (388)	880 (381)	843 (270)	959 (421)
With supplements	1572 (638)	1634 (641)	1577 (648)	1460 (799)	
Magnesium (mg)	351 (106)	369 (112)	338 (106)	296 (77)	309 (103)
With supplements	425 (153)	446 (159)	414 (145)	380 (186)	
Iron (mg)	14.1 (4.7)	14.7 (5.0)	13.4 (4.6)	13.7 (4.5)	13.8 (4.9)
With supplements	18.4 (10.1)	19.6 (10.6)	20 (10.7)	18.9 (12.3)	
Copper (mg)	1.5 (0.5)	1.6 (0.5)	1.5 (0.5)	1.3 (0.5)	1.4 (0.8)
With supplements	2.5 (1.3)	2.6 (1.3)	2.5 (1.3)	2.1 (1.1)	
Zinc (mg)	11.9 (3.7)	12.6 (3.9)	11.5 (3.7)	10.3 (3.2)	11.5 (4.6)
With supplements	21.7 (14.0)	22.5 (14.7)	22.4 (14.8)	19.9 (11.8)	
Phosphorus (mg)	1360 (414)	1425 (422)	1300 (415)	1187 (274)	1325 (409)
With supplements	1391 (418)	1458 (427)	1339 (418)	1209 (275)	
Choline (mg)	343 (110)	353 (105)	330 (111)	295 (79)	313 (105)
With supplements	348 (115)	358 (107)	335 (113)	298 (85)	
Potassium (mg)	3251 (965)	3389 (973)	3143 (965)	2632 (656)	2801 (868)
With supplements	3298 (976)	3437 (987)	3190 (972)	2670 (665)	
Sodium (mg)	2061 (660)	2152 (685)	1947 (638)	2647 (640)	3087 (913)
Caffeine (mg)	179 (139)	185 (136)	168 (129)	164 (112)	167 (134)

Note: data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012.
SD, standard deviation; FA, fatty acid; Eqvts, equivalents.
*Average of two 7DDRs;
**Average of four ASA24s.

Table 1.3. Rank intraclass correlations of daily nutrient intake from SFFQ, 7DDR, and ASA24

Nutrient	SFFQ*			7DDR*			ASA24*		
	Un-adjusted	Energy density	Residual method	Un-adjusted	Energy density	Residual method	Un-adjusted	Energy density	Residual method
Total energy (kcal)	0.70			0.64			0.29		
Total fat (g)	0.69	0.68	0.68	0.62	0.64	0.65	0.26	0.23	0.23
Saturated fat (g)	0.71	0.69	0.69	0.65	0.65	0.65	0.28	0.26	0.25
Polyunsaturated fat (g)	0.68	0.65	0.65	0.51	0.48	0.49	0.19	0.13	0.12
Monounsaturated fat (g)	0.67	0.64	0.64	0.60	0.58	0.58	0.26	0.22	0.21
Arachadonic FA (g)	0.66	0.63	0.62	0.55	0.51	0.51	0.17	0.16	0.16
Lauric FA (g)	0.67	0.66	0.66	0.48	0.44	0.43	0.24	0.22	0.21
Linoleic FA (g)	0.66	0.63	0.63	0.50	0.48	0.49	0.19	0.13	0.13
Linolenic FA (g)	0.69	0.66	0.66	0.50	0.44	0.44	0.21	0.14	0.13
N-3 (DHA+EPA) FA (g)	0.69	0.67	0.67	0.42	0.41	0.41	0.10	0.09	0.08
With supplements	0.77	0.74	0.73	0.63	0.61	0.63			
Oleic FA (g)	0.66	0.64	0.65	0.60	0.58	0.58	0.25	0.21	0.20
Cholesterol (mg)	0.69	0.62	0.62	0.61	0.58	0.58	0.20	0.16	0.16
Protein (g)	0.66	0.63	0.62	0.65	0.60	0.61	0.25	0.22	0.21
Carbohydrate (g)	0.72	0.73	0.73	0.69	0.74	0.74	0.36	0.34	0.34
Total sugar (g)	0.73	0.78	0.78	0.73	0.75	0.75	0.37	0.37	0.37
Fiber (g)	0.71	0.71	0.71	0.73	0.74	0.75	0.35	0.32	0.33
Alcohol (g)	0.91	0.91	0.86	0.81	0.81	0.73	0.47	0.49	0.46
Retinol activity eqvts (mcg)	0.65	0.65	0.64	0.42	0.37	0.37	0.22	0.20	0.20
With supplements	0.68	0.69	0.68	0.65	0.64	0.65			
Alpha carotene (mcg)	0.69	0.65	0.65	0.37	0.37	0.37	0.15	0.14	0.14
Beta carotene (mcg)	0.70	0.66	0.67	0.45	0.43	0.44	0.22	0.20	0.20
With supplements	0.70	0.67	0.67	0.47	0.46	0.46			
Lutein-zeaxanthin (mcg)	0.71	0.69	0.69	0.51	0.49	0.50	0.23	0.21	0.21
Lycopene (mcg)	0.64	0.57	0.57	0.24	0.23	0.23	0.11	0.11	0.11
Beta cryptoxanthin (mcg)	0.74	0.72	0.72	0.31	0.31	0.31	0.19	0.17	0.15
Vitamin B1 (mg)	0.65	0.63	0.62	0.55	0.53	0.52	0.20	0.19	0.19
With supplements	0.68	0.68	0.67	0.72	0.71	0.71			
Vitamin B2 (mg)	0.67	0.71	0.70	0.70	0.67	0.69	0.31	0.31	0.31
With supplements	0.67	0.68	0.68	0.75	0.74	0.75			
Vitamin B3 (mg)	0.64	0.62	0.59	0.57	0.55	0.54	0.17	0.19	0.17
With supplements	0.69	0.71	0.70	0.72	0.71	0.72			
Vitamin B6 (mg)	0.63	0.62	0.60	0.59	0.57	0.58	0.22	0.20	0.19
With supplements	0.68	0.70	0.68	0.73	0.71	0.72			
Vitamin B12 (mg)	0.59	0.59	0.58	0.50	0.48	0.48	0.13	0.12	0.11
With supplements	0.67	0.65	0.65	0.72	0.72	0.72			
Natural folate (mcg)	0.71	0.67	0.67	0.67	0.63	0.65	0.34	0.31	0.33

Table 1.3 (continued): Rank intraclass correlations of daily nutrient intake from SFFQ, 7DDR, ASA24

Nutrient	SFFQ*			7DDR*			ASA24*		
	Un-adjusted	Energy density	Residual method	Un-adjusted	Energy density	Residual method	Un-adjusted	Energy density	Residual method
Folic acid (mcg)	0.59	0.60	0.60	0.59	0.59	0.59	0.22	0.22	0.23
With supplements	0.64	0.65	0.64	0.72	0.71	0.72			
Dietary folate eqvts (mcg)	0.59	0.58	0.57	0.59	0.56	0.56	0.23	0.20	0.20
With supplements	0.65	0.65	0.64	0.74	0.71	0.73			
Vitamin C (mg)	0.71	0.68	0.68	0.59	0.56	0.57	0.30	0.27	0.28
With supplements	0.72	0.71	0.71	0.73	0.73	0.73			
Vitamin D (mg)	0.68	0.67	0.67	0.61	0.57	0.58	0.22	0.20	0.20
With supplements	0.56	0.56	0.54	0.72	0.72	0.72			
Vitamin E (mg)	0.65	0.64	0.64	0.55	0.53	0.53	0.26	0.23	0.23
With supplements	0.71	0.73	0.72	0.78	0.77	0.77			
Vitamin K (mg)	0.71	0.69	0.69	0.50	0.48	0.48	0.20	0.17	0.17
With supplements	0.71	0.68	0.68	0.54	0.53	0.53			
Calcium (mg)	0.64	0.65	0.64	0.71	0.68	0.70	0.30	0.25	0.26
With supplements	0.69	0.72	0.71	0.72	0.72	0.73			
Magnesium (mg)	0.68	0.68	0.67	0.72	0.72	0.73	0.37	0.35	0.37
With supplements	0.67	0.70	0.69	0.78	0.78	0.80			
Iron (mg)	0.62	0.56	0.55	0.59	0.54	0.54	0.21	0.19	0.19
With supplements	0.50	0.48	0.46	0.51	0.50	0.50			
Copper (mg)	0.65	0.69	0.67	0.62	0.60	0.60	0.28	0.25	0.25
With supplements	0.60	0.64	0.62	0.64	0.65	0.64			
Zinc (mg)	0.60	0.60	0.56	0.58	0.48	0.49	0.15	0.12	0.11
With supplements	0.61	0.61	0.60	0.67	0.66	0.66			
Phosphorus (mg)	0.67	0.68	0.67	0.71	0.65	0.68	0.29	0.24	0.24
With supplements	0.67	0.68	0.66	0.71	0.66	0.68			
Choline (mg)	0.69	0.62	0.62	0.68	0.61	0.63	0.24	0.19	0.19
With supplements	0.69	0.63	0.63	0.67	0.60	0.62			
Potassium (mg)	0.71	0.68	0.68	0.74	0.70	0.73	0.37	0.33	0.36
With supplements	0.72	0.69	0.69	0.75	0.70	0.73			
Sodium (mg)	0.73	0.68	0.69	0.58	0.50	0.50	0.24	0.20	0.20
Caffeine (mg)	0.78	0.78	0.78	0.88	0.87	0.86	0.62	0.57	0.50

Note: data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012.
*Two SFFQs, one year apart; Two 7DDRs, 6 months apart; Four ASA24s, every three months over one year.

Table 1.4. Spearman correlation coefficient for comparison of SFFQ2 with 7DDRs, and ASA24s

Nutrient	SFFQ2 vs. 7DDR						SFFQ2 vs. ASA24					
	Un-adjusted	Energy- adjusted					Un-adjusted	Energy- adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC *		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC *
Total energy (kcal)	0.28			0.31	(0.23, 0.38)	0.64	0.30			0.40	(0.31, 0.49)	0.29
Total fat (g)	0.36	0.59	0.59	0.67	(0.60, 0.71)	0.65	0.34	0.55	0.55	0.76	(0.64, 0.84)	0.23
Saturated fat (g)	0.44	0.62	0.61	0.69	(0.62, 0.73)	0.65	0.44	0.57	0.54	0.71	(0.60, 0.80)	0.25
Polyunsaturated fat (g)	0.28	0.45	0.47	0.57	(0.49, 0.64)	0.49	0.24	0.40	0.41	0.70	(0.52, 0.83)	0.12
Monounsaturated fat (g)	0.37	0.48	0.48	0.56	(0.49, 0.63)	0.58	0.30	0.40	0.40	0.60	(0.48, 0.69)	0.21
Arachadonic FA (g)	0.36	0.44	0.43	0.52	(0.44, 0.59)	0.51	0.25	0.33	0.32	0.52	(0.38, 0.63)	0.16
Lauric FA (g)	0.35	0.37	0.36	0.46	(0.37, 0.54)	0.43	0.34	0.34	0.33	0.48	(0.37, 0.58)	0.21
Linoleic FA (g)	0.29	0.44	0.45	0.55	(0.47, 0.62)	0.49	0.25	0.39	0.41	0.70	(0.52, 0.82)	0.13
Linolenic FA (g)	0.35	0.44	0.45	0.57	(0.48, 0.65)	0.44	0.27	0.33	0.34	0.58	(0.42, 0.70)	0.13
N-3 (DHA+EPA) FA (g)	0.53	0.53	0.52	0.67	(0.58, 0.74)	0.41	0.27	0.30	0.29	0.64	(0.40, 0.80)	0.08
With supplements	0.62	0.62	0.60	0.69	(0.62, 0.74)	0.63						
Oleic FA (g)	0.37	0.48	0.48	0.56	(0.48, 0.62)	0.58	0.30	0.40	0.40	0.60	(0.48, 0.69)	0.20
Cholesterol (mg)	0.50	0.56	0.56	0.65	(0.58, 0.71)	0.58	0.39	0.42	0.42	0.68	(0.55, 0.79)	0.16
Protein (g)	0.33	0.48	0.48	0.54	(0.47, 0.60)	0.61	0.30	0.38	0.37	0.54	(0.42, 0.64)	0.21
Carbohydrate (g)	0.41	0.66	0.65	0.69	(0.64, 0.73)	0.74	0.44	0.58	0.58	0.73	(0.65, 0.80)	0.34
Total sugar (g)	0.53	0.68	0.68	0.74	(0.69, 0.79)	0.75	0.54	0.64	0.63	0.75	(0.67, 0.81)	0.37
Fiber (g)	0.46	0.64	0.62	0.66	(0.60, 0.70)	0.75	0.41	0.52	0.51	0.65	(0.56, 0.72)	0.33
Alcohol (g)	0.86	0.86	0.77	0.84	(0.80, 0.87)	0.73	0.75	0.75	0.65	0.74	(0.68, 0.80)	0.46
Retinol activity eqvts (mcg)	0.45	0.48	0.49	0.66	(0.56, 0.74)	0.37	0.39	0.40	0.42	0.60	(0.48, 0.70)	0.20
With supplements	0.64	0.65	0.66	0.74	(0.68, 0.79)	0.65						
Alpha carotene (mcg)	0.49	0.48	0.47	0.64	(0.54, 0.72)	0.37	0.39	0.37	0.37	0.65	(0.50, 0.75)	0.14
Beta carotene (mcg)	0.51	0.53	0.52	0.66	(0.58, 0.73)	0.44	0.44	0.44	0.44	0.67	(0.55, 0.75)	0.20
With supplements	0.50	0.50	0.50	0.63	(0.54, 0.69)	0.46						
Lutein-zeaxanthin (mcg)	0.54	0.56	0.55	0.68	(0.60, 0.74)	0.50	0.44	0.47	0.47	0.70	(0.58, 0.79)	0.21

Table 1.4 (continued): Spearman correlation coefficient for comparison of SFFQ2 with 7DDRs, and ASA24s

Nutrient	SFFQ2 vs. 7DDR						SFFQ2 vs. ASA24					
	Un-adjusted	Energy intake adjusted					Un-adjusted	Energy intake adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC *		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Lycopene (mcg)	0.40	0.38	0.37	0.61	(0.46, 0.72)	0.23	0.23	0.20	0.15	0.29	(0.14, 0.43)	0.11
Beta cryptoxanthin (mcg)	0.36	0.38	0.38	0.54	(0.43, 0.64)	0.31	0.39	0.41	0.42	0.59	(0.44, 0.70)	0.15
Vitamin B1 (mg)	0.36	0.53	0.54	0.65	(0.57, 0.71)	0.52	0.32	0.42	0.42	0.65	(0.52, 0.74)	0.19
With supplements	0.70	0.71	0.71	0.78	(0.73, 0.82)	0.71						
Vitamin B2 (mg)	0.52	0.57	0.61	0.67	(0.62, 0.72)	0.69	0.46	0.50	0.53	0.69	(0.60, 0.77)	0.31
With supplements	0.71	0.71	0.73	0.79	(0.74, 0.83)	0.75						
Vitamin B3 (mg)	0.36	0.46	0.46	0.55	(0.47, 0.61)	0.54	0.29	0.40	0.38	0.59	(0.45, 0.69)	0.17
With supplements	0.67	0.70	0.71	0.78	(0.73, 0.82)	0.72						
Vitamin B6 (mg)	0.38	0.48	0.48	0.56	(0.49, 0.63)	0.58	0.33	0.38	0.38	0.57	(0.44, 0.67)	0.19
With supplements	0.68	0.67	0.68	0.74	(0.69, 0.79)	0.72						
Vitamin B12 (mg)	0.43	0.44	0.45	0.56	(0.48, 0.63)	0.48	0.32	0.31	0.31	0.56	(0.39, 0.69)	0.11
With supplements	0.69	0.67	0.68	0.74	(0.69, 0.79)	0.72						
Natural folate (mcg)	0.48	0.59	0.59	0.67	(0.61, 0.72)	0.65	0.40	0.51	0.50	0.64	(0.55, 0.71)	0.33
Folic acid (mcg)	0.56	0.59	0.59	0.69	(0.62, 0.74)	0.59	0.45	0.46	0.44	0.57	(0.46, 0.67)	0.23
With supplements	0.68	0.69	0.69	0.76	(0.71, 0.80)	0.72						
Dietary folate eqvts (mcg)	0.45	0.54	0.55	0.64	(0.57, 0.70)	0.56	0.36	0.39	0.40	0.56	(0.44, 0.66)	0.20
With supplements	0.68	0.70	0.71	0.77	(0.72, 0.81)	0.73						
Vitamin C (mg)	0.49	0.53	0.52	0.61	(0.54, 0.67)	0.57	0.47	0.46	0.46	0.62	(0.52, 0.70)	0.28
With supplements	0.70	0.71	0.71	0.77	(0.72, 0.82)	0.73						
Vitamin D (mg)	0.60	0.58	0.59	0.69	(0.62, 0.74)	0.58	0.51	0.51	0.52	0.77	(0.64, 0.85)	0.20
With supplements	0.63	0.59	0.60	0.66	(0.60, 0.71)	0.72						
Vitamin E (mg)	0.40	0.47	0.47	0.56	(0.48, 0.62)	0.53	0.29	0.36	0.35	0.51	(0.39, 0.60)	0.23
With supplements	0.72	0.72	0.72	0.78	(0.74, 0.82)	0.77						
Vitamin K (mg)	0.53	0.58	0.57	0.71	(0.63, 0.78)	0.48	0.37	0.41	0.41	0.64	(0.51, 0.73)	0.17
With supplements	0.53	0.56	0.56	0.67	(0.59, 0.73)	0.53						

Table 1.4 (continued): Spearman correlation coefficient for comparison of SFFQ2 with 7DDRs, and ASA24s

Nutrient	SFFQ2 vs. 7DDR						SFFQ2 vs. ASA24					
	Un-adjusted	Energy intake adjusted					Un-adjusted	Energy intake adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC *		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC *
Calcium (mg)	0.56	0.61	0.63	0.68	(0.62, 0.73)	0.70	0.48	0.54	0.55	0.68	(0.57, 0.77)	0.26
With supplements	0.66	0.64	0.68	0.74	(0.69, 0.79)	0.73						
Magnesium (mg)	0.43	0.65	0.66	0.72	(0.67, 0.77)	0.73	0.37	0.59	0.58	0.73	(0.65, 0.79)	0.37
With supplements	0.58	0.72	0.73	0.75	(0.70, 0.80)	0.80						
Iron (mg)	0.35	0.48	0.48	0.56	(0.49, 0.63)	0.54	0.30	0.39	0.37	0.55	(0.43, 0.65)	0.19
With supplements	0.40	0.50	0.48	0.58	(0.50, 0.65)	0.50						
Copper (mg)	0.36	0.51	0.50	0.58	(0.50, 0.64)	0.60	0.29	0.40	0.38	0.52	(0.41, 0.61)	0.25
With supplements	0.51	0.57	0.56	0.63	(0.56, 0.68)	0.64						
Zinc (mg)	0.36	0.41	0.42	0.52	(0.43, 0.59)	0.49	0.31	0.28	0.29	0.53	(0.36, 0.66)	0.11
With supplements	0.61	0.60	0.61	0.68	(0.62, 0.73)	0.66						
Phosphorus (mg)	0.41	0.58	0.60	0.67	(0.61, 0.71)	0.68	0.36	0.51	0.51	0.68	(0.57, 0.77)	0.24
With supplements	0.41	0.59	0.60	0.67	(0.61, 0.72)	0.68						
Choline (mg)	0.44	0.53	0.55	0.62	(0.56, 0.68)	0.63	0.35	0.40	0.40	0.61	(0.49, 0.71)	0.19
With supplements	0.43	0.52	0.54	0.61	(0.54, 0.67)	0.62						
Potassium (mg)	0.45	0.57	0.60	0.65	(0.60, 0.70)	0.73	0.41	0.52	0.53	0.64	(0.56, 0.71)	0.36
With supplements	0.45	0.56	0.60	0.65	(0.60, 0.70)	0.73						
Sodium (mg)	0.28	0.46	0.44	0.53	(0.44, 0.60)	0.50	0.26	0.31	0.29	0.43	(0.31, 0.53)	0.20
Caffeine (mg)	0.78	0.76	0.76	0.80	(0.77, 0.83)	0.86	0.72	0.72	0.70	0.77	(0.71, 0.82)	0.50

Note: data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012.
*ICC: Intraclass correlation coefficient for each comparison method is presented for energy-adjusted intake using the residual method in the log scale.

Table 1.5. Summary of Spearman correlation coefficient in overall and subgroups of participants

Group	Mean (Min, Median, Max)			
	Unadjusted	Energy-adjusted	De-attenuated	ICC**
Overall (N=632)*				
SFFQ2 vs 7DDR	0.44 (0.28, 0.43, 0.86)	0.53 (0.36, 0.52, 0.77)	0.63 (0.46, 0.65, 0.84)	0.56 (0.23, 0.58, 0.86)
SFFQ2 vs ASA24	0.38 (0.23, 0.36, 0.75)	0.43 (0.15, 0.42, 0.70)	0.62 (0.29, 0.64, 0.77)	0.22 (0.08, 0.20, 0.50)
SFFQ1 vs 7DDR	0.44 (0.31, 0.43, 0.84)	0.51 (0.37, 0.49, 0.77)	0.60 (0.48, 0.59, 0.84)	0.56 (0.23, 0.58, 0.86)
SFFQ1 vs ASA24	0.38 (0.18, 0.35, 0.75)	0.41 (0.11, 0.39, 0.74)	0.57 (0.18, 0.59, 0.79)	0.22 (0.08, 0.20, 0.50)
WEBFFQ vs 7DDR	0.42 (0.27, 0.39, 0.85)	0.52 (0.36, 0.50, 0.76)	0.61 (0.46, 0.61, 0.82)	0.56 (0.23, 0.58, 0.86)
WEBFFQ vs ASA24	0.37 (0.20, 0.34, 0.75)	0.42 (0.14, 0.40, 0.71)	0.60 (0.26, 0.60, 0.78)	0.22 (0.08, 0.20, 0.50)
22 nutrients without supplements				
SFFQ2 vs 7DDR	0.45 (0.35, 0.45, 0.60)	0.54 (0.42, 0.53, 0.66)	0.63 (0.52, 0.65, 0.72)	0.57 (0.37, 0.57, 0.73)
SFFQ1 vs 7DDR	0.44 (0.34, 0.44, 0.57)	0.50 (0.41, 0.49, 0.61)	0.59 (0.51, 0.59, 0.67)	0.57 (0.37, 0.57, 0.73)
WEBFFQ vs 7DDR	0.42 (0.32, 0.41, 0.56)	0.52 (0.43, 0.51, 0.61)	0.61 (0.52, 0.61, 0.67)	0.57 (0.37, 0.57, 0.73)
22 nutrients with supplements				
SFFQ2 vs 7DDR	0.60 (0.40, 0.64, 0.72)	0.64 (0.48, 0.67, 0.73)	0.71 (0.58, 0.74, 0.79)	0.68 (0.46, 0.72, 0.80)
SFFQ1 vs 7DDR	0.58 (0.43, 0.60, 0.72)	0.61 (0.48, 0.62, 0.72)	0.67 (0.58, 0.69, 0.76)	0.68 (0.46, 0.72, 0.80)
WEBFFQ vs 7DDR	0.57 (0.38, 0.62, 0.70)	0.61 (0.47, 0.65, 0.71)	0.68 (0.57, 0.71, 0.77)	0.68 (0.46, 0.72, 0.80)
Subgroup with complete four ASA24s (N=226)				
SFFQ2 vs 7DDR	0.44 (0.23, 0.44, 0.85)	0.54 (0.32, 0.55, 0.77)	0.64 (0.46, 0.64, 0.83)	0.57 (0.28, 0.55, 0.88)
SFFQ2 vs ASA24	0.41 (0.24, 0.40, 0.75)	0.45 (0.24, 0.45, 0.71)	0.63 (0.42, 0.65, 0.80)	0.22 (0.08, 0.20, 0.51)
Age 45-60 years old (N=309)				
SFFQ2 vs 7DDR	0.46 (0.24, 0.45, 0.85)	0.54 (0.28, 0.53, 0.78)	0.63 (0.41, 0.65, 0.83)	0.56 (0.15, 0.57, 0.87)
SFFQ2 vs ASA24	0.39 (0.20, 0.37, 0.75)	0.43 (0.19, 0.41, 0.75)	0.59 (0.31, 0.61, 0.80)	0.22 (0.08, 0.20, 0.49)
Age 61-80 years old (N=323)				
SFFQ2 vs 7DDR	0.42 (0.27, 0.39, 0.86)	0.53 (0.34, 0.51, 0.77)	0.62 (0.43, 0.61, 0.83)	0.57 (0.30, 0.56, 0.85)
SFFQ2 vs ASA24	0.38 (0.18, 0.36, 0.78)	0.46 (0.13, 0.44, 0.70)	0.61 (0.21, 0.60, 0.76)	0.23 (0.07, 0.22, 0.53)
BMI<25 (N=298)				
SFFQ2 vs 7DDR	0.45 (0.23, 0.43, 0.85)	0.56 (0.35, 0.56, 0.77)	0.65 (0.44, 0.64, 0.84)	0.59 (0.20, 0.61, 0.86)
SFFQ2 vs ASA24	0.43 (0.24, 0.43, 0.75)	0.46 (0.12, 0.46, 0.73)	0.62 (0.25, 0.62, 0.80)	0.25 (0.08, 0.24, 0.53)
BMI>=25 (N=334)				
SFFQ2 vs 7DDR	0.44 (0.29, 0.44, 0.86)	0.50 (0.34, 0.49, 0.76)	0.61 (0.48, 0.61, 0.82)	0.54 (0.25, 0.53, 0.86)
SFFQ2 vs ASA24	0.35 (0.15, 0.34, 0.74)	0.42 (0.19, 0.40, 0.71)	0.61 (0.40, 0.62, 0.76)	0.20 (0.04, 0.18, 0.48)
BMI>=30 (N=127)				
SFFQ2 vs 7DDR***	0.39 (0.16, 0.40, 0.85)	0.47 (0.27, 0.46, 0.75)	0.59 (0.34, 0.58, 0.78)	0.49 (0.16, 0.48, 0.86)
SFFQ2 vs ASA24	0.37 (0.09, 0.35, 0.75)	0.42 (0.17, 0.41, 0.72)	0.57 (0.34, 0.57, 0.84)	0.18 (0.04, 0.17, 0.40)

* The table shows the summary statistics for 44 common nutrients without supplements among SFFQ, 7DDR and ASA24 unless otherwise indicated;
**ICC: Intraclass correlation coefficient for each comparison method is presented for energy-adjusted intake using the residual method in the log scale.
**The ICC for Retinol activity equivalent in 7DDR was -0.02 among subgroup of participants with BMI>=30kg/m2, thus de-attenuated Spearman correlation coefficient was not available.

Paper II

Validation of Long-term Intakes of Energy, Protein, Sodium and Potassium Among Women Assessed by Questionnaire and 24-hour Recalls Using Biomarkers and Diet Records as Standards

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Abstract

In the Women's Lifestyle Validation Study, we collected recovery biomarkers for total energy, protein, sodium and potassium with replicates at 3-12 months interval and utilized these biomarkers to evaluate the performance of our semi-quantitative food frequency questionnaire (SFFQ) and a web-based 24-hour recall (ASA24) among 624 women. Three SFFQ's (paper SFFQ1 administered at the beginning; paper SFFQ2 and WebFFQ administered approximately one year later at the end of the study's diet data collection), four ASA24s, two 7-day dietary records (7DDR), four 24-hour urine samples for intakes of protein, sodium and potassium, and one assessment of energy intake by doubly labeled water (DLW) (repeated in a sample of 76 participants) were obtained over approximately one year. Compared to the DLW measurements, energy intake was underreported by all self-reported methods. The correlations of total energy intake assessed by SFFQs and averaged ASA24s with DLW were relatively low (r=0.11-0.20). The SFFQ reported comparable protein and potassium intakes to biomarker, while ASA24 and 7DDR underreported those intakes. Sodium intake was underreported by all self-reported methods, especially by SFFQ. For energy-adjusted protein, sodium and potassium, large within-person variation was observed for the ASA24 and biomarkers. After correcting for random within-person variations in the reference methods, each SFFQ, especially the average of SFFQ1 & 2, and averaged ASA24s had modest-to-strong correlations with the biomarkers (for SFFQ2 vs. biomarker: r=0.46 for protein, 0.48 for sodium and 0.49 for potassium); for averaged ASA24s (mean of 2.9 days) vs. biomarker: these correlations were r=0.54 , 0.36, and 0.52). These correlations were slightly higher when the 7DDR's were used as the reference method, and higher correlations were observed between 7DDR and biomarkers after correcting for random within-person variations in the latter (r=0.46 for total energy, 0.67 for protein, 0.54 for sodium

and 0.64 for potassium). Using the method of triads, the estimated correlations of both the FFQs and ASA24s with true intake were higher than these correlations, irrespective of whether biomarkers or 7DDRs were served as the reference methods. These data indicate that the SFFQ and averaged ASA24's are reasonably valid measurements for energy-adjusted protein, sodium and potassium compared to multiple recovery biomarkers or dietary records among women.

I. Introduction

In large cohort studies of diet and health outcomes, food frequency questionnaires have been the primary method of dietary assessment because they can be self-administered, efficiently processed, and provide data on individual intakes of both foods and nutrients over an extended period of time. [1] The validity of these questionnaires (FFQ) for measuring long-term diet has been evaluated extensively in many studies, indicating a strong concordance with dietary records or 24-hr dietary recalls.[1,2] However, correlated errors between FFQs, diet records and recalls may tend to overestimate the validity of FFQ's. Conversely, both comparison methods have their own sources of error, which would tend to underestimate validity of the FFQ. To overcome these limitations, biomarkers has been utilized as the reference method to evaluate the performance of other dietary methods.

Recovery biomarkers, based on the balance between intake and output to estimate absolute intakes over a certain time period, are in principle unlikely to have errors that are correlated with errors in reported intake.[3] Because few recovery biomarkers have been identified, existing validation studies have been mainly based on doubly-labeled water (DLW) (as a measure of energy intake) and 24-hr urinary nitrogen (UN) (as a measure of protein intake), sodium, and potassium. Most recently, a study pooled the results from five validation studies, in which the study design and the reported correlations for energy, protein, sodium and potassium between FFQ or 24-hr recalls and biomarkers varied across included studies. The pooled results found that, in general, FFQs have stronger correlations with biomarkers for protein density (percent of energy from protein) than for absolute protein intake, multiple 24-hour recalls perform better than a single recall in measuring energy and protein density; FFQs and 24-recalls measured potassium intake better than sodium, and modest correlations were observed between energy-

adjusted sodium and potassium intakes assessed by FFQ's or multiple 24-hr recalls and the biomarkers.[4-10] However, limitations in the analyzed studies include that replicate biomarkers were only collected in small subgroups (e.g., less than 25% participants) with a relative short time interval (mostly from several days to several months) from the first measurement, [11] which were unlikely to adequately account for within-person variation over a year or more.[12-14] Furthermore, the biomarkers were often collected close in time with 24-hour recalls, which may tend to overestimate the validity of 24-hour recalls due to correlations between within-person variations in the two methods if they both reflect short term intake.[13]

To address these issues, we aimed to evaluate the validity of multiple dietary assessment methods over a one-year period by comparisons with recovery biomarkers using repeated samples obtained over this one-year interval. The performance of our semi-quantitative food frequency questionnaire (SFFQ), which has been widely used in many large cohort studies, and the web-based self-administered 24-hour recall (ASA24), a relatively new, easily accessible, free method developed by the National Cancer Institute [15], was evaluated. Triangulation analyses (methods of triads) including SFFQ (or ASA24), biomarkers, and two 7-day dietary records (7DDR) were also conducted to estimate the validity of each method with the underlying true intakes.

II. Materials and Methods

A. Study Population

The current study was based on the Women's Lifestyle Validation Study (WLVS), one of the three studies that comprise The Multi-Cohort Eating and Activity Study for Understanding Reporting Error (MEASURE), which was designed to investigate the measurement error

structure associated with self-reported dietary and physical activity assessments, and how objective measures might be used to minimize error.[16] In 2010, we randomly selected a subset of Nurses' Health Study [17] and Nurses' Health Study II[18] participants aged 45-80 years, from all geographical regions of the US, who had completed the 2006/2007 cohort FFQ, had previously provided blood samples, had access to broadband internet, and were not planning to change their diet nor their physical activity levels. Women with medical history of coronary heart disease, stroke, cancer, or major neurological disease were excluded. The sample selection was stratified by age, and African-Americans were oversampled. A total of 5,509 female nurses were invited via email, by regular US mail method or both. Of these, 796 consented to participate, and received information on the timeline and activities. Study instructions were provided on the study website. Participants received support from study staff throughout their participation via the internet, telephone, and mail. Study participants were offered $600 for completion of the study due to the many complex activities. This study was approved by the Human Subjects Committees of the Harvard T.H. Chan School of Public Health and Brigham and Women's Hospital.

This study used doubly-labeled water (DLW) (as a measure of energy intake); 24-hr excretion of urinary nitrogen (as a measure of protein intake), sodium, and potassium; and 7-day dietary records (7DDR) to evaluate the performance of both our SFFQ and ASA24s (see Figure 2). To represent the one-year period typically used as the time frame for dietary questionnaires, we spread the dietary and biomarker measurements over a period of approximately one-year and altered the order of measurements by dividing the participants into four groups. We collected one paper SFFQ (SFFQ1) at baseline, a second paper SFFQ (SFFQ2) and the WebFFQ (two weeks apart from the second paper SFFQ and in random order) at the end of data collection and

approximately one-year after SFFQ1, one ASA24 in each season and two 7DDRs 6 months apart to capture seasonal variability, DLW once for all participants randomly over one year with a subset of 100 women having a repeated measure obtained at an interval of 6, 9 or 12 months (33 per subgroup), and four 24-hour urines (once each season) to capture within-person variation over one-year period . By design, three ASA24s were collected on weekdays and one on a weekend to capture differences in eating habits; within the same phase, the 7DDRs, ASA24s, DLW and 24-hr urine measurements were collected 1-4 weeks apart from each other and in random order to avoid artificially high correlations.[13,19]

Among 796 participants who completed the first activity (SFFQ1), 795 completed SFFQ1, 774 completed at least one 7DDR, 692 completed at least one ASA24, 755 completed at least one DLW, 777 completed at least one 24-hr urine collection, 759 completed SFFQ2, and 747 completed WebFFQ. Among 755 participants with first DLW measurement (N=192, 189, 187 and 187 across four phases), 90 (12%) of them have a replicate DLW at least 6 months from the first measurement. Four repeated 24-hr urines were requested from participants (we obtained 770, 763, 759 and 755 across the four phases). Of the 692 participants completing ASA24s, 93 (13%) competed one ASA24, 136 (20%) completed two, 219 (32%) competed three, and 244 (35%) completed four ASA24s. The distribution of ASA24 over the four phases was 512, 496, 497 and 493, respectively. Lower completion rates for the ASA24 were likely related, at least in part, to difficulties with the internet interface, which has been addressed in further versions of ASA24. In the quality control of SFFQ nutrient measurements, we excluded those participants with total daily energy intake <600 kcal or >3500 kcal or with more than 70 blanks. Overall, 771 SFFQ1 (97%), 742 SFFQ2 (98%), and 721 WebFFQ (97%) were included. These same restrictions for kilocalories and blank food items have been applied in analyses of diet and disease in the NHS

and NHSII studies. We also excluded one urine measurement with a reported urine volume of 17714 ml. Our primary analysis included 624 participants with complete data for SFFQ1, SFFQ2, WebFFQ, at least one 7DDR, one ASA24, one DLW and one 24-hr urinary measurement for nitrogen, potassium and sodium. Information on year of birth, height, weight, ethnicity and smoking status was collected at enrollment. Weight information was also collected every four months during follow up.

B. The semi-quantitative food frequency questionnaire (SFFQ)

The 152-food-item semi-quantitative SFFQ was a slightly expanded version of the questionnaire previously validated in the NHS and Health Professionals Follow-up Study (www.channing.harvard.edu/nhs/questionnaires/pdfs/NHSII/2007.pdf).[1,20,21] Small modifications since 1986 were made to account for changes in the food supply (e.g., more low fat versions) based on pilot surveys conducted every 4 years. Respondents were asked how often, on average, they had consumed the specified amount of each type of food or beverage during the past year; nine possible frequency categories ranged from never/almost never to 6 or more times per day. Participants also reported any additional 3 foods not included on the questionnaire that they consumed more than once per week. Nutrient intakes were calculated from the questionnaire by multiplying a weight assigned to the frequency of use (where once per day is equal to one) by the nutrient composition for the portion size specified for each food or vitamin supplement based on an extensive and regularly updated food composition database (https://regepi.bwh.harvard.edu/health/nutrition/repeatUser.html) maintained at Harvard T.H Chan School of Public Health. The web-based version of this questionnaire was similar to the standard paper questionnaire and was enhanced by the use of branched questions. Daily intakes

of energy (kcal/day), protein (g/day), sodium (mg/day) and potassium (mg/day, from food and supplements) were obtained from SFFQ data.

C. 7 Day Dietary Records (7DDR)

WLVS research dietitians developed the 7DDRs with detailed instructions and a DVD instructional video. Each participant received an Escali food scale and ruler, the instructional DVD, and instructions via telephone by our research dietitians explaining how to keep detailed 7DDRs. A computerized reminder system was used to track participant study activities, sent reminder emails to participants on day 2, day 3 or 4 and day 6 of the diet recording week and to encourage participants to complete their diet record, to ask study research dietitians for clarifications on how to complete their diet records, and to review the DVD. Participants reported the gram weights for food intake and provided recipes of all home prepared foods including the number of recipe servings, and the portion of the recipes they each consumed. Participants also reported gram weights of foods before and after consumption, as relevant, so actual intake could be computed. Additionally, participants collected labels of store brand products and returned them with their records for analysis. A research dietitian contacted each participant during the recording week to discuss any questions and review procedures. Issues found in their first 7DDR provided after coding by the Nutrition Coordinating Center (NCC) at the University of Minnesota[22] were specifically addressed during their second 7DDR instructional session with the research dietitian. The NCC used Nutrition Data System for Research (NDSR) 2011 to analyze the 7DDRs.[23,24] Daily intakes of energy (kcal/day), protein (g/day), sodium (mg/day) and potassium (mg/day from food and supplements) were obtained from 7DDR data.

D. Web-based, self-administered 24-hour dietary recall (ASA24)

Participants logged into the Beta version of ASA24 website to complete their 24hr recall entries using accounts created by the NCI. Participants were reminded on assigned days to complete the web-based recall by a computerized reminder system to ensure their readiness and availability to do so that day. The USDA's Food and Nutrient Database for Dietary Studies (FNDDS 4.1) was used to derive the nutrient data.[25] Daily energy (kcal/day), protein (g/day), sodium (mg/day) and potassium (mg/day, from food only) intakes were obtained from ASA24 data. The potassium intake from ASA24 didn't account for supplements; however, the amounts used were quite small and could be considered as ignorable compared to intake from the food.

E. Doubly-labeled water (DLW)

The DLW method is considered the gold standard for measuring total energy expenditure in free-living persons. Energy intake is estimated as the amount of carbon dioxide produced over a 10-14 day period, assuming that the individuals are in energy balance.[26] WLVS participants were provided with a specimen collection kit (including bottled DLW) for DLW Urine collection. Participants first provided two spot urine samples (10 ml each) before oral intake of water labeled with nonradioactive $^{2}H_{2}{}^{18}O$, and then provided two spot urine samples (10 ml each) at 4.5 hours and 6 hours later. After 10 to 14 days, participants provided two more post dose urine samples following the same procedures; the two urine samples were separated by at least 30 minutes or more. Participant weight, date, time and volume of each urine collection, and date and time of DLW dose were collected. Urine specimens were assessed at Dr. Jennifer Rood's Mass Spectrometry Laboratory at Pennington Biomedical Research Center via mass spectroscopic analysis for deuterium (^{2}H) and heavy oxygen (^{18}O).[27-29] The relative decrease in deuterium and oxygen-18 concentrations was used to calculate total daily energy expenditure (TDEE) as

described by Schoeller.[28,29] Due to the high cost of this measurement, we collected the first DLW assessments randomly across four phases among all participants, and collected replicate measures for a subset of 100 women at 6, 9 or 12 months from the first DLW measurement to assess within-person variability over one year. TDEE (kcal/day) were obtained from DLW data.

F. 24-hour Urine

Biochemical indicators of protein (g/day), sodium (mg/day) and potassium (mg/day) intakes were obtained from 24-hr urinary data. WLVS participants were provided with a specimen collection kit to self-collect 24-hour urine samples. The urine collection started after the first morning void and continued until the first morning void of the following morning. Participants were asked to not take medications containing, acetaminophen, sulphonamides, or vitamin supplements during the 24-hour urine collection period. Starting and ending time/date of collection, the time and approximate amount of the missed sample and total volume collected were recorded. Participants' sample collection kits was picked-up by FedEx and shipped with the cold pack by overnight mail to Fisher BioServices, Inc.50 ml of urine samples were stored for immediate and further analyses. Sample collection, return and processing were carefully monitored to reduce error by Fisher BioServices. Urinary nitrogen, sodium and potassium concentrations were measured at the Tufts University Nutrition Evaluation Laboratory. Urinary potassium and sodium were measured using ion selective electrode methods with the Olympus AU400 analyzer (Olympus, Tokyo, Japan). Urinary nitrogen was measured by a Dumas method of combustion–detection by a Thermal Conductivity Cell assay (FP2000 Nitrogen Analyzer, LECO Corporation, LECO Corporation, St. Joseph, MI). Urinary values for nitrogen, sodium and potassium were calculated multiplying the urinary concentrations by reported total urine volumes. Urinary nitrogen in grams was divided by 0.81 to convert the measurement to dietary

nitrogen and was then multiplied by 6.25 to obtain the dietary protein intake (g/day).[30] Urinary values were divided by 0.86 for sodium, and 0.80 for potassium to convert them to the corresponding dietary values (mg/day).[10,31]

G. Statistical Analysis

Average daily intakes of energy, protein, sodium and potassium estimated from the SFFQs, ASA24s were compared with the corresponding recovery biomarkers and 7DDRs. Means and standard deviations for total daily nutrient intakes from SFFQ1, SFFQ2, WebFFQ, ASA24s (day1-4, and days average), 7DDRs (week 1, week 2 and week average), DLW (Phase 1-4, and days average), and 24-hr urinary measurements (phase 1-4, and averaged value) were calculated. Log transformation was performed for nutrients to increase normality because their distributions were skewed toward higher values. Energy-adjusted intakes are of greatest importance because individuals primarily alter their intakes of specific nutrients by changing the composition of their diet, keeping total energy constant.[1,32] Therefore, we calculated energy-adjusted protein (percent of total energy from protein), sodium and potassium (the ratio of nutrient intake (mg) to energy intake (1000 kcal)) [33].

As the first DLW measurement was randomly assessment across four phases, we utilized two energy adjustment strategies. The primary method used concurrent DLW measurements to calculate nutrient density for nutrient biomarkers, which led to a reduced sample size at each phase and the use of one 24-hr urine for most women; only the 76 women with a second DLW measurement had two energy-adjusted measurements that provide data on within-person variation. The second strategy assumed that the first DLW measure (obtained for all participants) represents the level across the one-year period, therefore each 24-hr urinary measurement was adjusted for the first DLW measure, no matter when it was collected. To further reduce the

influence of the extreme nutrient distributions, the subsequent analyses were based on the ranks of the log-transformed nutrient and nutrient density value.

To assess the reproducibility of repeated SFFQ, ASA24, 7DDR and recovery biomarkers, rank intraclass correlation coefficients (ICC) were calculated for each nutrient at both unadjusted and adjusted for energy intake. To assess nutrient validity, the averaged recovery biomarker, or 7DDR intakes, repeated at an interval of at least several months over a one-year period, were assumed as unbiased estimates of the true underlying nutrient intakes. Spearman correlation coefficients (r_0) between nutrient intakes reported on each surrogate method (SFFQ2, SFFQ1, averaged SFFQ, WebFFQ, averaged ASA24), and the corresponding intakes reported on the biomarkers and 7DDRs were calculated. Random within-person variation in the biomarkers or 7DDRs can attenuate correlations between these measures and the surrogate methods. Therefore, we calculated de-attenuated Spearman correlation coefficients (r_c) to reduce the effect of random within-person variation in the reference methods. The standard method first calculates the within- and between- person components of variation in nutrient intakes of the reference methods,[34,35] and corrects correlation coefficients for the within-person variation using $r_c = r_0\sqrt{(1 + \gamma / k)}$, where r_0 represent the observed correlation coefficient, r_c represents the corrected correlation coefficient, and γ equals the ratio of estimated within-person variation and between-person variation, k is the number of repeated observations of the reference methods.[34,35] As the number of biomarkers was unbalanced among individuals, we used an extension of the methods [36] by Rosner & Glynn [37] and Perisic & Rosner [38] to correct for random within-person measurement error in the presence of unbalanced data and applied the method by Rosner and Willett [35] to calculate corresponding 95% CI for de-attenuated correlation coefficients.

To further correct for bias due to potential correlated errors in the repeated measurements from the reference method, and simultaneously compare the relative performance of both surrogate methods and the reference methods, a triangular comparison (the method of triads) was used to calculate the validity coefficient (VC), an estimate of the correlation of each method with true intake, making the assumption that all dietary measurements were linearly related to true intake and that their measurement errors were mutually independent. [39] The following formula was derived based on the factor analysis model, in which the validity coefficient (ρ) was calculated from the observed correlation (r) estimated above.

The "method of triads" formulae are [39]:

$$\rho_{\mathrm{QT}} = \sqrt{r_{QR} \times r_{QB} / r_{RB}} \quad \rho_{\mathrm{RT}} = \sqrt{r_{QR} \times r_{RB} / r_{QB}} \quad \rho_{\mathrm{BT}} = \sqrt{r_{BQ} \times r_{BR} / r_{QR}},$$

where Q indicates SFFQ or ASA24 intakes, R indicates diet record intake, and B indicates recovery biomarkers, T indicates true underlying intakes, ρ is the validity coefficient, and r is the observed correlation coefficient in the current analyses. In our case, we used the Spearman correlation coefficient (r). As noted elsewhere, correlations can be estimated as greater than 1.0 due to random errors; as suggested, these have been set to 1.0.[39]

To understand how temporal relations would affect the relative validity of different methods, we examined correlations between any one, or any two days of ASA24-assesed intakes (e.g. ASA24, Day 1, Day 2, or averaged over Day 1 & 2) and any one, or two measurements of the biomarker for each nutrient (e.g. UN, Day 3, Day 4, and averaged over Day 3 & 4).

III. Results

Descriptive statistics

Study participants had a mean age of 61 years and a mean BMI of 26.6 kg/m^2 at WLVS enrollment, 90.2% were white and 1.9% were current smokers. 76 participants had two measurements of DLW, and this subgroup had characteristics similar to the overall population (Table 2.1). The distribution of mean daily intakes of energy, protein, sodium and potassium (absolute and energy-adjusted intakes), and the reproducibility of each method are described in Table 2.2. Compared to recovery biomarkers, intakes of total energy were under-reported by all self-reported methods (SFFQ, ASA24, and 7DDR). The paper SFFQ reported protein intake similar to the biochemical measures, while WebFFQ, 7DDR and ASA24 tended to underestimate protein intake. Furthermore, protein density tended to be slightly over-reported by these methods as a consequence of greater underreporting of total energy than that of protein intake. Compared with 24-hr urinary sodium, all self-reported methods, especially the SFFQ, under-reported the intake. For potassium, both ASA24 and 7DDRs underreported intake while intakes measured by SFFQ were slightly higher compared to potassium biomarkers. Nutrients estimated by ASA24 and biomarkers had relatively wider distributions, which were mainly due to the large day-to-day variation of dietary intakes.

As shown in Table 2.2, total energy intake was highly reproducible by two SFFQs (ICC=0.70) spaced one-year apart, and by two 7DDRs (ICC=0.63) with a six months interval. Total energy intake estimated by ASA24 repeated every three months over a year was relatively low (ICC=0.28). For DLW, all participants randomly provided one assessment, with collection spread evenly across the year. Among the 164 women who provided a DLW in the first three months, 76 women provided a second DLW randomly at approximately 6, 9, or 12 months from

the first assessment. For the 76 participants with two DLWs, the ICC was 0.72 if repeated after 6 months, 0.78 if repeated after 9 months, and 0.64 if repeated after 12 months; for all 76 women the ICC for total energy was 0.71 with an average of interval of 8.8 months between replicated measurements. For absolute intakes of protein, sodium, and potassium, we observed a high degree of reproducibility by SFFQs (ICCs=0.65-0.73), and by 7DDRs (ICCs=0.64-0.75). The reproducibility of corresponding biomarkers (0.37-0.56) was lower and much lower in ASA24 (0.23-0.37) measured every three months over one year. Almost all ICCs were reduced after adjustment for total energy intake using the nutrient density method. The lower ICCs for most nutrients measured by ASA24 and biomarkers indicated large within-person variation in nutrient intakes over the one-year interval.

Spearman correlation coefficients between intakes assessed by SFFQ or ASA24 and by biomarkers and7DDR

When using biomarkers as the reference methods, the Spearman correlation coefficients (r) between the absolute nutrient intakes from SFFQ2 (which assesses intake over the same period as the biomarker collection),), and those from the four biochemical measures were 0.09 for total energy, 0.25 for protein, 0.16 for sodium intake, and 0.32 for potassium (Table 2.3). The correlation was improved when using nutrient density instead of absolute intakes for protein (r=0.29), sodium (r=0.25) and potassium (r=0.38). As the random within-person variation over a year was relatively larger for protein, sodium and potassium biomarkers at nutrient density scale, the de-attenuated correlations increased substantially for the two nutrients (r_c=0.46 for protein density, 0.48 for sodium density, and 0.49 for potassium density). The correlations for energy also increased after correcting for random within-person variation in the biomarker measurements (r = 0.11). Similar patterns of correlations were observed in SFFQ1 and WebFFQ,

although as expected the correlations were slightly lower in SFFQ1 (except protein) because this referred to the year before collection of the biomarkers. When using the averaged nutrient intakes of SFFQ1 and SFFQ2, the correlations with reference methods were slightly increased. The average of ASA24's (mean = 2.9 days), as a surrogate method, performed slightly better than SFFQ2 in estimating total energy (r_c=0.20), protein density (r_c=0.54) and potassium density (r_c=0.52), but was lower for sodium density (r_c=0.36). When 7DDRs were used as the reference method, the correlations of SFFQ, and average of ASA24's with the mean of two 7DDRs were higher compared to the corresponding correlations when biomarkers were instead used as the reference method. Higher correlations were observed between 7DDR and biomarkers after correcting for random within-person variations in the latter (r=0.46 for total energy, 0.67 for protein density, 0.54 for sodium density and 0.64 for potassium density) (Table 2.3 & Figure 3a).

We also compared each single 7DDR, single ASA24, the average intake of different combinations of two ASA24s, with each single and two measurements of biomarkers. Due to random fluctuations in a single day measurement of total energy intake, we observed highly variable correlations between a single ASA24 and a single DLW measurement, even when they were measured within the same phase (ranged from -0.01 to 0.37). Also DLW measured at phase 2 (Day 2) had consistently higher correlations with any single measure of ASA24 (0.24-0.36) (Supplementary Table 2.1). For absolute protein, sodium and potassium, the correlations of the biomarker measured closer in time to the ASA24 were slightly higher, although by design these measurements were at least several weeks apart. This pattern disappeared when the nutrient was adjusted for total energy intakes (Supplementary Tables 2.2a-f). Of note, the energy adjustment method used concurrent DLW measurements, which led to a reduced sample size at each phase and the use of one 24-hr urine measurement for most participants.

Instead of using concurrent DLW to derive nutrient density values, applying the first DLW measurement to each urinary biomarker tended to underestimate the random within-person variations in protein density and sodium density and slightly overestimated the random within-person variations in potassium density. Taking protein and potassium as an example, the total energy intake contributed relatively more to the variation in the ratio compared to potassium. Thus, keeping the total energy intake constant (denominator) tended to artificially create a higher ICC for protein than for potassium. Compared to the concurrent adjustment method, this method gave relatively narrow confidence intervals for the de-attenuated correlations due to its larger sample size at each phase (four 24-hr urine measurements were used for all participants) (Supplementary Table 2.3-2.5). The pattern that the relatively higher correlation between a single ASA24 day and a single biomarker measured within the same phase somewhat remained if using the first DLW measurement to adjust for total energy intakes for the four repeated biomarkers (Supplementary Tables 2.6a-c).

Validity coefficient

The validity coefficients for each dietary assessment method are shown in Table 2.4 and Figure 3b.After accounting for potential positive correlated errors between repeated assessments of the reference methods, the estimated validity coefficients for SFFQs and ASA24s with true intake were higher than the observed correlations, irrespective of whether biomarkers or 7DDRs were served as the reference methods. Relative to the underlying true intakes, the validity coefficients were highest for 7DDR in estimating the total energy, potassium, sodium and protein intakes (de-attenuated ρ>=0.79). For total energy intake, SFFQ, DLW, and averaged ASA24 had relatively lower validity coefficients (de-attenuated ρ ranged from 0.26 to 0.47). The SFFQ, averaged

ASA24, and biomarkers had similar VCs for energy-adjusted protein, sodium and potassium (de-attenuated ρ>=0.61).

IV. Discussion

Utilizing recovery biomarkers for total energy, protein, sodium and potassium, we evaluated the performance of multiple dietary assessment methods, including our semi-quantitative FFQs (both paper and web version) , the web-based ASA24s and two 7DDRs kept over a one-year period among women aged 45-80 years old in Women's Lifestyle Validation Study. Total energy was underreported by all self-reported methods and its correlations between SFFQ, ASA24 with DLW were relatively low. For energy-adjusted protein, sodium and potassium, large within-person variations were observed for short-term intake measures such as ASA24 and biomarkers. After correcting for random within-person variation in the reference methods, SFFQs, especially the average of SFFQ1 & 2, and ASA24s had modest-to-strong correlations with both biomarkers and 7DDRs measurements, and the correlations with the latter one were slightly higher. Further adjustment for systematic errors in the biomarkers using the method of triads gave increased estimates for correlations with true intake of energy-adjusted protein, sodium, and potassium that were similar for the SFFQ, averaged ASA24's , and biomarkers (all validity correlations >= 0.58).

Recovery biomarkers of dietary intake are considered as an objective 'gold standard' to validate other dietary methods; however, they still have many limitations due to high day-to-day variations and very high costs of data collection. Therefore, few large validations using recovery biomarkers have been published so far. In those studies, after accounting for within-person

variation in the biomarker, the correlations between energy intake and absolute protein intake measured by FFQ and by biomarkers were low (mean r=0.25 and 0.29 in women). [4] However, for energy-adjusted protein intake (protein density as a percent of energy), the correlations were considerably higher (mean r=0.43 in women). For protein density, multiple 24-hour recalls substantially increased correlations over a single 24-hour recall, and had similar correlations (mean r=0.40-0.47 in women) with that of a FFQ.[4] Our study has similar findings with the pooled analysis and some specific studies which had realistic intervals (5-6 months)[5,7] between repeated biomarker measurements. The correlation of SFFQ2 with biomarker was higher after adjusting protein for total energy intake (0.29 vs 0.25); Correcting for within-person variation in biomarker substantially increased this correlation (0.46 vs 0.29). And the correlation was higher when we using the mean of SFFQ1 and SFFQ as a measure of long-term dietary intake for protein density (r=0.52). Furthermore, the correlations for energy-adjusted protein intake were quite similar when using diet records as the reference method (SFFQ2: r=0.55 in this study; r=0.47 in our earlier validation study in women[21]). Multiple ASA24s used in our study (average = 2.9 days) had a slightly higher correlation with biomarkers (r=0.54). Although we tried to avoid artificially correlated errors between short-term methods in our design by having several weeks between urine collection and dietary assessments, the biomarker assessments were still more proximal to the period assessed by the ASA24 than the period assessment by the SFFQs; This could potentially cause overestimation of ASA24 correlations with long-term true intake and underestimation of SFFQ correlations (submitted manuscript).[19] Our study also observed moderate correlations for energy-adjusted sodium and potassium between SFFQ2, averaged ASA24s and biomarkers (r=0.48 and 0.49 for SFFQ2; r=0.36 and 0.52 for ASA24s). Our observed correlations were similar to the reported validity of multiple 24-hr recalls (mean r=0.38

and 0.53) but were higher than reported validities of SFFQ (mean r=0.32 and 0.47) in previous studies. [10]

As mentioned earlier, true day to day variation or random laboratory error in biomarker can attenuate its correlations with other dietary methods. Our study collected replicate biomarkers at least 3 months apart from each other over a one-year period and statistically corrected for the estimated random error. Our study observed an ICC of 0.71 for DLW measurements (an average of 8.8 months interval), which is consistent with previous reported reproducibility (0.72 for DLW repeated at an interval of 6 months and 0.43 when measurements repeated 16 months apart).[7,14] The ICCs were much lower for energy-adjusted protein (0.33), sodium (0.26) and potassium (0.56). Some previous studies did not adequately capture the long-term within person variation in the biomarkers, either due to small sample size of repeated measurements[9], or the relatively short time intervals between repeated measurement[6,8,9], or lack of adjustment for total energy intake [3,11].

Two reference methods, biomarkers and 7DDRs, were utilized in our study, and 7DDR had slightly higher correlations with both SFFQs and ASA24s. However, those reference methods are also subject to potential error that may not be entirely accounted for by correction for random within-person error, therefore typical validation studies using biomarkers or other dietary assessments as gold standards may underestimate true validity.[39,40] In our triangulation analyses, validity coefficients for SFFQs and ASA24s with true intake were higher than the observed correlations with biomarkers or 7DDRs. Although 7DDRs had the highest validity correlations (ranged from 0.79-1.00), both biomarkers and 7DDRs have some measurement errors, especially the DLW methods (validity coefficient=0.39, suggesting substantial measurement errors). Therefore, the observed correlations between SFFQ and ASA24s with biomarker, after

correcting for random with-in person variation, can be considered as the lower limits of the true validity coefficients. On the other hand, the validity coefficient from the 'method of triads' can be interpreted as the upper limits of the true validity coefficient, as there may be non-ignorable correlated positive errors between SFFQ or ASA24s and 7DDRs, which tended to overestimate the validity coefficient of SFFQs and ASA4s.[39,41]

Although biochemical indicators of diet have potential advantages as a reference method and provide valuable information, accurate biochemical markers of dietary intake are available for only a limited number of foods and food components [3] and are also subject to potential errors. Differences in the homeostatic, metabolic processes, and lifestyle factors between subjects can cause variability in biochemical indicators unrelated to dietary intake.[3] The effects of these factors may operate systematically within subjects, therefore errors between replicated biomarker samples tend to be positively correlated, producing what has also been called "person-specific bias". [3,39] Among the intake methods used in this study, the two 7DDR's had the strongest correlations with biomarkers of intake, presumably due to a total of 14 days of open-ended, weighed and recorded intake and the detailed coding of foods. Unfortunately the high costs and participant burden of this method preclude use in large studies, particularly if repeated assessments over time are needed. Participants had the most difficulty completing the requested four ASA24 assessments, resulting in lower completion rates (over 30% completed only one or two ASA24s); whether this can be improved by software enhancements needs to be evaluated.

To the best of our knowledge, this is the largest validation study using recovery biomarkers among women. By design, all short-term dietary methods were repeated at reasonable time intervals (at least three months apart) to capture the variation over one year, and those methods were not collected close in time (1-4 weeks apart from each other and in random order) to avoid

artificially high correlations. Future validation studies should consider these temporal relationships when being designed.[13] Our study also has some limitations. Our analysis is based only on total energy, protein, sodium, and potassium, and findings do not necessarily generalize to other nutrients. However, the correlations for protein intake tend to be lower than for other nutrients due to low between-person variation. Furthermore, errors in dietary assessments with the SFFQ, diet records, dietary recalls and biomarkers may lead to over- or under-estimate of the validities of different methods; our findings suggest that biomarkers, including DLW should not be considered to be without error, including systematic within-person error.

In conclusion, our data document that the semi-quantitative food frequency questionnaire and multiple automated self-administered 24-hour recalls provide reasonably valid measurements for energy-adjusted intakes of protein, sodium and potassium compared to multiple recovery biomarkers or dietary records among women.

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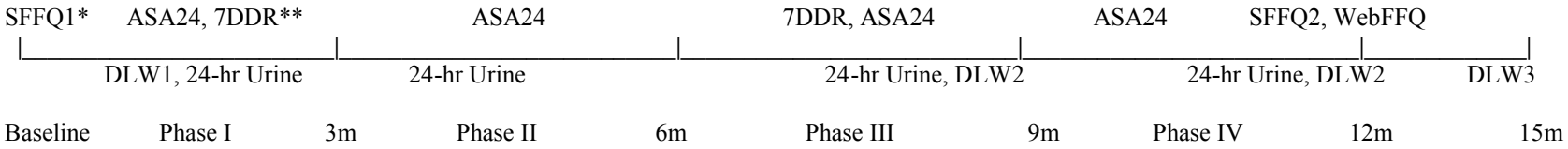
VI. Figures and Tables

Figure 2. Timeline of the dietary assessment activity in project 2.



* This figure showed the timeline for group 1 participants; group 3 went through the same data collection timeline as group 1, except all group 3 participants completed DLW in phase III, and had no DLW subgroup repeats; groups 2 and 4 went through similar data collection timeline as group 1, except Groups 2 and 4 participants completed DLW in phase II and IV, respectively, and had no DLW subgroup repeats. Group 2 and 4 also completed the 7DDR in phase II and IV.
**Within the same phase, 7DDRs and ASA24s, SFFQ2 and WebFFQ were in random order; 7DDR, ASA24, DLW and 24-hr Urine were collected 1-5 weeks apart from each other to avoid artificially high correlations.

Table 2.1. Subject characteristics among WLVS participants in project 2

Variables	All participants (N=624)	Participants with only one DLW measurements (N=548)	Participants with two DLW measurements (N=76)
Age at enrollment, mean(sd), year	61.4 (9.5)	61.5 (9.5)	60.8 (9.5)
Height, mean (sd), m	1.64 (0.07)	1.64 (0.07)	1.63 (0.07)
Weight at baseline, mean (sd), kg	71.6 (15.5)	71.9 (15.6)	69.7 (14.6)
Weight change at year 1, mean (sd), kg	-0.2 (2.7)	-0.2 (2.6)	-0.4 (3.3)
BMI at baseline, mean (sd), kg/m2	26.6 (5.4)	26.6 (5.5)	26.2 (5.0)
White, %	90.2	89.2	97.4
Current smokers, %	1.9	1.5	5.3
Total energy (kcal/day)			
7DDR	1737 (335)	1738 (336)	1732 (335)
DLW	2195 (369)	2202 (371)	2146 (349)
Protein (% total energy)			
7DDR	16.9 (2.9)	16.9 (2.9)	16.7 (2.5)
Biomarker	14.9 (4.0)	14.8 (4.1)	15.1 (3.8)
Sodium (mg/day)			
7DDR	2645 (641)	2650 (650)	2608 (573)
Biomarker	3388 (1004)	3411 (1023)	3221 (838)
Potassium (mg/day)			
7DDR	2661 (655)	2665 (665)	2637 (818)
Biomarker	3043 (866)	3047 (882)	2971 (1032)

Note: data provided by 624 U.S. women aged 45-80 years, 2010-2012
7DDR:7 Day Dietary Records
DLW: doubly-labeled water
Sd: standard deviation.

Table 2.2. Energy, protein, sodium and potassium intake distributions by different dietary methods

Method	Absolute level											Energy-adjusted level §						
	Total Energy (kcal)			N	Protein (g/day)		N	Sodium (mg/day)		Potassium (mg/day)		N	Protein (% energy)		N	Sodium (mg/1,000 kcal)		Potassium (mg/1,000 kcal)	
	N	Mean	(SD)		Mean	(SD)		Mean	(SD)	Mean	(SD)		Mean	(SD)		Mean	(SD)	Mean	(SD)
FFQ																			
SFFQ2	624	1853	(523)	624	80.4	(23.9)	624	2060	(661)	3286	(966)	624	17.5	(3.0)	624	1121	(215)	1794	(290)
SFFQ1	624	1927	(524)	624	83.9	(23.9)	624	2149	(682)	3427	(977)	624	17.6	(3.0)	624	1125	(230)	1798	(305)
SFFQ1&2	624	1890	(485)	624	82.2	(21.8)	624	2104	(628)	3357	(901)	624	17.6	(2.7)	624	1123	(205)	1796	(275)
WebFFQ	624	1765	(515)	624	76.9	(24.0)	624	1945	(637)	3180	(964)	624	17.6	(2.9)	624	1110	(215)	1824	(316)
$ICC_{ffq1\&2}$		0.70			0.65			0.73		0.71			0.63			0.68		0.69	
Diet record																			
Week 1	623	1766	(363)	623	73.2	(17.4)	623	2694	(719)	2693	(680)	623	16.7	(3.1)	623	1539	(328)	1543	(328)
Week 2	623	1710	(376)	623	71.7	(17.8)	623	2597	(719)	2631	(716)	623	17.0	(3.3)	623	1533	(312)	1560	(356)
Week 1&2	624	1737	(335)	624	72.4	(16.1)	624	2645	(641)	2661	(655)	624	16.9	(2.9)	624	1536	(277)	1551	(316)
ICC		0.63			0.64			0.58		0.75			0.60			0.50		0.70	
ASA24																			
Day 1	462	1791	(623)	462	75.8	(31.7)	462	2998	(1184)	2745	(1126)	462	17.2	(4.9)	462	1721	(522)	1567	(446)
Day 2	454	1832	(571)	454	77.5	(29.7)	454	3210	(1235)	2805	(1032)	454	17.3	(5.2)	454	1785	(522)	1582	(499)
Day 3	452	1814	(610)	452	76.9	(29.5)	452	3054	(1245)	2799	(1039)	452	17.5	(5.6)	452	1739	(693)	1604	(517)
Day 4	452	1800	(576)	452	75.2	(28.1)	452	2997	(1111)	2734	(935)	452	17.2	(5.1)	452	1719	(542)	1576	(492)
Days, all	624	1815	(471)	624	77.2	(23.7)	624	3079	(904)	2788	(856)	624	17.4	(3.8)	624	1742	(409)	1585	(376)
ICC		0.28			0.25			0.23		0.37			0.21			0.20		0.32	
Biomarker																			
Day 1	164	2111	(338)	607	80.2	(20.8)	607	3546	(1319)	3068	(1047)	156	14.7	(3.7)	157	1664	(620)	1462	(500)
Day 2	162	2232	(363)	600	80.8	(24.7)	603	3486	(1484)	3064	(1061)	157	14.8	(4.1)	157	1622	(637)	1419	(527)
Day 3	181*	2250	(397)	605	78.8	(22.1)	606	3220	(1319)	3021	(1052)	177	14.5	(3.5)	176	1497	(543)	1428	(481)
Day 4	193**	2166	(372)	609	79.2	(24.2)	605	3298	(1285)	3011	(1120)	189	15.4	(4.8)	187	1587	(539)	1435	(586)
Days, all	624	2195	(369)	624	79.8	(18.9)	624	3388	(1004)	3043	(866)	609	14.9	(4.0)	609	1590	(581)	1433	(515)
ICC		0.71			0.56			0.37		0.49			0.33			0.26		0.56	

*28 out of the 181 participants had the first measurement in phase 1; **48 out of the193 participants had the first measurement in phase 1;
§Nutrient density was calculated using the concurrent DLW assessment. Therefore, the sample size for energy-adjusted nutrients was reduced and was similar to that of the energy biomarkers.

Table 2.3. Spearman correlation coefficients for protein, sodium, potassium and energy estimated by different methods

Nutrient	Biomarker (Reference 1)				7DDR (Reference 2)			
	Absolute intake	Nutrient density	De-attenuated (ρ & 95%CI)	ICC *	Absolute intake	Nutrient density	De-attenuated (ρ & 95%CI)	ICC*
Protein								
SFFQ2	0.25	0.29	0.46 (0.27, 0.62)	0.33	0.32	0.48	0.55 (0.47, 0.61)	0.60
SFFQ1	0.29	0.31	0.50 (0.30, 0.65)	0.33	0.36	0.48	0.54 (0.47, 0.61)	0.60
SFFQ1&2	0.29	0.33	0.52 (0.31, 0.68)	0.33	0.37	0.53	0.60 (0.53, 0.66)	0.60
WebFFQ	0.26	0.28	0.46 (0.27, 0.61)	0.33	0.34	0.47	0.54 (0.46, 0.60)	0.60
Single ASA24	0.31	0.25	0.37 (0.19, 0.52)	0.33	0.36	0.38	0.44 (0.35, 0.52)	0.60
Averaged ASA24	0.39	0.33	0.54 (0.34, 0.70)	0.33	0.47	0.52	0.60 (0.53, 0.66)	0.60
Averaged 7DDR	0.65	0.42	0.67 (0.41, 0.83)	0.33				
Sodium								
SFFQ2	0.16	0.25	0.48 (0.25, 0.65)	0.26	0.28	0.45	0.56 (0.47, 0.63)	0.50
SFFQ1	0.18	0.23	0.41 (0.19, 0.59)	0.26	0.31	0.42	0.50 (0.42, 0.58)	0.50
SFFQ1&2	0.18	0.26	0.49 (0.25, 0.67)	0.26	0.32	0.47	0.58 (0.50, 0.65)	0.50
WebFFQ	0.15	0.26	0.46 (0.23, 0.65)	0.26	0.27	0.44	0.53 (0.44, 0.60)	0.50
Single ASA24	0.24	0.15	0.25 (0.08, 0.40)	0.26	0.32	0.30	0.37 (0.26, 0.45)	0.50
Averaged ASA24	0.29	0.19	0.36 (0.17, 0.53)	0.26	0.45	0.41	0.50 (0.41, 0.57)	0.50
Averaged 7DDR	0.48	0.32	0.54 (0.26, 0.74)	0.26				
Potassium								
SFFQ2	0.32	0.38	0.49 (0.38, 0.58)	0.56	0.45	0.57	0.63 (0.56, 0.68)	0.70
SFFQ1	0.33	0.34	0.43 (0.33, 0.52)	0.56	0.48	0.52	0.56 (0.49, 0.61)	0.70
SFFQ1&2	0.34	0.39	0.49 (0.38, 0.58)	0.56	0.50	0.59	0.63 (0.57, 0.68)	0.70
WebFFQ	0.32	0.32	0.42 (0.32, 0.51)	0.56	0.42	0.50	0.56 (0.49, 0.62)	0.70
Single ASA24	0.40	0.33	0.41 (0.29, 0.51)	0.56	0.49	0.49	0.53 (0.45, 0.60)	0.70
Averaged ASA24	0.48	0.43	0.52 (0.41, 0.61)	0.56	0.61	0.62	0.67 (0.61, 0.72)	0.70
Averaged 7DDR	0.65	0.50	0.64 (0.52, 0.72)	0.56				

Table 2.3 (continued): Spearman correlation coefficients for protein, sodium, potassium and energy estimated by different methods

Nutrient	Biomarker (Reference 1)				7DDR (Reference 2)			
	Absolute intake	Nutrient density	De-attenuated (ρ & 95%CI)	ICC *	Absolute intake	Nutrient density	De-attenuated (ρ & 95%CI)	ICC *
Energy								
SFFQ2	0.09		0.11 (0.02, 0.19)	0.71	0.27		0.30 (0.22, 0.37)	0.63
SFFQ1	0.09		0.10 (0.02, 0.19)	0.71	0.28		0.30 (0.22, 0.38)	0.63
SFFQ1&2	0.10		0.12 (0.03, 0.20)	0.71	0.30		0.33 (0.25, 0.40)	0.63
WebFFQ	0.12		0.14 (0.05, 0.22)	0.71	0.27		0.29 (0.21, 0.37)	0.63
Single ASA24	0.14		0.16 (0.06, 0.26)	0.71	0.37		0.41 (0.33, 0.50)	0.63
Averaged ASA24	0.18		0.20 (0.11, 0.29)	0.71	0.47		0.54 (0.46, 0.60)	0.63
Averaged 7DDR	0.40		0.46 (0.37, 0.53)	0.71				

*ICC: Rank intraclass correlation coefficient for each reference method is presented for energy-adjusted intake using nutrient density method.

Table 2.4. Validity coefficients for energy, protein, sodium, potassium estimated by different methods

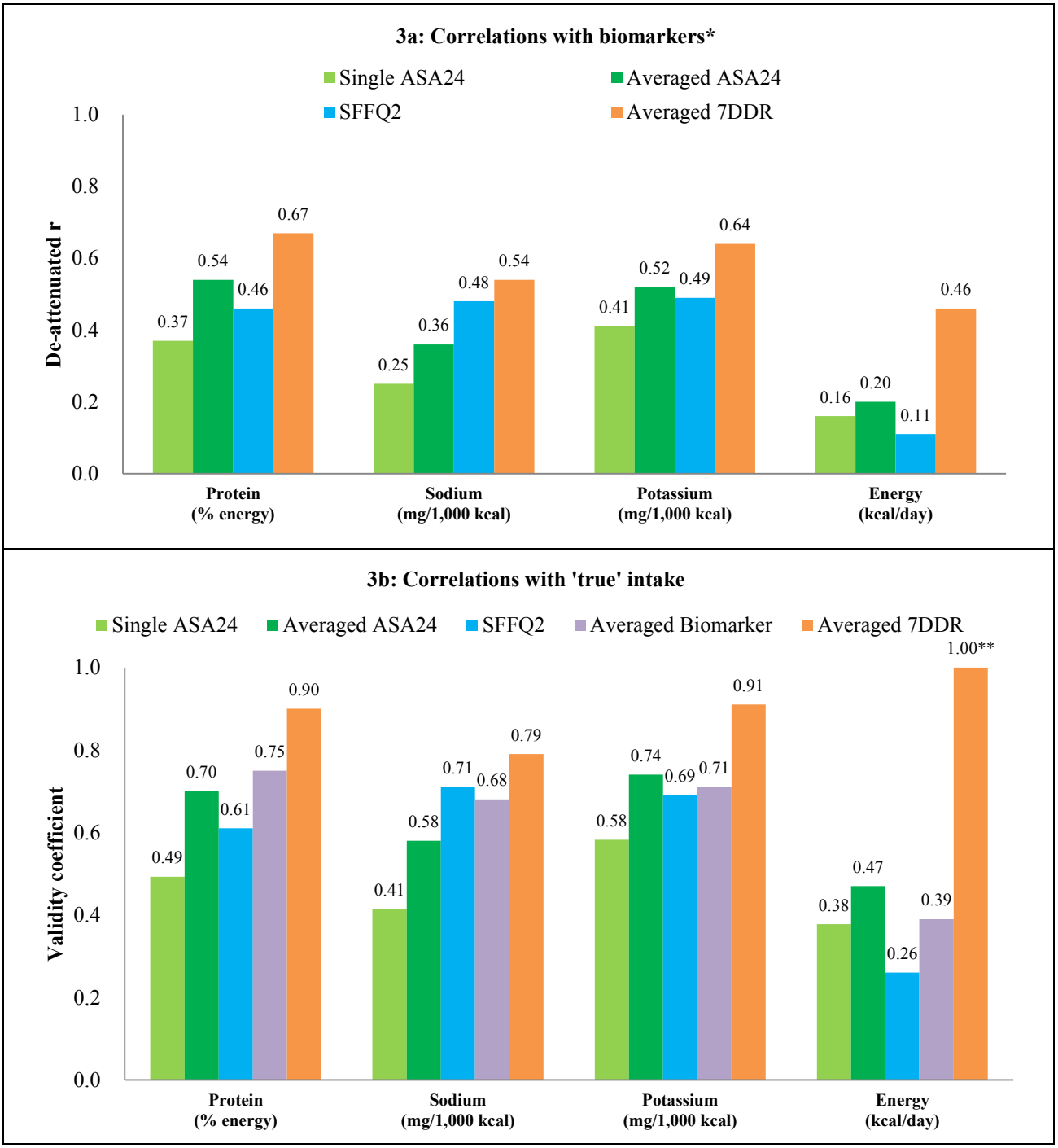
Nutrient	Validity coefficient (Method of triads)								
	Surrogate vs true			Averaged Biomarker vs true			Averaged 7DDR vs true		
	Absolute intake	Nutrient density	De-[a] attenuated	Absolute intake	Nutrient density	De-[a] attenuated	Absolute intake	Nutrient density	De-[a] attenuated
Protein									
SFFQ2	0.35	0.58	0.61	0.71	0.50	0.75	0.91	0.83	0.90
SFFQ1	0.40	0.60	0.63	0.72	0.52	0.79	0.90	0.81	0.85
SFFQ1&2	0.41	0.65	0.68	0.71	0.51	0.76	0.91	0.82	0.88
WebFFQ	0.37	0.56	0.61	0.71	0.50	0.76	0.92	0.84	0.89
Single ASA24	0.41	0.48	0.49	0.75	0.53	0.75	0.87	0.80	0.89
Averaged ASA24	0.53	0.64	0.70	0.73	0.52	0.78	0.89	0.81	0.86
Sodium									
SFFQ2	0.31	0.59	0.71	0.52	0.42	0.68	0.92	0.76	0.79
SFFQ1	0.34	0.55	0.62	0.53	0.42	0.67	0.91	0.76	0.81
SFFQ1&2	0.35	0.62	0.73	0.52	0.42	0.68	0.92	0.76	0.80
WebFFQ	0.29	0.60	0.67	0.52	0.43	0.68	0.93	0.74	0.79
Single ASA24	0.40	0.38	0.41	0.60	0.40	0.60	0.80	0.80	0.89
Averaged ASA24	0.52	0.49	0.58	0.56	0.39	0.62	0.86	0.83	0.87
Potassium									
SFFQ2	0.47	0.66	0.69	0.68	0.58	0.71	0.96	0.87	0.91
SFFQ1	0.49	0.59	0.61	0.67	0.57	0.70	0.97	0.87	0.91
SFFQ1&2	0.51	0.68	0.69	0.66	0.57	0.71	0.98	0.87	0.91
WebFFQ	0.45	0.57	0.61	0.70	0.57	0.69	0.92	0.88	0.92
Single ASA24	0.55	0.57	0.58	0.73	0.58	0.70	0.89	0.86	0.91
Averaged ASA24	0.67	0.73	0.74	0.72	0.59	0.70	0.91	0.85	0.91
Energy									
SFFQ2	0.25		0.26	0.37		0.39	1.00[b]		1.00[b]
SFFQ1	0.25		0.26	0.36		0.39	1.00[b]		1.00[b]
SFFQ1&2	0.28		0.29	0.36		0.39	1.00[b]		1.00[b]
WebFFQ	0.27		0.28	0.40		0.44	0.99		1.00[b]
Single ASA24	0.36		0.38	0.39		0.42	1.00[b]		1.00[b]
Averaged ASA24	0.45		0.47	0.38		0.40	1.00[b]		1.00[b]

Table 2.4 (continued): Validity coefficients for energy, protein, sodium, potassium estimated from different methods

Note: a. This de-attenuated validity coefficient was calculated based on the correlation coefficient de-attenuated for the variations in each reference method (biomarkers and 7DDR), as shown in Table 2.3.

b. Validity coefficients >1 (Heywood cases) were set to 1.00.

Figure 3. Spearman correlation coefficients with biomarkers and validity coefficients for protein, sodium, potassium and energy intakes



*Correlations were energy adjusted and de-attenuated for the random-within variations in the reference method.
**Validity coefficients >1 (Heywood cases) were set to 1.00.

Paper III

Validation of Nutrients Assessed by Questionnaire and 24-hour Recalls Among Women Compared With Intake by the Diet Records and Plasma Fatty Acids, Carotenoids, Retinol, Tocopherols and Folate

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Abstract

This study aimed to evaluate the performance of a semi-quantitative food frequency questionnaire (SFFQ) and a web-based 24-hour recall (ASA24) by comparing intakes of fatty acids, carotenoids, retinol, tocopherols and folate with plasma levels and 7-day weighed dietary records (7DDR) over a one-year period among 627 women. Three SFFQ's (paper SFFQ1 administered at the beginning; paper SFFQ2 and WebFFQ administered approximately one year later after other data collection) and four ASA24s, two 7DDRs, two fasting blood samples were obtained over approximately one year. SFFQ2 tended to overestimate absolute intakes of carotenoids, alpha-tocopherol and folate compared to 7DDR and ASA24s. The reproducibility was high for SFFQs (all nutrients included in our study), 7DDRs (except for several carotenoids), and plasma biomarkers (except saturated fatty acids and trans fat).The reproducibility of the ASA24 over one year was much lower compared to other methods. After correcting for random within-person variations in the reference method and adjustment for several dietary and non-dietary factors, Spearman correlation coefficients between SFFQ2 and biomarkers were weak for saturated fatty acids, MUFAs and retinol ($r<0.18$). Correlations were higher for other fatty acids, especially long chain N-3 fatty acids ($r=0.62$; $r=0.58$ among women without taking N-3 fatty acid supplements); and were modest-to-strong for carotenoids, alpha-tocopherol and folate, ranging from 0.33 for lycopene to 0.65 for alpha-tocopherol ($r=0.14$ among women who didn't take supplements for alpha-tocopherol). Further adjustment for systematic errors in the biomarkers using the method of triads gave substantially increased estimated correlations between nutrients assessed by SFFQ and true intake, especially for nutrient intakes not well reflected by the plasma concentrations (e.g. saturated fatty acid, MUFA and retinol). Estimated correlations with true intake were highest for intakes assessed by 7DDR's. Among women who

didn’t take supplements, most nutrients assessed by averaged ASA24 had lower correlations with biomarkers, 7DDRs and underlying ‘true’ intakes than did these nutrients assessed by SFFQ. These data indicate that the SFFQ provides reasonably valid measurements for specific fatty acid composition, most carotenoids, alpha-tocopherol and folate compared to concentration biomarkers or dietary records among women. The ASA24 needs further evaluation of its use in large population studies.

I. Introduction

Food frequency questionnaires (FFQs) can be self-administered, efficiently processed, and provide data on individual intakes of both foods and nutrients over an extended period of time; thus they have been the primary method of dietary assessment in large cohort studies.[1] An extensive literature on the validity of FFQs for measuring long-term diet has documented a strong concordance of FFQ with dietary records or 24-hour dietary recalls, but has mixed results with biomarkers of diet.[1,2]

The fundamental advantage of using biomarkers of diet as a reference method in validation studies is that errors affecting their measurement are generally uncorrelated with errors in self-reported dietary methods.[3] Up to date, the majority of intake biomarkers identified are 'concentration biomarkers', which are measures of concentration of a specific substance in plasma, adipose or other biological tissues. Unlike recovery biomarkers such as doubly labeled water, and urinary nitrogen (to assess protein intake), potassium, and sodium, which reflect absolute nutrient intakes over a certain time period, concentration biomarker levels reflect dietary composition and cannot be translated to absolute intakes because levels are influenced by numerous physiological and environmental factors in addition to diet. [3,4] Despite these limitations, concentration biomarkers can serve as an objective reference to assess the relative validity of different dietary assessment methods and to estimate correlations of these methods with true intake using the method of triads.[5,6 7]

Interpretation of findings from validation studies using concentration biomarkers need careful consideration of their sensitivity to intake, variability over time and time-integrating capacity, variation in diet by season, time frame of data collection, and the temporal relations of each dietary method.[3,6,8-10] Our group has evaluated the performance of the semi-quantitative FFQ

(SFFQ) against several concentration biomarkers in both the Nurse Health Study and Health Professional Follow Up Study and documented that intakes of specific carotenoids, fatty acids and vitamins are correlated with their corresponding biological fluid or tissue levels, which provide qualitative evidence of validity.[11-18] After modifications to incorporate changes in the food supply and eating patterns, our current version of this SFFQ includes 152 food items; we are using this in both a paper and a web-based format (WebFFQ). The web-based 24-hour recall (ASA24), recently developed by the National Cancer Institute (NCI),[19] is self-administered over the internet with minimal cost. The present study aimed to evaluate the validity of nutrient intakes measured by our SFFQs and by the ASA24 compared with plasma levels of fatty acids, carotenoids, retinol, and folate. Triangulation analyses including SFFQ (or ASA24), biomarkers, and two 7-day dietary records (7DDR) were also conducted to estimate the validity of each method with the underlying true intakes.

II. Materials and Methods

A. Study Population

The current study was based on the Women's Lifestyle Validation Study (WLVS), one of the three studies that comprise The Multi-Cohort Eating and Activity Study for Understanding Reporting Error (MEASURE), which was designed to investigate the measurement error structure associated with self-reported dietary and physical activity assessments, and how objective measures might be used to minimize error.[20] In 2010, we randomly selected a subset of Nurses' Health Study [21] and Nurses' Health Study II[22] participants aged 45-80 years, from all geographical regions of the US, who had completed the 2006/2007 cohort FFQ, had previously provided blood samples, had access to broadband internet, and were not planning to change their

diet. Women with medical history of coronary heart disease, stroke, cancer, or major neurological disease were excluded. The sample selection was stratified by age, and African-Americans were oversampled. A total of 5,509 female nurses were invited via email, by regular US mail method or both. Of these, 796 consented to participate, and received information on the timeline and activities. Study instructions were provided on the study website. Participants received support from study staff throughout their participation via the internet, telephone, and mail. Study participants were offered $600 for completion of the study due to the many complex activities. This study was approved by the Human Subjects Committees of the Harvard T.H. Chan School of Public Health and Brigham and Women's Hospital.

This analysis used plasma levels of biomarkers and 7-day dietary records (7DDR) to evaluate the performance of both our SFFQ and the ASA24s (see Figure 4). To represent the one-year period typically used as the time frame for dietary questionnaires, we spread the dietary and biomarker measurements over a period of approximately one-year and altered the order of measurements by dividing the participants into four groups. We collected one paper SFFQ (SFFQ1) at baseline, a second paper SFFQ (SFFQ2) and the WebFFQ (two weeks apart from the second paper SFFQ and in random order) at the end of data collection and approximately one-year after SFFQ1, one ASA24 in each season, and two 7DDRs and two blood samples 6 months apart to capture seasonal variability over one-year period. By design, three ASA24s were collected on weekdays and one on a weekend to capture differences in eating habits; within the same phase, the 7DDRs, ASA24s, blood samples were collected 1-5 weeks apart from each other and in random order to avoid artificially high correlations.[6,23]

Among 795 participants who completed the first activity (SFFQ1), 775 completed at least one fasting blood collection, 774 completed at least one 7DDR, 692 completed at least one ASA24,

759 completed SFFQ2, and 747 completed WebFFQ. Among 775 participants with fasting blood samples, 768 provided the first blood sample and 759 participants provided the second one. Of the 692 participants completing ASA24s, 93 (13%) competed one ASA24, 136 (20%) completed two, 219 (32%) competed three, and 244 (35%) completed four ASA24s. The distribution of ASA24 over the four phases was 512, 496, 497 and 493, respectively. Lower completion rates for the ASA24 were likely related, at least in part, to difficulties with the internet interface, which has been addressed in further versions of ASA24. In the quality control of SFFQ nutrient measurements, we excluded those participants with total daily energy intake <600 kcal or >3500 kcal or with more than 70 blanks. Overall, 771 SFFQ1 (97%), 742 SFFQ2 (98%), and 721 WebFFQ (97%) were included. These same restrictions for kilocalories and blank food items have been applied in analyses of diet and disease in the NHS and NHSII studies. Our primary analysis included 627 participants with complete data for SFFQ1, SFFQ2, WebFFQ, at least one 7DDR, one ASA24, and one fasting blood measurement. Information on year of birth, height, weight, ethnicity and smoking status was collected at enrollment. Weight information was also collected every four months during follow up.

B. The semi-quantitative food frequency questionnaire (SFFQ)

The 152-food-item semi-quantitative SFFQ was a slightly expanded version of the questionnaire previously validated in the NHS and Health Professionals Follow-up Study (www.channing.harvard.edu/nhs/questionnaires/pdfs/NHSII/2007.pdf).[1,24,25] Small modifications since 1986 were made to account for changes in the food supply (e.g., more low fat versions) based on pilot surveys conducted every 4 years. Respondents were asked how often, on average, they had consumed the specified amount of each type of food or beverage during the past year; nine possible frequency categories ranged from never/almost never to 6 or more times per day.

Participants also reported additional 3 foods (up to 3) not included on the questionnaire that they consumed more than once per week. Nutrient intakes were calculated from the questionnaire by multiplying a weight assigned to the frequency of use (where once per day is equal to one) by the nutrient composition for the portion size specified for each food or vitamin supplement based on an extensive and regularly updated food composition database (https://regepi.bwh.harvard.edu/health/nutrition/repeatUser.html) maintained at Harvard School of Public Health. The web-based version of this questionnaire was similar to the standard paper questionnaire and was enhanced by the use of branched questions.

C. **7-Day Weighed Dietary Records (7DDRs)**

WLVS research dietitians developed the 7DDRs with detailed instructions and a DVD instructional video. Each participant received an Escali food scale and ruler, the instructional DVD, and instructions via telephone by our research dietitians explaining how to keep detailed 7DDRs. A computerized reminder system was used to track participant study activities, sent reminder emails to participants on day 2, day 3 or 4 and day 6 of the diet recording week and to encourage participants to complete their diet record, to ask study research dietitians for clarifications on how to complete their diet records, and to review the DVD. Participants reported the gram weights for food intake and provided recipes of all home prepared foods including the number of recipe servings, and the portion of the recipes they each consumed. Participants also reported gram weights of foods before and after consumption, as relevant, so actual intake could be computed. Additionally, participants collected labels of store brand products and returned them with their records for analysis. A research dietitian contacted each participant during the recording week to discuss any questions and review procedures. Issues found in their first 7DDR provided after coding by the Nutrition Coordinating Center (NCC) at

the University of Minnesota[26] were specifically addressed during their second 7DDR instructional session with the research dietitian. The NCC used Nutrition Data System for Research (NDSR) 2011 to analyze the 7DDRs.[27,28] Nutrient intakes were calculated using the NCC food composition data base that is derived primarily from USDA sources.

D. Web-based, self-administered 24-hour dietary recall (ASA24)

Participants logged into the Beta version of ASA24 website to complete their 24hr recall entries using accounts created by the NCI. Participants were reminded on assigned days to complete the web-based recall by a computerized reminder system to ensure their readiness and availability to do so that day. The USDA's Food and Nutrient Database for Dietary Studies (FNDDS 4.1) was used to derive the nutrient data.[29] Daily energy (kcal/day), protein (g/day), sodium (mg/day) and potassium (mg/day, from food only) intakes were obtained from ASA24 data. The potassium intake from ASA24 didn't account for supplements; however, the amounts used were quite small and could be considered as ignorable compared to intake from the food.

E. Fasting blood collection

We collected plasma samples to measure specific fatty acids, specific carotenoids, retinol, tocopherols, and folate. Standard measures of blood lipids (total cholesterol, HDL, and triglycerides) were used adjust for variation in carotenoids, retinol, and tocopherols. Participants were asked to have their blood collected by a friend, colleague, or a local laboratory. Blood specimens were collected twice about six months apart. Participants were provided with a specimen collection kit to collect fasting blood samples. The first blood collection was 30 mL consisting of three 10 mL Heparin tubes, and the second blood collection was 40 mL consisting of four 10 mL Heparin blood tubes. Participants were asked to have their blood drawn in the

morning after fasting for 12 hours. For each blood sample, information on fasting status, blood collection time and date, menopausal status, smoking status, use of replacement hormones, physical activity, and weight,., was recorded. Participants' sample collection kits was picked-up by FedEx and shipped with a cold pack by overnight mail to Fisher BioServices, Inc. Then, whole blood samples were centrifuged for 20 minutes at 2500 RPM and 4 degrees C. Plasma was aliquoted into three small cryovials: two 0.5mL (one for folate, and one for carotenoids and fatty acids determinations), one 0.2mL (for lipids determinations).

Plasma fatty acids, expressed as percentages of total fatty acids, were measured by gas liquid chromatography in the laboratory of Dr. Campos at the Department of Nutrition, Harvard T.H. Chan School of Public Health as previously described.[30,31] Around 40 fatty acids were identified (mean coefficient of variation (CV) =18.1%). Our analysis focused on the specific fatty acids of greatest epidemiologic interest, recognizing that the concentrations of some in plasma, such as saturated and monounsaturated fatty acids, are minimally influenced by diet because they can be endogenously synthesized. Also, the saturated fat 15:0, 17:0, and trans fatty acid 16:1 were compared with dairy fat intake assessed by our SFFQ[32-35]. Plasma carotenoids (mean CV=9.7%), retinol (8.6%) and tocopherols (mean CV=9.3%) were measured by high-performance liquid chromatography as described by Hess [36] with some modifications. Because lutein and zeaxanthin co-elute on the chromatogram, the two are grouped and presented as lutein+zeaxanthin. Plasma folate (CV=9.2%) was determined by chemiluminescence using Immulite 1000 at the Vitamin Metabolism Laboratory at Tufts University. Blood lipids (total cholesterol, HDL cholesterol, and triglycerides) were determined enzymatically by the Lipid Metabolism Laboratory at Tufts University.

F. Statistical Analysis

A total of 28 specific fatty acids, 5 specific carotenoids, retinol and alpha- and gama-tocopherols, and folate intakes were used to evaluate the validity of the SFFQs and ASA24. Because nutrients estimated by ASA24 didn't include supplements, and blood concentrations reflect intakes from both foods and supplements, we conducted analyses limited to women who did not consume supplements for the nutrients to which supplements contributed.

Average daily intakes of fatty acids (% of total fatty acids), carotenoids, retinol, tocopherols and folate assessed by the SFFQs and ASA24s were compared with the corresponding nutrients assessed by plasma biomarkers and 7DDRs. Means and standard deviations for total daily nutrient intakes from SFFQ1, SFFQ2, WebFFQ, ASA24s (individual days1-4, and average days), 7DDRs (week1, week2 and average weeks), blood samples (blood1, blood2 and average) were calculated. Log transformation was performed for nutrients to increase normality because their distributions were skewed toward higher values. To remove variation in plasma nutrient levels due to non-dietary factors, we obtained fatty acid residuals from multivariate linear regression of each fatty acid composition on the following covariates: age (years), BMI (kg/m2) at enrollment; current weight (lbs), current smoking status (yes, no), menopausal status (yes, no) and use of hormone replacement within 6 months (yes, no), fasting status (>=8 hrs, <8 hrs) at each blood drawing; fat soluble carotenoids, retinol and tocopherol residuals were calculated by additionally adjusting for plasma lipids levels (total triglycerides, total cholesterol). Self-reported nutrient residuals were calculated using similar models adjusted for total energy intake, age, BMI at enrollment; current weight, and current smoking status. To further reduce the influence of the extreme nutrient distributions, the following analyses were based on the ranks of the log-transformed nutrient and multivariable-adjusted nutrient values.

To assess the reproducibility of repeated SFFQ's, ASA24's, 7DDR's and biomarkers, rank intraclass correlation coefficients (ICC) were calculated for each nutrient using both unadjusted and adjusted level. To assess nutrient validity, the averaged biomarker levels, or 7DDR intakes, repeated at an interval of around six months over a one-year period, were assumed as unbiased estimates of the true underlying nutrient intakes. Spearman correlation coefficients (r_0) between nutrient intakes based on each surrogate method (SFFQ2, SFFQ1, averaged SFFQ, WebFFQ, averaged ASA24), and the corresponding biomarkers or 7DDRs were calculated. Random within-person variation in the biomarkers or 7DDRs can attenuate correlations between these measures and the surrogate methods. Therefore, we calculated de-attenuated Spearman correlation coefficients (r_c) to reduce the effect of random within-person variation in the reference methods. The standard method first calculates the within- and between- person components of variation in nutrient intakes of the reference methods,[37,38] and corrects correlation coefficients for the within-person variation using $r_c = r_0\sqrt{(1 + \gamma / k)}$, where r_0 represent the observed correlation coefficient, r_c represents the corrected correlation coefficient, and γ equals the ratio of estimated within-person variation and between-person variation, k is the number of repeated observations of the reference methods.[37,38] As the number of biomarkers was unbalanced among individuals (costs precluded measurements on all samples), we used an extension of these methods [39] by Rosner & Glynn [40] and Perisic & Rosner [41] to correct for random within-person measurement error in the presence of unbalanced data and applied the method by Rosner and Willett [38] to calculate corresponding 95% CI for de-attenuated correlation coefficients.

To understand how temporal relations affected the validity of different methods, we examined correlations between each week of 7DDRs, or any one, or any two days of ASA24-assesed

intakes (e.g. ASA24, Day 1, Day 2, or averaged over Day 1 & 2) and each one, or two measurements of the biomarker for each nutrient. To further correct for systematic with-person errors (also referred to as person-specific bias) in the reference method, and simultaneously compare the relative performance of both surrogate methods and the reference methods, a triangular comparison (the method of triads) was used to estimate the correlation of each method with true intake. This method assumes that all dietary measurements were linearly related to true intake and that their measurement errors are mutually independent. [7] The following formulae were derived based on the factor analysis model, in which the validity coefficients (ρ) are calculated from the observed correlations (r) estimated above [7]:

$$\rho_{\mathrm{QT}} = \sqrt{r_{QR} \times r_{QB} / r_{RB}} \quad \rho_{\mathrm{RT}} = \sqrt{r_{QR} \times r_{RB} / r_{QB}} \quad \rho_{\mathrm{BT}} = \sqrt{r_{BQ} \times r_{BR} / r_{QR}},$$

where Q indicates SFFQ or ASA24 intakes, R indicates diet record intake, and B indicates biomarkers, T indicates true underlying intakes, ρ is the validity coefficient, and r is the observed correlation coefficient in the current analyses. In our case, we used the Spearman correlation coefficient (r). As noted elsewhere, correlations can be estimated as greater than 1.0 due to random errors; as suggested, these have been set to 1.0.[7]

In our SFFQ, two sets of fatty acid variables were available. One type was derived based primarily on USDA sources while the other one used updated analyses of commonly consumed brands and types of margarines , cooking fats, and processed foods conducted in the same laboratory as used for our plasma fatty acid analyses; the latter one was used in this study. 7DDR fatty acid variables were derived based on the NDSR2011 database; therefor, some specific isomers of certain fatty acids were not derived. The following results for fatty acids compositions focused on total saturated fatty acids, total monounsaturated fatty acids (MUFA), total

polyunsaturated fatty acids (PUFA), total trans fatty acids and several specific PUFA's(linoleic acid, alpha-linolenic acid, the long chain N-3 fatty acids). The results for all specific fatty acids are shown in the supplementary materials (Supplementary Table 3.1 & 3.2). Carotenoids and tocopherols analyzed in this study included alpha-carotene, beta-carotene, beta-cryptoxanthin, lutin-zeaxanthin, lycopene, alpha-tocopherol, and gama-tocopherol; we also included retinol. Plasma folate was measured in a subgroup of 459 women with two repeated measurements per individual. Dietary folate equivalent (sum of food natural folate and 1.7*total folic acid), which was justified by different bioavailability between folic acid and naturally occurring food folate, were compared with the plasma folate level. Nutrients assessed by ASA24 didn't account for supplements; however, the amounts used for most types of fatty acids (except N-3 fatty acid), alpha-carotene, beta-cryptoxanthin, lutin-zeaxanthin and lycopene were quite small and could be considered as ignorable compared to intake from the food. For long chain N-3 fatty acid, beta-carotene, retinol, alpha-tocopherol, and folate, we conducted separate analyses for women not taking the corresponding supplements.

III. Results

Descriptive statistics

Study participants had a mean age of 61 years and a mean BMI of 26.5 kg/m^2 at WLVS enrollment, 90.6% were white and 1.9% were current smokers. Among 627 participants who had at least one measurement of plasma fatty acids, carotenoids, retinol and tocopherols, 306 participants had two measurements of them; 459 participants had two measurements of plasma folate, and these subgroups had characteristics similar to the overall population (Table 3.1). The distributions of mean daily nutrient intakes and plasma concentrations are described in Table 3.2. Plasma concentrations for each nutrient could not be directly compared with the intakes

estimated by self-reported methods. Fatty acid (percentage of total fat) intakes estimated by each SFFQ, by two 7DDRs, and by four ASA24s, were approximately comparable. SFFQ2 tended to overestimate absolute intakes of carotenoids, alpha-tocopherol and folate compared to 7DDR and ASA24s (Table 3.2).

As shown in Table 3.3, we observed a high degree of reproducibility for all presented nutrient intakes including supplements by two FFQs spaced one-year apart (ICC>0.50). The reproducibility of unadjusted nutrient intakes measured by two 7DDRs with a six months interval was high for most fatty acids and folate and relatively low for many carotenoids. Most plasma nutrients concentrations were highly reproducible, except total saturated fat and trans fat, The reproducibility of the ASA24 over one year was much lower compared to other methods, indicating large within-person variation in nutrient intakes over the one-year interval.

Spearman correlation coefficients between intakes assessed by SFFQ or ASA24 and by biomarkers and 7DDRs

As expected, Spearman correlation coefficients between dietary saturated fatty acids and MUFAs assessed by SFFQ2 and corresponding plasma levels were weak even after correcting the random within-person variations in the reference method (r=0.15 and 0.08) (Supplementary Table 3.1 & Figure 5a). The correlations were higher for PUFAs (r=0.27), linoleic acid (r=0.25), trans fat (r=0.33), alpha-linolenic acid (r=0.34), and much higher for long chain N-3 fatty acids (r=0.62). For specific carotenoids, retinol and tocopherols, after adjusting nutrient intakes for total caloric intake and plasma nutrients for plasma lipid values, the de-attenuated correlations between SFFQ2 and biomarkers were lowest for gama-tocopherol, and retinol (all r<0.10), for other nutrients, the correlations ranged from 0.33 for lycopene to 0.65 for alpha-tocopherol. Dietary folate equivalents (r=0.54) were strongly correlated with plasma folate (Supplementary Table 3.3

& Figure 6a). The correlations of SFFQ2 with the mean of two 7DDRs were higher compared to the corresponding correlations when biomarkers were instead used as the reference method, although for many nutrients the difference was modest. Similar patterns of correlations were observed for WebFFQ and SFFQ1, although as expected the correlations were slightly lower with SFFQ1 because this referred to the year before collection of the biomarkers and 7DDRs. For nutrients with minimal contributions to intake by supplements, the de-attenuated and adjusted correlations between averaged ASA24 and biomarkers were 0.10 for saturated fat, 0.05 for MUFA, 0.27 for alpha-linolenic acid, 0.13 for linoleic acid, and 0.14 for PUFA, and 0.25-0.45 for most carotenoids. Among women who didn't take supplements, averaged ASA24 performed slightly better than SFFQ2 in estimating alpha-tocopherol and folate. However, compared to nutrients assessed by SFFQ2, correlations for almost all nutrients assessed by averaged ASA24s were lower with 7DDRs and biomarkers (Figure 5a & 6a, Supplementary Table 3.1, 3.3 & 3.6).

When comparing each single 7DDR, single ASA24, and the average intake of different combinations of two ASA24s, with each single and two measurements of biomarkers, we didn't observe an obvious pattern in which correlations of the biomarker measured closer in time to the 7DDRs or ASA24 were higher (Supplementary Tables 3.7-3.10).

Validity Assessed by the Method of Triads

The validity coefficients for each dietary assessment method are shown in Supplementary Tables 3.2, 3.4 and Figure 5b & 6b. After accounting for potential correlated errors between repeated assessments of the reference methods, the estimated validity coefficients for SFFQs and ASA24s with true intake were higher than the observed correlations, especially for nutrients with very low correlations between intakes and biomarkers, such as saturated fat, MUFA, lycopene and

retinol. Relative to the underlying true intakes, the validity coefficients were highest for SFFQs in estimating the MUFA, alpha-linolenic acid, long chain N-3 fatty acid, linoleic acid, PUFA, all carotenoids, alpha-tocopherol, and folate intakes (de-attenuated $\rho >= 0.64$). 7DDRs had the highest validity coefficients for almost all nutrients. For saturated fat, MUFA, retinol, and gama-tocopherols intake, biomarkers had very lower validity coefficients (de-attenuated $\rho < 0.30$), consistent with expectations. The averaged ASA24 had lower validity coefficients compared to SFFQ2 for almost all nutrients except for several nutrients without supplements.

IV. Discussion

Utilizing concentration biomarkers for various nutrients including fatty acids, carotenoids, retinol, tocopherols and folate, we evaluated the performance of our semi-quantitative FFQ, the web-based ASA24s and two 7DDRs kept over a one-year period among women participating in the Nurses' Health Studies. Correlations between nutrient intakes assessed by SFFQ and blood concentrations varied from weak to high after adjusting for energy and several dietary and non-dietary factors and correcting for random within-person variation in the biomarkers. Further adjustment for systematic errors in the biomarkers using the method of triads substantially increased estimates of correlations with true intake, especially for dietary nutrient intakes not well reflected by the plasma concentrations (e.g. saturated fatty acid, MUFA and retinol). Higher correlations were observed between SFFQ2 and averaged 7DDRs, and 7DDRs had the highest validity correlations. Compared to SFFQ, almost all nutrients estimated by an average of 3 days ofASA24 had relatively low correlations with biomarkers, 7DDRs and estimated 'true' underlying intakes. These finding greatly extend the comparison of different dietary assessment methods using only recovery biomarkers[42-48], which exist for only a very limited number of nutrients.

Although concentration biomarkers of dietary intake are relatively objective measurements, they are likely to be influenced by factors including individual differences in the nutrient absorption, metabolism, endogenous synthesis, interactions with other nutrients, turn over in the targeted tissue, day-to-day or seasonal variations in dietary intakes, or technical error associated with laboratory measurement. The overall effect of all these sources of error is to weaken correlations between a dietary questionnaire used to assess long-term intake and the biochemical indicator. Therefore, a modest correlation with the concentration biomarkers does not necessary mean that the dietary measurements are not accurate and precise.[3,6] Our study presented the pair-wise correlations between different dietary assessments methods, mainly focused on specific fatty acids, specific carotenoids, retinol, tocopherols, and folate; these nutritional factors have been hypothesized to be related to risks of multiple diseases and address the validity of different aspects of diet. To remove variation in plasma nutrient levels due to some non-dietary factors, we obtained nutrients residuals adjusted for various non-dietary factors (e.g. age, BMI, smoking status), and plasma lipids level (for fat soluble nutrients). To focus on dietary composition, which is of primary interest in nutritional epidemiology, we also adjusted for total energy intake. [1,49] To account for random within-person variations in the reference methods, we calculated de-attenuated correlations; To further adjust for systematic errors in those reference methods, we used methods of triads to estimate the validity coefficients which reflected the degree of correlations with the underlying 'true intakes'. The validity coefficient from the 'method of triads' can be interpreted as the upper limits of the true validity coefficient, as there may be non-ignorable correlated positive errors between SFFQ or ASA24s and 7DDRs, which tended to overestimate the validity coefficient of SFFQs and ASA4s.[7,50] However, by design, we avoided collecting biomarkers and short term dietary measures at the same time (at least several weeks

apart) and we did not find evidence of residual important temporal relations between methods. Also, we found similar de-attenuated correlations between SFFQ and intakes assessed by ASA24 and 7DDRs, and these reference methods have very different sources of error. Therefore, the validity correlation coefficients were less subject to the overestimation due to correlated errors between different dietary methods.

In our study, we observed weak correlations for saturated fatty acid, MUFA and several other endogenously synthesized fatty acids, whereas fatty acids with mainly exogenous origin had higher correlations between SFFQ and biomarkers. Correlations ranged from 0.25 for linoleic acid to 0.65 for long chain N-3 fatty acids with supplements. Previous studies also reported modest-to-strong correlations for N-3 fatty acid, trans fat, and linoleic acid using biomarkers measured in plasma [51], erythrocytes [52] or adipose tissue.[9,11,53] For carotenoids, alpha-tocopherol and folate, we observed high correlations between SFFQ and biomarkers (r ranged from 0.33 for lycopene to 0.65 for alpha-tocopherol), similar to most existing studies.[6,8] Our study also found that multiple ASA24s had lower correlations with biomarkers, compared to nutrients estimated by SFFQs; this may be due to large day-to-day variations in nutrient intakes, whereas an average of 2.9 days of ASA24 collected in our study did not sufficiently capture the long-term variations.

Because concentration biomarkers have its own errors in representing long-term dietary intake, we performed triangulation analyses to compare the validity coefficients for different methods. Correlations of SFFQs and ASA24s with true intake were higher than the observed correlations with biomarkers. Among different dietary assessment methods, SFFQs and 7DDRs had very high validity correlations, while both biomarkers and ASA24 have large measurement errors. In particular, biomarkers had very low validity correlations for saturated fatty acids, MUFA, retinol,

delta- and gama-tocopherols, which also lead to the low correlations between nutrients by SFFQs and these concentrations biomarkers.

To the best of our knowledge, this is the largest validation study of multiple dietary assessment methods using concentration biomarkers for many different nutrients. By design, all short-term dietary methods were repeated at reasonable time intervals (at least three months apart) to capture the variation over one year, and those methods were not collected close in time (1-5 weeks apart from each other and in random order) to avoid artificially high correlations. Future validation studies should consider these temporal relationships when being designed.[6] Our study also has some limitations. Our analysis for ASA24 is based on nutrients only from food, which could not provide a complete evaluation of its overall performance. Furthermore, because all standards for assessment of validity have errors, the validity of questionnaires cannot be known exactly. However, the use of multiple methods provides a range of validity that can be useful. Our findings confirmed that most concentration biomarker should not be considered to be without error, including systematic with-person error.

Among the intake methods used in this study, the two 7DDR's had the strongest correlations with biomarkers of intake, presumably due to a total of 14 days of open-ended, weighed and recorded intake and the detailed coding of foods. Unfortunately the high costs and participant burden of this method preclude use in large studies, particularly if repeated assessments over time are needed. Multiple ASA24 didn't perform as well as our dietary questionnaire, future studies could evaluate the optimal number of ASA24 needed to be comparable to FFQs; Furthermore, participants had the most difficulty completing the requested four ASA24 assessments; whether this can be improved by software enhancements needs to be evaluated. In addition to the limitation that concentration biomarkers subject to many potential errors, the lack

of any marker for many dietary factors of major interest, in part due to strong homeostatic regulation, constrains their use as standards in a validation study. Further validation studies could incorporate additional dietary biomarkers and evaluate the changes of biomarkers levels in response to changes in diet.

In conclusion, our data document that the semi-quantitative food frequency questionnaire provided reasonably valid measurements for specific fatty acid composition, most carotenoids, and folate compared to concentrations biomarkers or dietary records among women. Multiple automated self-administered 24-hour recalls didn't perform as well as the SFFQ in estimating these nutrients; the potential to be used as a dietary assessment method in large population studies needs further evaluation. Future analyses are needed to address other important practical questions including the number of ASA24s needed, the appropriate time interval between each measurement to be representative of long-term dietary intake, and the potential improvement by combining its use with other dietary assessment methods .

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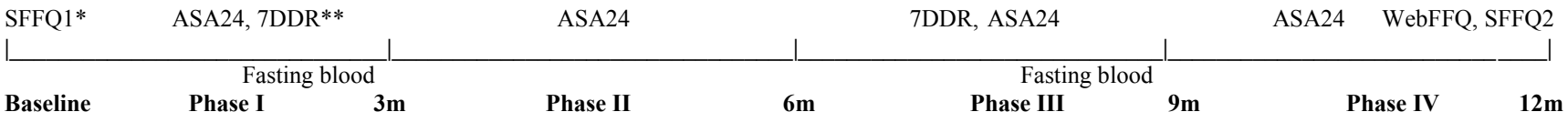
VI. Figures and Tables

Figure 4. Timeline of the dietary assessment activity in project 3



* This figure showed the timeline for group 1 participants; group 3 went through the same data collection timeline as group 1; groups 2 and 4 went through similar data collection timeline as group 1, except groups 2 and 4 participants completed the 7DDR and fasting blood collections in phase II and IV.
**Within the same phase, 7DDRs and ASA24s, SFFQ2 and WebFFQ were in random order; 7DDR, ASA24 and fasting blood were collected 1-5 weeks apart from each other to avoid artificially high correlations.

Table 3.1. Subject characteristics among WLVS participants in project 3

Variables	Overall (n=627)	Fatty acid and carotenoid biomarkers		Folate biomarker
		One blood (n=321)	Two bloods (n=306)	Two bloods (n=459)
Age at enrollment, mean(sd), year	61.4 (9.5)	62.3 (9.9)	60.5 (9.1)	60.8 (9.3)
Height, mean (sd), meter	1.64 (0.07)	1.64 (0.07)	1.64 (0.07)	1.64 (0.07)
Weight at baseline, mean (sd), kg	71.5 (15.5)	71.6 (15.6)	72.7 (15.5)	71.6 (15.3)
Weight change at year 1, mean (sd), kg	-0.2 (2.6)	-0.1 (2.6)	-0.3 (2.6)	-0.2 (2.7)
BMI at baseline, mean(sd), kg/m2	26.5 (5.4)	26.6 (5.4)	26.9 (5.4)	26.5 (5.4)
White, %	90.6	92.8	88.2	90.6
Current smokers, %	1.9	1.0	3.0	2.2
Use of hormone replacement in the past 6 months, %	24.4	24.6	24.3	23.7
Menopause, %	83.8	84.2	83.3	83.3
Plasma total triglycerides, mean(sd), mg/dl	112 (53)	111 (53)	114 (54)	112 (54)
Plasma total cholesterol, mean(sd), mg/dl	214 (36)	210 (35)	218 (36)	211 (35)
Plasma HDL cholesterol, mean(sd), mg/dl	70 (16)	70 (17)	69 (16)	69 (16)
Plasma TG/HDL ratio, mean(sd), mg/dl	1.8 (1.3)	1.8 (1.3)	1.8 (1.4)	1.8 (1.4)

Note: data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012
sd: standard deviation

Table 3.2. Distribution of specific fatty acids, carotenoids, retinol, tocopherols and folate

Nutrients	Biomarker	SFFQ	7DDR	ASA24
	Blood 1&2 (N=627)	SFFQ2 (N=627)	week1&2 (N=627)	days, all (N=627)
Fatty acids (% total fatty acids or fat)				
Saturated fatty acids	31.4 ± 2.7	35.6 ± 6.2	33.1 ± 4.7	33.8 ± 6.2
Monounsaturated fatty acids	22.2 ± 2.8	38.0 ± 4.2	36.1 ± 3.1	35.6 ± 3.8
Polyunsaturated fatty acids				
Alpha-linolenic acid	0.6 ± 0.2	2.3 ± 1.1	2.3 ± 1.0	2.2 ± 1.0
Long Chain N-3 (DHA,DPA,EPA)	3.5 ± 1.6	0.8 ± 0.7	0.5 ± 0.7	N/A
Long Chain N-3* (N=363)	2.8 ± 1.0	0.4 ± 0.4	0.3 ± 0.3	0.3 ± 0.6
Linoleic acid (18:2n-6cc)	30.0 ± 3.7	18.1 ± 3.7	19.5 ± 3.7	19.0 ± 4.9
Arachidonic acid (20:4n-6c)	8.2 ± 1.9	0.2 ± 0.1	0.2 ± 0.1	0.2 ± 0.1
Total PUFA	44.8 ± 4.0	22.2 ± 4.6	22.4 ± 4.2	21.9 ± 5.6
Trans Fatty acids	1.1 ± 0.4	2.7 ± 0.7	3.3 ± 1.1	N/A
Carotenoids (ug/L or ug/day)				
Alpha carotene	101 ± 94	855 ± 767	613 ± 490	498 ± 771
Beta carotene	397 ± 398	6498 ± 4055	4291 ± 2828	N/A
Beta carotene* (N=335)	348 ± 349	6342 ± 3844	3987 ± 2707	3434 ± 2966
Beta cryptoxanthin	160 ± 142	111 ± 96	154 ± 182	100 ± 138
Lutein-zeaxanthin	284 ± 146	3926 ± 3182	2700 ± 2799	2598 ± 2940
Lycopene	431 ± 172	5613 ± 3987	4986 ± 3570	5091 ± 5412
Retinol activity equivalents (ug/L or day)	620 ± 136	1810 ± 1302	1480 ± 1297	N/A
Retinol activity equivalents* (N=207)	591 ± 126	931 ± 383	787 ± 399	762 ± 605
Tocopherols (mg)				
Alpha- tocopherol	14.1 ± 4.8	42.3 ± 71.9	37.3 ± 54.1	N/A
Alpha- tocopherol * (N=145)	11.4 ± 2.7	9.1 ± 4.5	9.0 ± 3.6	7.8 ± 3.9
Gama - tocopherol	1.4 ± 0.8	10.1 ± 4.3	11.2 ± 3.9	N/A
Folate (ng/ml or ug/day) (N=459)				
Dietary folate equivalents	31.9 ± 21.6	1150 ± 664	1073 ± 629	N/A
Dietary folate equivalents* (N=135)	19.2 ± 16.1	557 ± 374	517 ± 216	487± 211

*Subgroups of women who didn't take supplements for this nutrient.
Note: PUFA: Polyunsaturated fatty acids.

Table 3.3. Reproducibility of nutrient intakes estimated by SFFQ, 7DDR and ASA24 and biomarkers

Variables	Biomarker		SFFQ		7DDR		ASA24	
	Un-adjusted	Adjusted*	Un-adjusted	Adjusted*	Un-adjusted	Adjusted*	Un-adjusted	Adjusted*
Fatty acids (% total fatty acids or fat)								
Saturated fatty acids	0.21	0.20	0.65	0.64	0.58	0.58	0.19	0.19
Monounsaturated fatty acids	0.69	0.64	0.62	0.62	0.41	0.41	0.11	0.11
Polyunsaturated fatty acids								
Alpha-linolenic acid	0.59	0.57	0.59	0.58	0.48	0.47	0.18	0.17
Long Chain N-3 (DHA+DPA+EPA)	0.78	0.75	0.75	0.73	0.61	0.59	N/A	N/A
Long Chain N-3** (N=363)	0.67	0.61	0.69	0.67	0.53	0.51	0.11	0.12
Linoleic acid	0.58	0.53	0.54	0.53	0.44	0.44	0.14	0.13
Total PUFA	0.43	0.38	0.58	0.58	0.46	0.46	0.15	0.14
Trans Fatty acids	0.25	0.23	0.65	0.65	0.41	0.40		
Carotenoids (ug/L or day)								
Alpha carotene	0.83	0.77	0.69	0.64	0.37	0.35	0.15	0.12
Beta carotene	0.86	0.81	0.70	0.65	0.47	0.43	N/A	N/A
Beta carotene** (N=335)	0.85	0.78	0.69	0.68	0.52	0.48	0.25	0.16
Lutein-zeaxanthin	0.81	0.75	0.71	0.68	0.53	0.50	0.24	0.20
Beta cryptoxanthin	0.77	0.71	0.74	0.70	0.32	0.29	0.19	0.14
Lycopene	0.65	0.56	0.63	0.55	0.24	0.21	0.11	0.11
Retinol activity equivalents (ug/L or day)	0.68	0.64	0.68	0.67	0.65	0.64	N/A	N/A
Retinol activity equivalents** (N=207)	0.63	0.48	0.54	0.55	0.48	0.32	0.21	0.15

Table 3.3 (continued): Reproducibility of nutrient intakes estimated by SFFQ, 7DDR and ASA24 and biomarkers

Variables	Biomarker		SFFQ		7DDR		ASA24	
	Un-adjusted	Adjusted*	Un-adjusted	Adjusted*	Un-adjusted	Adjusted*	Un-adjusted	Adjusted*
Tocopherols (mg)								
Alpha- tocopherol	0.63	0.53	0.71	0.71	0.78	0.77	N/A	N/A
Alpha- tocopherol * (N=145)	0.56	0.44	0.46	0.49	0.58	0.44	0.24	0.21
Gama - tocopherol	0.71	0.67	0.66	0.60	0.44	0.38	N/A	N/A
Folate (ng/ml or ug/day) (N=459)								
Dietary folate equivalents	0.70	0.67	0.66	0.63	0.78	0.77	N/A	N/A
Dietary folate equivalents** (N=135)	0.73	0.69	0.40	0.35	0.69	0.61	0.21	0.18

* Nutrients were adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; self-reported nutrients were further adjusted for total energy intake (except for fatty acid composition); plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing; Plasma carotenoids, retinol and was further adjusted for plasma lipids. **Subgroups of women who didn't take supplements for this nutrient.

Figure 5. Spearman correlation coefficients and validity coefficients for specific fatty acids

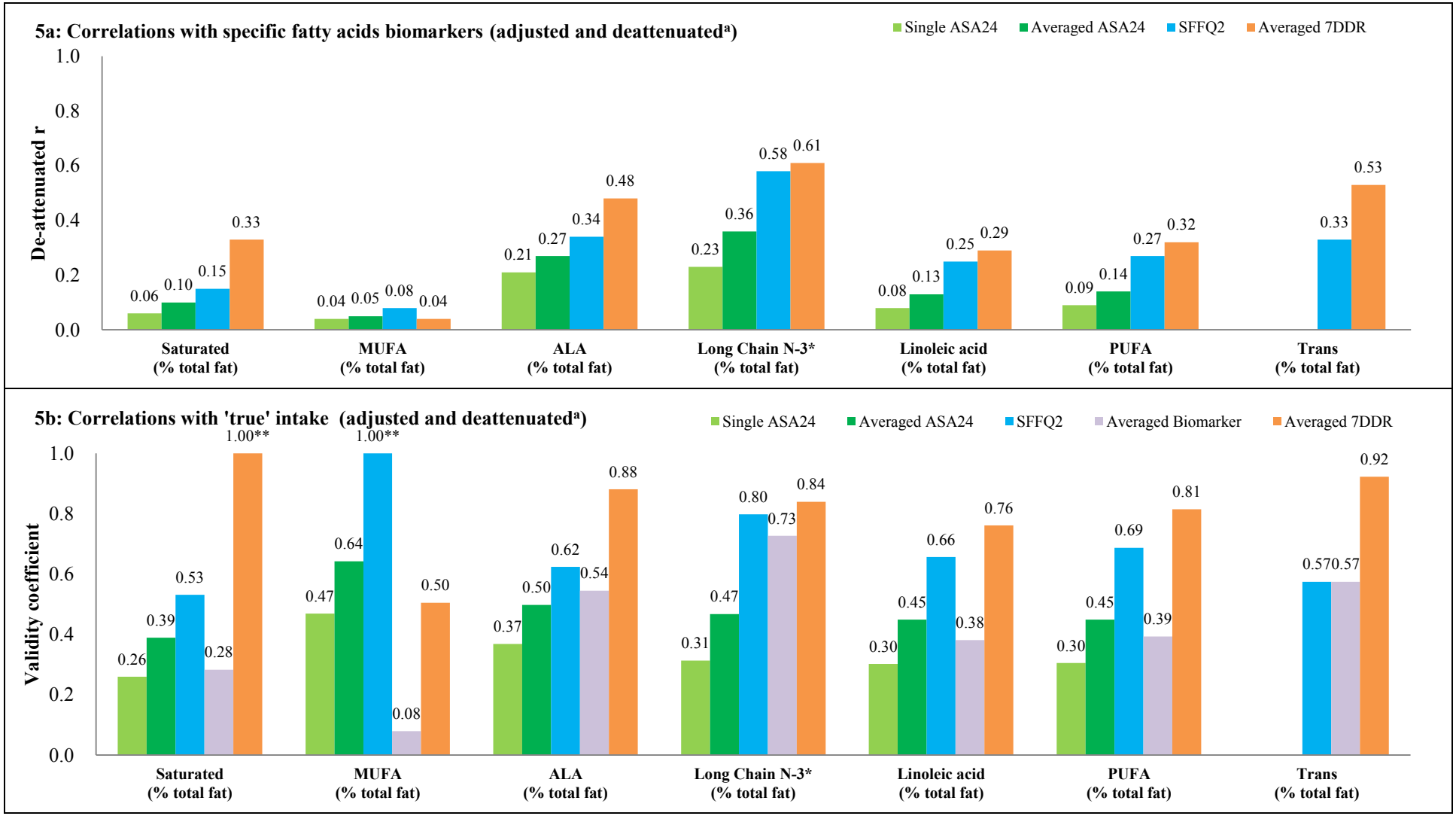


Figure 5 Footnote

a: Nutrients were adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing; correlations were de-attenuated for the random-within variations in the reference method.

[*] Subgroups of women who didn't take supplements for this nutrient (N=363).

[**] Validity coefficients >1 (Heywood cases) were set to 1.00.

Figure 6. Spearman correlation coefficients and validity coefficients for specific carotenoids, retinol, tocopherols and folate

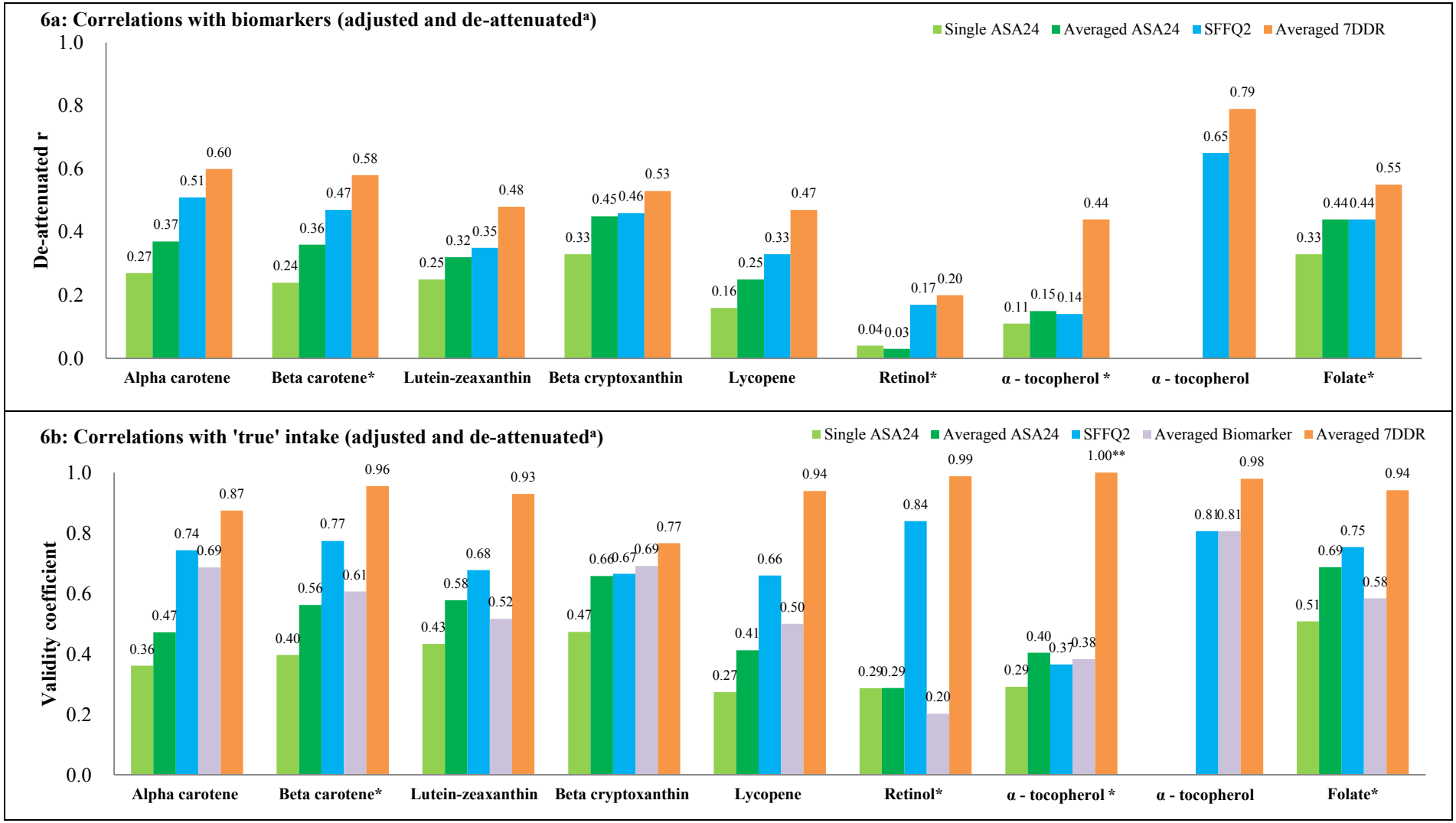


Figure 6 Footnote

a: Nutrients were adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; self-reported nutrients were further adjusted for total energy intake; plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing; Plasma carotenoids, retinol and was further adjusted for plasma lipids; correlations were de-attenuated for the random-within variations in the reference method.

[*] Subgroups of women who didn't take supplements for this nutrient (N=335 for beta-carotene, 207 for retinol, 145 for alpha-tocopherol, and 135 for folate).

[**] Validity coefficients >1 (Heywood cases) were set to 1.00.

Supplementary Tables

Supplementary Table 1.1a: Mean absolute daily nutrient intakes estimated by SFFQs, two 7DDRs, and four ASA24s, nutrients not included in the main table 1.2 (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ2		SFFQ1		WebFFQ		7DDR		ASA24	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Butyric FA (g)	0.46	(0.28)	0.47	(0.29)	0.45	(0.26)	0.54	(0.28)	0.62	(0.43)
Capric FA (g)	0.36	(0.21)	0.37	(0.21)	0.35	(0.2)	0.47	(0.25)	0.51	(0.35)
Caproic FA (g)	0.24	(0.15)	0.24	(0.16)	0.23	(0.14)	0.27	(0.16)	0.32	(0.22)
Caprylic FA (g)	0.18	(0.11)	0.18	(0.11)	0.17	(0.1)	0.27	(0.17)	0.27	(0.20)
Gadoleic FA (g)	0.24	(0.12)	0.25	(0.13)	0.23	(0.12)	0.21	(0.09)	0.21	(0.12)
Myristic FA (g)	1.7	(0.9)	1.8	(0.9)	1.7	(0.8)	2.1	(0.9)	2.2	(1.3)
Palmitic FA (g)	12.3	(4.5)	12.6	(4.6)	11.8	(4.4)	11.6	(3.5)	12.8	(5.1)
Palmitoleic FA (g)	1.0	(0.4)	1.0	(0.4)	1.0	(0.4)	1.0	(0.4)	1.1	(0.5)
Stearic FA (g)	5.5	(2.4)	5.6	(2.4)	5.3	(2.3)	5.3	(1.8)	6.2	(3.0)
Total folate (mcg)	458	(181)	478	(170)	443	(159)	388	(122)	376	(147)
With supplements	743	(355)	772	(323)	722	(308)	671	(366)		
Alpha carotene with supplements (mcg)	855	(766)	920	(872)	850	(767)	611	(489)		
Lutein-zeaxanthin with supplements (mcg)	3925	(3178)	3877	(2638)	3818	(2523)	2702	(2788)		
Lycopene with supplements (mcg)	5602	(3980)	5743	(4487)	5713	(4610)	4976	(3565)		
Beta cryptoxanthin with supplements (mcg)	111	(96)	120	(103)	107	(94)	154	(181)		

Supplementary Table 1.1b: Rank intraclass correlations of one-week diet records, the semi-quantitative food frequency questionnaire and ASA24, nutrients not included in the main table 1.3 (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ			Record			ASA24		
	Un-adjusted	Energy density	Residual method	Un-adjusted	Energy density	Residual method	Un-adjusted	Energy density	Residual method
Butyric FA (g)	0.66	0.62	0.61	0.59	0.55	0.53	0.25	0.22	0.19
Capric FA (g)	0.68	0.65	0.65	0.58	0.53	0.52	0.26	0.23	0.21
Caproic FA (g)	0.67	0.63	0.63	0.59	0.56	0.55	0.26	0.23	0.21
Caprylic FA (g)	0.66	0.64	0.64	0.54	0.49	0.48	0.23	0.20	0.18
Gadoleic FA (g)	0.64	0.60	0.60	0.44	0.38	0.38	0.15	0.11	0.11
Myristic FA (g)	0.68	0.64	0.64	0.61	0.57	0.55	0.25	0.21	0.19
Palmitic FA (g)	0.71	0.68	0.68	0.65	0.66	0.65	0.28	0.25	0.25
Palmitoleic FA (g)	0.68	0.61	0.61	0.62	0.58	0.58	0.24	0.22	0.21
Stearic FA (g)	0.70	0.70	0.70	0.64	0.64	0.64	0.27	0.25	0.24
Total folate (mcg)	0.62	0.59	0.58	0.61	0.58	0.58	0.25	0.22	0.22
With supplements	0.65	0.65	0.64	0.74	0.72	0.74			
Alpha carotene with supplements (mcg)	0.69	0.65	0.65	0.37	0.37	0.37			
Lutein-zeaxanthin with supplements (mcg)	0.71	0.69	0.69	0.52	0.51	0.52			
Lycopene with supplements (mcg)	0.63	0.56	0.56	0.24	0.23	0.23			
Beta cryptoxanthin with supplements (mcg)	0.74	0.72	0.72	0.31	0.31	0.31			

Supplementary Table 1.1c: Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with 7DDRs, and ASA24s, unadjusted and energy-adjusted using energy density and residual method, nutrients not included in the main table 1.4 (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR						SFFQ2 vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Butyric FA (g)	0.53	0.55	0.52	0.63	(0.55, 0.69)	0.53	0.45	0.44	0.39	0.60	(0.47, 0.69)	0.19
Capric FA (g)	0.48	0.50	0.48	0.58	(0.50, 0.64)	0.52	0.42	0.40	0.37	0.56	(0.44, 0.65)	0.21
Caproic FA (g)	0.55	0.56	0.53	0.63	(0.56, 0.69)	0.55	0.48	0.47	0.42	0.63	(0.52, 0.72)	0.21
Caprylic FA (g)	0.42	0.42	0.41	0.50	(0.41, 0.57)	0.48	0.39	0.35	0.32	0.51	(0.38, 0.61)	0.18
Gadoleic FA (g)	0.26	0.33	0.33	0.44	(0.35, 0.53)	0.38	0.16	0.23	0.24	0.45	(0.30, 0.59)	0.11
Myristic FA (g)	0.48	0.52	0.51	0.60	(0.53, 0.67)	0.55	0.44	0.43	0.42	0.65	(0.53, 0.74)	0.19
Palmitic FA (g)	0.42	0.62	0.62	0.70	(0.64, 0.74)	0.65	0.42	0.59	0.56	0.74	(0.63, 0.82)	0.25
Palmitoleic FA (g)	0.41	0.51	0.51	0.58	(0.51, 0.65)	0.58	0.36	0.43	0.43	0.64	(0.53, 0.72)	0.21
Stearic FA (g)	0.43	0.59	0.60	0.68	(0.61, 0.73)	0.64	0.44	0.55	0.51	0.70	(0.60, 0.79)	0.24
Total folate	0.44	0.54	0.55	0.63	(0.56, 0.69)	0.58	0.35	0.41	0.41	0.57	(0.46, 0.66)	0.22
With supplements	0.66	0.69	0.70	0.76	(0.71, 0.80)	0.74						
Alpha carotene with supplements (mcg)	0.49	0.48	0.47	0.64	(0.54, 0.71)	0.37						
Lutein-zeaxanthin with supplements (mcg)	0.51	0.53	0.52	0.63	(0.56, 0.70)	0.52						
Lycopene with supplements (mcg)	0.40	0.38	0.37	0.61	(0.45, 0.72)	0.23						
Beta cryptoxanthin with supplements (mcg)	0.36	0.38	0.38	0.55	(0.43, 0.64)	0.31						

Supplementary Table 1.2: Pearson correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR						SFFQ2 vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Total energy (kcal)	0.30			0.33	(0.25, 0.41)	0.64	0.32			0.43	(0.33, 0.52)	0.30
Total fat (g)	0.37	0.62	0.60	0.68	(0.62, 0.73)	0.67	0.35	0.58	0.56	0.77	(0.64, 0.85)	0.22
Saturated fat (g)	0.46	0.63	0.63	0.70	(0.65, 0.75)	0.67	0.46	0.55	0.58	0.74	(0.63, 0.82)	0.25
Polyunsaturated fat (g)	0.30	0.47	0.49	0.60	(0.52, 0.67)	0.50	0.25	0.44	0.44	0.74	(0.55, 0.86)	0.13
Monounsaturated fat (g)	0.39	0.52	0.51	0.59	(0.52, 0.65)	0.61	0.32	0.43	0.42	0.63	(0.51, 0.73)	0.20
Arachadonic FA (g)	0.43	0.39	0.48	0.57	(0.49, 0.64)	0.54	0.31	0.29	0.36	0.49	(0.32, 0.63)	0.14
Lauric FA (g)	0.36	0.30	0.37	0.47	(0.37, 0.55)	0.45	0.35	0.15	0.33	0.48	(0.36, 0.59)	0.21
Linoleic FA (g)	0.31	0.45	0.47	0.58	(0.49, 0.65)	0.50	0.26	0.43	0.44	0.75	(0.56, 0.87)	0.12
Linolenic FA (g)	0.38	0.57	0.52	0.64	(0.56, 0.71)	0.50	0.29	0.41	0.37	0.61	(0.46, 0.72)	0.15
N-3 (DHA+EPA) FA (g)	0.54	0.43	0.54	0.70	(0.61, 0.77)	0.44	0.24	0.30	0.28	0.57	(0.32, 0.75)	0.08
With supplements	0.65	0.46	0.64	0.73	(0.67, 0.79)	0.61						
Oleic FA (g)	0.39	0.51	0.50	0.58	(0.51, 0.64)	0.60	0.32	0.42	0.41	0.63	(0.50, 0.73)	0.19
Cholesterol (mg)	0.52	0.56	0.58	0.68	(0.61, 0.74)	0.59	0.40	0.40	0.42	0.73	(0.57, 0.84)	0.14
Protein (g)	0.34	0.50	0.50	0.56	(0.48, 0.62)	0.63	0.31	0.39	0.39	0.56	(0.45, 0.66)	0.22
Carbohydrate (g)	0.44	0.69	0.71	0.75	(0.70, 0.79)	0.78	0.46	0.61	0.65	0.78	(0.71, 0.84)	0.39
Total sugar (g)	0.56	0.69	0.71	0.77	(0.73, 0.81)	0.77	0.56	0.64	0.64	0.77	(0.70, 0.83)	0.38
Fiber (g)	0.48	0.64	0.64	0.68	(0.62, 0.73)	0.75	0.42	0.53	0.53	0.67	(0.58, 0.75)	0.32
Alcohol (g)	0.73	0.83	0.72	0.82	(0.77, 0.86)	0.65	0.60	0.75	0.59	0.67	(0.61, 0.73)	0.51
Retinol activity eqvts (mcg)	0.45	0.32	0.49	0.68	(0.57, 0.76)	0.35	0.39	0.51	0.41	0.59	(0.47, 0.69)	0.21
With supplements	0.66	0.42	0.65	0.74	(0.68, 0.79)	0.63						
Alpha carotene (mcg)	0.51	0.34	0.50	0.69	(0.58, 0.78)	0.35	0.33	0.74	0.32	0.61	(0.44, 0.75)	0.11
Beta carotene (mcg)	0.53	0.47	0.54	0.68	(0.60, 0.75)	0.46	0.44	0.61	0.45	0.67	(0.55, 0.76)	0.21
With supplements	0.52	0.38	0.50	0.63	(0.54, 0.69)	0.46						

Supplementary Table 1.2 (continued): Pearson correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR						SFFQ2 vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Lutein-zeaxanthin (mcg)	0.55	0.53	0.57	0.70	(0.62, 0.76)	0.51	0.46	0.45	0.49	0.72	(0.61, 0.81)	0.22
Lycopene (mcg)	0.37	0.42	0.35	0.71	(0.42, 0.87)	0.14	0.12	0.23	0.08	0.18	(0.01, 0.35)	0.09
Beta cryptoxanthin (mcg)	0.36	0.22	0.39	0.57	(0.45, 0.67)	0.30	0.33	0.37	0.36	0.49	(0.33, 0.62)	0.14
Vitamin B1 (mg)	0.37	0.31	0.49	0.59	(0.51, 0.66)	0.52	0.33	0.32	0.40	0.62	(0.49, 0.73)	0.18
With supplements	0.75	0.36	0.74	0.80	(0.75, 0.83)	0.77						
Vitamin B2 (mg)	0.53	0.48	0.60	0.67	(0.61, 0.72)	0.69	0.47	0.48	0.53	0.70	(0.61, 0.77)	0.31
With supplements	0.72	0.48	0.71	0.78	(0.73, 0.82)	0.73						
Vitamin B3 (mg)	0.38	0.45	0.49	0.58	(0.50, 0.64)	0.57	0.31	0.35	0.39	0.60	(0.47, 0.71)	0.18
With supplements	0.75	0.75	0.76	0.83	(0.80, 0.86)	0.73						
Vitamin B6 (mg)	0.39	0.41	0.48	0.57	(0.50, 0.64)	0.56	0.35	0.36	0.40	0.57	(0.45, 0.67)	0.21
With supplements	0.67	0.54	0.65	0.71	(0.66, 0.75)	0.71						
Vitamin B12 (mg)	0.44	0.37	0.46	0.57	(0.49, 0.65)	0.48	0.30	0.23	0.30	0.63	(0.41, 0.78)	0.09
With supplements	0.68	0.53	0.66	0.72	(0.67, 0.77)	0.73						
Natural folate (mcg)	0.50	0.57	0.60	0.68	(0.62, 0.73)	0.67	0.42	0.49	0.51	0.66	(0.57, 0.73)	0.34
Folic acid (mcg)	0.69	0.51	0.72	0.84	(0.79, 0.89)	0.57	0.41	0.39	0.39	0.63	(0.49, 0.74)	0.16
With supplements	0.70	0.54	0.69	0.89	(0.80, 0.95)	0.42						
Dietary folate eqvts (mcg)	0.47	0.50	0.56	0.64	(0.57, 0.70)	0.60	0.37	0.37	0.41	0.56	(0.44, 0.66)	0.22
With supplements	0.73	0.55	0.73	0.79	(0.74, 0.83)	0.75						
Vitamin C (mg)	0.51	0.49	0.54	0.63	(0.56, 0.69)	0.58	0.47	0.43	0.47	0.66	(0.56, 0.74)	0.26
With supplements	0.77	0.71	0.76	0.82	(0.79, 0.86)	0.77						
Vitamin D (mg)	0.61	0.47	0.60	0.70	(0.63, 0.75)	0.60	0.48	0.41	0.49	0.78	(0.65, 0.87)	0.18
With supplements	0.70	0.40	0.67	0.74	(0.68, 0.78)	0.72						

Supplementary Table 1.2 (continued): Pearson correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR						SFFQ2 vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Vitamin E (mg)	0.42	0.47	0.50	0.59	(0.51, 0.65)	0.56	0.31	0.31	0.35	0.50	(0.39, 0.60)	0.24
With supplements	0.76	0.63	0.75	0.81	(0.77, 0.84)	0.79						
Vitamin K (mg)	0.55	0.55	0.59	0.72	(0.65, 0.78)	0.50	0.39	0.38	0.43	0.66	(0.53, 0.76)	0.19
With supplements	0.51	0.10	0.52	0.60	(0.53, 0.66)	0.60						
Calcium (mg)	0.57	0.61	0.64	0.69	(0.64, 0.74)	0.72	0.49	0.50	0.55	0.69	(0.58, 0.78)	0.26
With supplements	0.68	0.39	0.68	0.75	(0.70, 0.80)	0.71						
Magnesium (mg)	0.45	0.65	0.67	0.73	(0.68, 0.78)	0.75	0.38	0.60	0.60	0.74	(0.67, 0.80)	0.38
With supplements	0.60	0.70	0.73	0.74	(0.69, 0.79)	0.81						
Iron (mg)	0.37	0.49	0.50	0.59	(0.51, 0.65)	0.59	0.32	0.41	0.40	0.57	(0.44, 0.67)	0.19
With supplements	0.44	0.47	0.50	0.61	(0.53, 0.67)	0.51						
Copper (mg)	0.37	0.38	0.50	0.61	(0.53, 0.68)	0.51	0.29	0.35	0.39	0.54	(0.43, 0.64)	0.23
With supplements	0.53	0.44	0.54	0.62	(0.55, 0.67)	0.63						
Zinc (mg)	0.38	0.41	0.44	0.54	(0.46, 0.61)	0.53	0.32	0.23	0.30	0.56	(0.37, 0.71)	0.10
With supplements	0.65	0.49	0.64	0.72	(0.66, 0.77)	0.68						
Phosphorus (mg)	0.42	0.59	0.61	0.68	(0.62, 0.73)	0.70	0.37	0.52	0.52	0.69	(0.57, 0.77)	0.25
With supplements	0.43	0.58	0.60	0.66	(0.60, 0.71)	0.69						
Choline (mg)	0.45	0.53	0.56	0.63	(0.57, 0.69)	0.65	0.36	0.39	0.41	0.63	(0.51, 0.73)	0.19
With supplements	0.44	0.46	0.51	0.59	(0.52, 0.65)	0.61						
Potassium (mg)	0.47	0.58	0.61	0.67	(0.61, 0.72)	0.75	0.43	0.52	0.55	0.67	(0.59, 0.74)	0.36
With supplements	0.47	0.58	0.59	0.65	(0.59, 0.70)	0.73						
Sodium (mg)	0.30	0.47	0.46	0.55	(0.47, 0.62)	0.53	0.27	0.31	0.28	0.41	(0.29, 0.52)	0.21
Caffeine (mg)	0.73	0.70	0.73	0.78	(0.74, 0.82)	0.80	0.70	0.65	0.70	0.81	(0.75, 0.86)	0.42

Supplementary Table 1.3: Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ1 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ1 vs. 7DDR						SFFQ1 vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Total energy (kcal)	0.28			0.31	(0.23, 0.38)	0.64	0.31			0.42	(0.33, 0.51)	0.29
Total fat (g)	0.37	0.57	0.58	0.65	(0.59, 0.70)	0.65	0.34	0.52	0.53	0.76	(0.64, 0.84)	0.23
Saturated fat (g)	0.43	0.60	0.60	0.67	(0.61, 0.72)	0.65	0.42	0.52	0.51	0.71	(0.61, 0.79)	0.25
Polyunsaturated fat (g)	0.35	0.47	0.48	0.59	(0.50, 0.65)	0.49	0.29	0.39	0.39	0.68	(0.51, 0.81)	0.12
Monounsaturated fat (g)	0.38	0.49	0.49	0.57	(0.50, 0.63)	0.58	0.31	0.40	0.42	0.62	(0.51, 0.71)	0.21
Arachadonic FA (g)	0.38	0.46	0.45	0.54	(0.46, 0.61)	0.51	0.27	0.28	0.29	0.41	(0.28, 0.53)	0.16
Lauric FA (g)	0.35	0.37	0.37	0.48	(0.38, 0.56)	0.43	0.39	0.38	0.37	0.56	(0.44, 0.65)	0.21
Linoleic FA (g)	0.35	0.47	0.48	0.58	(0.50, 0.65)	0.49	0.30	0.39	0.39	0.66	(0.48, 0.79)	0.13
Linolenic FA (g)	0.39	0.44	0.44	0.56	(0.47, 0.63)	0.44	0.27	0.30	0.31	0.55	(0.40, 0.67)	0.13
N-3 (DHA+EPA)	0.49	0.50	0.48	0.62	(0.53, 0.70)	0.41	0.28	0.28	0.28	0.52	(0.29, 0.69)	0.08
With supplements	0.59	0.59	0.58	0.65	(0.58, 0.70)	0.63						
Oleic FA (g)	0.38	0.49	0.49	0.57	(0.49, 0.63)	0.58	0.31	0.40	0.42	0.63	(0.51, 0.72)	0.20
Cholesterol (mg)	0.53	0.57	0.58	0.68	(0.61, 0.73)	0.58	0.37	0.34	0.34	0.55	(0.41, 0.66)	0.16
Protein (g)	0.36	0.48	0.48	0.55	(0.47, 0.61)	0.61	0.35	0.38	0.38	0.47	(0.36, 0.58)	0.21
Carbohydrates (g)	0.40	0.66	0.65	0.69	(0.64, 0.73)	0.74	0.41	0.52	0.52	0.66	(0.58, 0.73)	0.34
Total sugar (g)	0.53	0.69	0.69	0.75	(0.70, 0.79)	0.75	0.51	0.59	0.59	0.71	(0.63, 0.77)	0.37
Fiber (g)	0.50	0.65	0.63	0.66	(0.61, 0.71)	0.75	0.45	0.53	0.53	0.64	(0.55, 0.71)	0.33
Alcohol (g)	0.84	0.86	0.77	0.84	(0.80, 0.87)	0.73	0.75	0.74	0.66	0.77	(0.70, 0.82)	0.46
Retinol activity eqvts (mcg)	0.43	0.47	0.48	0.65	(0.55, 0.72)	0.37	0.37	0.39	0.39	0.56	(0.44, 0.66)	0.20
With supplements	0.60	0.62	0.62	0.70	(0.64, 0.75)	0.65						
Alpha carotene (mcg)	0.43	0.41	0.40	0.54	(0.44, 0.62)	0.37	0.32	0.31	0.32	0.52	(0.38, 0.64)	0.14
Beta carotene (mcg)	0.49	0.50	0.49	0.63	(0.54, 0.70)	0.44	0.40	0.41	0.42	0.63	(0.51, 0.72)	0.20
With supplements	0.48	0.48	0.48	0.60	(0.51, 0.67)	0.46						
Lutein-zeaxanthin (mcg)	0.49	0.50	0.50	0.61	(0.53, 0.68)	0.50	0.39	0.41	0.41	0.60	(0.49, 0.69)	0.21

Supplementary Table 1.3 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ1 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ1 vs. 7DDR						SFFQ1 vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Lycopene (mcg)	0.40	0.37	0.37	0.59	(0.44, 0.71)	0.23	0.18	0.15	0.11	0.18	(0.04, 0.33)	0.11
Beta cryptoxanthin (mcg)	0.38	0.39	0.38	0.56	(0.44, 0.65)	0.31	0.40	0.40	0.41	0.59	(0.44, 0.70)	0.15
Vitamin B1 (mg)	0.35	0.47	0.47	0.56	(0.48, 0.63)	0.52	0.29	0.34	0.34	0.50	(0.37, 0.60)	0.19
With supplements	0.66	0.69	0.67	0.73	(0.67, 0.78)	0.71						
Vitamin B2 (mg)	0.52	0.57	0.60	0.67	(0.61, 0.72)	0.69	0.47	0.49	0.52	0.60	(0.50, 0.68)	0.31
With supplements	0.68	0.68	0.70	0.74	(0.68, 0.78)	0.75						
Vitamin B3 (mg)	0.34	0.46	0.45	0.54	(0.46, 0.60)	0.54	0.32	0.38	0.38	0.47	(0.34, 0.59)	0.17
With supplements	0.67	0.70	0.69	0.76	(0.71, 0.80)	0.72						
Vitamin B6 (mg)	0.36	0.45	0.44	0.51	(0.43, 0.58)	0.58	0.31	0.32	0.33	0.49	(0.37, 0.60)	0.19
With supplements	0.65	0.65	0.65	0.72	(0.66, 0.77)	0.72						
Vitamin B12 (mg)	0.41	0.41	0.42	0.52	(0.43, 0.59)	0.48	0.34	0.32	0.33	0.45	(0.29, 0.59)	0.11
With supplements	0.68	0.67	0.66	0.73	(0.67, 0.78)	0.72						
Natural folate (mcg)	0.49	0.53	0.54	0.61	(0.54, 0.67)	0.65	0.39	0.46	0.45	0.59	(0.50, 0.66)	0.33
Folic acid (mcg)	0.49	0.54	0.53	0.61	(0.54, 0.67)	0.59	0.35	0.37	0.36	0.50	(0.38, 0.59)	0.23
With supplements	0.61	0.62	0.62	0.68	(0.63, 0.73)	0.72						
Dietary folate eqvts (mcg)	0.40	0.49	0.48	0.57	(0.49, 0.63)	0.56	0.30	0.36	0.35	0.52	(0.40, 0.61)	0.20
With supplements	0.62	0.63	0.63	0.69	(0.64, 0.73)	0.73						
Vitamin C (mg)	0.48	0.49	0.49	0.57	(0.5, 0.64)	0.57	0.43	0.43	0.43	0.59	(0.49, 0.67)	0.28
With supplements	0.67	0.67	0.66	0.72	(0.67, 0.77)	0.73						
Vitamin D (mg)	0.57	0.54	0.55	0.64	(0.57, 0.69)	0.58	0.53	0.50	0.50	0.63	(0.50, 0.73)	0.20
With supplements	0.58	0.52	0.56	0.62	(0.56, 0.67)	0.72						
Vitamin E (mg)	0.43	0.47	0.47	0.57	(0.49, 0.63)	0.53	0.29	0.32	0.32	0.45	(0.35, 0.55)	0.23
With supplements	0.72	0.72	0.72	0.75	(0.71, 0.80)	0.77						

Supplementary Table 1.3 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ1 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ1 vs. 7DDR						SFFQ1 vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual	De-attenuated		ICC		Energy density	Residual	De-attenuated		ICC
Vitamin K (mg)	0.51	0.53	0.53	0.65	(0.57, 0.72)	0.48	0.35	0.38	0.38	0.61	(0.48, 0.70)	0.17
With supplements	0.50	0.51	0.51	0.61	(0.54, 0.68)	0.53						
Calcium (mg)	0.54	0.59	0.61	0.67	(0.61, 0.72)	0.70	0.50	0.56	0.57	0.64	(0.53, 0.73)	0.26
With supplements	0.64	0.62	0.66	0.72	(0.67, 0.77)	0.73						
Magnesium (mg)	0.45	0.60	0.60	0.66	(0.60, 0.71)	0.73	0.43	0.55	0.55	0.70	(0.62, 0.77)	0.37
With supplements	0.57	0.67	0.68	0.73	(0.68, 0.78)	0.80						
Iron (mg)	0.37	0.47	0.47	0.55	(0.48, 0.62)	0.54	0.28	0.35	0.34	0.53	(0.41, 0.63)	0.19
With supplements	0.44	0.50	0.49	0.59	(0.51, 0.66)	0.50						
Copper (mg)	0.37	0.46	0.45	0.51	(0.43, 0.58)	0.60	0.34	0.35	0.35	0.49	(0.38, 0.58)	0.25
With supplements	0.51	0.54	0.54	0.61	(0.54, 0.66)	0.64						
Zinc (mg)	0.38	0.40	0.41	0.51	(0.42, 0.58)	0.49	0.32	0.29	0.30	0.52	(0.35, 0.66)	0.11
With supplements	0.58	0.57	0.57	0.64	(0.58, 0.70)	0.66						
Phosphorus (mg)	0.42	0.52	0.54	0.60	(0.54, 0.66)	0.68	0.42	0.49	0.50	0.61	(0.49, 0.70)	0.24
With supplements	0.43	0.53	0.56	0.62	(0.56, 0.67)	0.68						
Choline (mg)	0.47	0.51	0.53	0.61	(0.54, 0.66)	0.63	0.39	0.35	0.36	0.53	(0.40, 0.63)	0.19
With supplements	0.47	0.52	0.54	0.62	(0.55, 0.67)	0.62						
Potassium (mg)	0.47	0.51	0.54	0.57	(0.51, 0.63)	0.73	0.44	0.47	0.49	0.61	(0.52, 0.67)	0.36
With supplements	0.47	0.52	0.55	0.58	(0.52, 0.64)	0.73						
Sodium (mg)	0.31	0.42	0.41	0.49	(0.41, 0.57)	0.50	0.26	0.26	0.25	0.37	(0.25, 0.47)	0.20
Caffeine (mg)	0.78	0.76	0.77	0.81	(0.78, 0.84)	0.86	0.74	0.75	0.74	0.79	(0.73, 0.84)	0.50

Supplementary Table 1.4: Spearman correlation coefficient for comparison of daily nutrient intake from WebFFQ with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	WebFFQ vs. 7DDR						WebFFQ vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Total energy (kcal)	0.27			0.30	(0.22, 0.37)	0.64	0.30			0.39	(0.29, 0.48)	0.29
Total fat (g)	0.32	0.55	0.56	0.63	(0.56, 0.68)	0.65	0.31	0.50	0.51	0.73	(0.62, 0.82)	0.23
Saturated fat (g)	0.39	0.59	0.60	0.67	(0.61, 0.72)	0.65	0.39	0.52	0.51	0.68	(0.57, 0.77)	0.25
Polyunsaturated fat (g)	0.27	0.47	0.48	0.59	(0.51, 0.66)	0.49	0.26	0.38	0.39	0.61	(0.43, 0.74)	0.12
Monounsaturated fat (g)	0.35	0.48	0.49	0.57	(0.49, 0.63)	0.58	0.28	0.39	0.40	0.60	(0.48, 0.69)	0.21
Arachadonic FA (g)	0.39	0.46	0.45	0.55	(0.46, 0.61)	0.51	0.27	0.29	0.29	0.47	(0.35, 0.58)	0.16
Lauric FA (g)	0.33	0.36	0.36	0.46	(0.37, 0.54)	0.43	0.36	0.36	0.36	0.54	(0.43, 0.63)	0.21
Linoleic FA (g)	0.28	0.45	0.47	0.58	(0.49, 0.64)	0.49	0.25	0.37	0.38	0.61	(0.44, 0.74)	0.13
Linolenic FA (g)	0.34	0.45	0.45	0.58	(0.49, 0.65)	0.44	0.27	0.33	0.34	0.54	(0.37, 0.66)	0.13
N-3 (DHA+EPA) FA (g)	0.50	0.52	0.51	0.66	(0.57, 0.73)	0.41	0.30	0.31	0.30	0.63	(0.38, 0.80)	0.08
With supplements	0.60	0.62	0.60	0.68	(0.62, 0.73)	0.63						
Oleic FA (g)	0.35	0.48	0.48	0.56	(0.49, 0.62)	0.58	0.28	0.38	0.39	0.59	(0.47, 0.69)	0.20
Cholesterol (mg)	0.49	0.55	0.55	0.65	(0.58, 0.70)	0.58	0.38	0.38	0.38	0.62	(0.48, 0.72)	0.16
Protein (g)	0.34	0.47	0.48	0.54	(0.46, 0.60)	0.61	0.32	0.39	0.39	0.53	(0.41, 0.63)	0.21
Carbohydrates (g)	0.40	0.64	0.64	0.67	(0.62, 0.72)	0.74	0.40	0.56	0.56	0.71	(0.62, 0.78)	0.34
Total sugar (g)	0.51	0.67	0.67	0.73	(0.68, 0.78)	0.75	0.49	0.61	0.61	0.72	(0.64, 0.79)	0.37
Fiber (g)	0.46	0.64	0.62	0.65	(0.59, 0.70)	0.75	0.41	0.53	0.53	0.67	(0.59, 0.74)	0.33
Alcohol (g)	0.85	0.86	0.75	0.82	(0.78, 0.85)	0.73	0.75	0.74	0.65	0.75	(0.69, 0.81)	0.46
Retinol activity eqvts (mcg)	0.39	0.46	0.46	0.61	(0.51, 0.69)	0.37	0.32	0.37	0.37	0.56	(0.44, 0.66)	0.20
With supplements	0.63	0.67	0.66	0.74	(0.68, 0.79)	0.65						
Alpha carotene (mcg)	0.45	0.45	0.43	0.58	(0.48, 0.66)	0.37	0.34	0.34	0.35	0.59	(0.45, 0.70)	0.14
Beta carotene (mcg)	0.47	0.51	0.50	0.63	(0.55, 0.70)	0.44	0.42	0.43	0.44	0.63	(0.51, 0.72)	0.20
With supplements	0.47	0.50	0.49	0.61	(0.52, 0.68)	0.46						
Lutein-zeaxanthin (mcg)	0.50	0.52	0.51	0.63	(0.55, 0.69)	0.50	0.46	0.49	0.49	0.61	(0.48, 0.71)	0.21

Supplementary Table 1.4 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from WebFFQ with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	WebFFQ vs. 7DDR						WebFFQ vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Lycopene (mcg)	0.42	0.41	0.40	0.65	(0.49, 0.77)	0.23	0.20	0.17	0.14	0.26	(0.11, 0.39)	0.11
Beta cryptoxanthin (mcg)	0.36	0.36	0.36	0.52	(0.40, 0.61)	0.31	0.39	0.40	0.40	0.66	(0.51, 0.77)	0.15
Vitamin B1 (mg)	0.34	0.51	0.50	0.61	(0.53, 0.67)	0.52	0.30	0.37	0.36	0.55	(0.41, 0.65)	0.19
With supplements	0.66	0.68	0.67	0.74	(0.69, 0.79)	0.71						
Vitamin B2 (mg)	0.49	0.57	0.60	0.66	(0.60, 0.71)	0.69	0.47	0.55	0.57	0.70	(0.60, 0.77)	0.31
With supplements	0.65	0.68	0.68	0.74	(0.69, 0.79)	0.75						
Vitamin B3 (mg)	0.32	0.46	0.46	0.54	(0.46, 0.61)	0.54	0.25	0.32	0.31	0.49	(0.36, 0.60)	0.17
With supplements	0.65	0.69	0.69	0.76	(0.71, 0.80)	0.72						
Vitamin B6 (mg)	0.35	0.48	0.48	0.56	(0.48, 0.62)	0.58	0.33	0.40	0.40	0.60	(0.47, 0.70)	0.19
With supplements	0.64	0.65	0.65	0.71	(0.65, 0.75)	0.72						
Vitamin B12 (mg)	0.42	0.46	0.46	0.57	(0.49, 0.64)	0.48	0.31	0.30	0.30	0.53	(0.37, 0.66)	0.11
With supplements	0.66	0.65	0.65	0.72	(0.66, 0.77)	0.72						
Natural folate (mcg)	0.44	0.56	0.56	0.64	(0.57, 0.69)	0.65	0.40	0.50	0.49	0.60	(0.50, 0.67)	0.33
Folic acid (mcg)	0.51	0.57	0.57	0.66	(0.59, 0.71)	0.59	0.42	0.45	0.44	0.57	(0.45, 0.66)	0.23
With supplements	0.65	0.68	0.67	0.73	(0.68, 0.78)	0.72						
Dietary folate eqvts (mcg)	0.38	0.50	0.50	0.58	(0.50, 0.64)	0.56	0.33	0.42	0.42	0.58	(0.45, 0.67)	0.20
With supplements	0.65	0.68	0.68	0.74	(0.69, 0.79)	0.73						
Vitamin C (mg)	0.47	0.51	0.50	0.59	(0.52, 0.65)	0.57	0.49	0.49	0.49	0.65	(0.55, 0.72)	0.28
With supplements	0.68	0.69	0.69	0.76	(0.71, 0.80)	0.73						
Vitamin D (mg)	0.56	0.57	0.58	0.67	(0.60, 0.72)	0.58	0.52	0.49	0.49	0.74	(0.62, 0.83)	0.20
With supplements	0.62	0.57	0.59	0.65	(0.59, 0.70)	0.72						
Vitamin E (mg)	0.33	0.44	0.43	0.52	(0.43, 0.59)	0.53	0.26	0.34	0.33	0.47	(0.37, 0.57)	0.23
With supplements	0.70	0.72	0.71	0.77	(0.72, 0.81)	0.77						

Supplementary Table 1.4 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from WebFFQ with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	WebFFQ vs. 7DDR						WebFFQ vs. ASA24					
	Un-adjusted	Energy-adjusted					Un-adjusted	Energy-adjusted				
		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC		Energy density	Residual method	De-attenuated (ρ & 95%CI)		ICC
Vitamin K (mg)	0.50	0.55	0.54	0.67	(0.59, 0.73)	0.48	0.36	0.42	0.41	0.58	(0.44, 0.69)	0.17
With supplements	0.48	0.52	0.51	0.61	(0.54, 0.68)	0.53						
Calcium (mg)	0.51	0.60	0.61	0.66	(0.60, 0.70)	0.70	0.46	0.51	0.51	0.64	(0.53, 0.73)	0.26
With supplements	0.62	0.66	0.67	0.71	(0.66, 0.75)	0.73						
Magnesium (mg)	0.38	0.61	0.61	0.66	(0.61, 0.71)	0.73	0.34	0.52	0.51	0.65	(0.57, 0.71)	0.37
With supplements	0.53	0.70	0.70	0.75	(0.70, 0.79)	0.80						
Iron (mg)	0.37	0.55	0.54	0.64	(0.57, 0.70)	0.54	0.30	0.44	0.42	0.61	(0.48, 0.71)	0.19
With supplements	0.39	0.48	0.47	0.57	(0.49, 0.64)	0.50						
Copper (mg)	0.34	0.48	0.46	0.54	(0.46, 0.60)	0.60	0.27	0.37	0.35	0.50	(0.39, 0.59)	0.25
With supplements	0.46	0.53	0.50	0.57	(0.50, 0.63)	0.64						
Zinc (mg)	0.35	0.42	0.43	0.53	(0.44, 0.60)	0.49	0.29	0.28	0.28	0.53	(0.36, 0.66)	0.11
With supplements	0.55	0.57	0.58	0.65	(0.59, 0.70)	0.66						
Phosphorus (mg)	0.38	0.55	0.56	0.62	(0.56, 0.67)	0.68	0.37	0.50	0.51	0.59	(0.47, 0.68)	0.24
With supplements	0.38	0.55	0.56	0.62	(0.56, 0.67)	0.68						
Choline (mg)	0.44	0.52	0.54	0.61	(0.54, 0.67)	0.63	0.37	0.38	0.38	0.58	(0.45, 0.68)	0.19
With supplements	0.44	0.52	0.54	0.61	(0.55, 0.67)	0.62						
Potassium (mg)	0.43	0.51	0.53	0.59	(0.52, 0.64)	0.73	0.42	0.47	0.49	0.60	(0.52, 0.67)	0.36
With supplements	0.42	0.51	0.53	0.58	(0.52, 0.64)	0.73						
Sodium (mg)	0.27	0.44	0.42	0.51	(0.42, 0.58)	0.50	0.27	0.33	0.32	0.47	(0.36, 0.57)	0.20
Caffeine (mg)	0.77	0.76	0.76	0.80	(0.77, 0.83)	0.86	0.72	0.74	0.71	0.78	(0.72, 0.83)	0.50

Supplementary Table 1.5: Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 226 U.S. female nurses with complete four ASA24, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Total energy (kcal)	0.29		0.32	(0.19, 0.44)	0.67	0.39		0.50	(0.35, 0.63)	0.27
Total fat (g)	0.33	0.56	0.63	(0.52, 0.71)	0.65	0.39	0.56	0.75	(0.58, 0.86)	0.23
Saturated fat (g)	0.44	0.61	0.68	(0.58, 0.77)	0.65	0.45	0.51	0.68	(0.52, 0.79)	0.25
Polyunsaturated fat (g)	0.23	0.47	0.57	(0.43, 0.68)	0.48	0.32	0.45	0.70	(0.45, 0.85)	0.14
Monounsaturated fat (g)	0.36	0.46	0.55	(0.41, 0.66)	0.55	0.34	0.40	0.55	(0.37, 0.68)	0.21
Arachadonic FA (g)	0.28	0.38	0.46	(0.32, 0.59)	0.49	0.25	0.31	0.45	(0.26, 0.60)	0.18
Lauric FA (g)	0.40	0.42	0.53	(0.38, 0.65)	0.44	0.40	0.38	0.55	(0.36, 0.70)	0.18
Linoleic FA (g)	0.24	0.46	0.57	(0.42, 0.68)	0.48	0.32	0.43	0.70	(0.43, 0.86)	0.13
Linolenic FA (g)	0.33	0.47	0.62	(0.46, 0.74)	0.40	0.33	0.39	0.58	(0.37, 0.72)	0.17
N-3 (DHA+EPA) FA (g)	0.50	0.52	0.67	(0.51, 0.79)	0.41	0.29	0.31	0.62	(0.22, 0.84)	0.08
With supplements	0.58	0.56	0.64	(0.53, 0.73)	0.59					
Oleic FA (g)	0.36	0.46	0.55	(0.41, 0.65)	0.54	0.34	0.38	0.54	(0.36, 0.68)	0.20
Cholesterol (mg)	0.46	0.55	0.63	(0.51, 0.72)	0.58	0.43	0.45	0.64	(0.44, 0.78)	0.19
Protein (g)	0.27	0.47	0.53	(0.40, 0.63)	0.64	0.33	0.40	0.54	(0.37, 0.67)	0.22
Carbohydrate (g)	0.47	0.63	0.68	(0.59, 0.75)	0.73	0.51	0.60	0.73	(0.60, 0.82)	0.34
Total sugar (g)	0.57	0.70	0.75	(0.67, 0.82)	0.73	0.59	0.66	0.80	(0.68, 0.87)	0.37
Fiber (g)	0.51	0.65	0.69	(0.60, 0.75)	0.79	0.46	0.54	0.65	(0.52, 0.74)	0.36
Alcohol (g)	0.85	0.77	0.83	(0.77, 0.88)	0.75	0.75	0.67	0.77	(0.66, 0.84)	0.43
Retinol activity eqvts (mcg)	0.51	0.54	0.73	(0.55, 0.85)	0.37	0.37	0.38	0.65	(0.37, 0.83)	0.11
With supplements	0.67	0.69	0.81	(0.70, 0.88)	0.58					
Alpha carotene (mcg)	0.56	0.55	0.70	(0.55, 0.81)	0.44	0.44	0.41	0.67	(0.40, 0.83)	0.13
Beta carotene (mcg)	0.58	0.59	0.73	(0.59, 0.83)	0.46	0.47	0.46	0.68	(0.46, 0.83)	0.16
With supplements	0.55	0.55	0.70	(0.55, 0.81)	0.44					
Lutein-zeaxanthin (mcg)	0.54	0.57	0.70	(0.57, 0.80)	0.50	0.48	0.50	0.70	(0.51, 0.83)	0.19
Lycopene (mcg)	0.47	0.45	0.68	(0.44, 0.82)	0.29	0.31	0.24	0.42	(0.17, 0.61)	0.11
Beta cryptoxanthin (mcg)	0.32	0.32	0.48	(0.28, 0.65)	0.28	0.43	0.38	0.53	(0.35, 0.67)	0.20
Vitamin B1 (mg)	0.39	0.52	0.61	(0.48, 0.71)	0.55	0.39	0.44	0.62	(0.43, 0.77)	0.19
With supplements	0.70	0.71	0.78	(0.69, 0.84)	0.72					

Supplementary Table 1.5 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted, energy-adjusted and de-attenuated results (data provided by 226 U.S. female nurses with complete four ASA24, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Vitamin B2 (mg)	0.52	0.59	0.64	(0.53, 0.72)	0.72	0.51	0.56	0.71	(0.57, 0.81)	0.30
With supplements	0.70	0.73	0.79	(0.71, 0.85)	0.75					
Vitamin B3 (mg)	0.29	0.41	0.48	(0.34, 0.59)	0.55	0.34	0.37	0.56	(0.36, 0.71)	0.16
With supplements	0.66	0.70	0.77	(0.68, 0.83)	0.71					
Vitamin B6 (mg)	0.32	0.47	0.55	(0.42, 0.66)	0.55	0.37	0.36	0.54	(0.34, 0.69)	0.17
With supplements	0.65	0.66	0.71	(0.62, 0.79)	0.73					
Vitamin B12 (mg)	0.37	0.44	0.54	(0.39, 0.66)	0.46	0.26	0.29	0.54	(0.23, 0.75)	0.09
With supplements	0.68	0.69	0.75	(0.66, 0.81)	0.73					
Natural folate (mcg)	0.53	0.64	0.72	(0.62, 0.80)	0.66	0.48	0.53	0.65	(0.52, 0.75)	0.33
Folic acid (mcg)	0.56	0.61	0.71	(0.60, 0.80)	0.59	0.51	0.51	0.70	(0.52, 0.82)	0.22
With supplements	0.67	0.69	0.73	(0.65, 0.80)	0.79					
Dietary folate eqvts (mcg)	0.44	0.54	0.62	(0.50, 0.71)	0.59	0.42	0.48	0.71	(0.50, 0.84)	0.18
With supplements	0.64	0.68	0.73	(0.64, 0.80)	0.79					
Vitamin C (mg)	0.55	0.55	0.63	(0.51, 0.72)	0.61	0.54	0.50	0.65	(0.49, 0.77)	0.26
With supplements	0.72	0.71	0.76	(0.68, 0.83)	0.75					
Vitamin D (mg)	0.57	0.60	0.72	(0.59, 0.81)	0.54	0.51	0.52	0.73	(0.53, 0.85)	0.20
With supplements	0.64	0.61	0.66	(0.56, 0.73)	0.75					
Vitamin E (mg)	0.38	0.42	0.52	(0.37, 0.65)	0.44	0.34	0.36	0.52	(0.33, 0.66)	0.18
With supplements	0.73	0.72	0.76	(0.69, 0.82)	0.80					
Vitamin K (mg)	0.54	0.60	0.73	(0.60, 0.82)	0.51	0.43	0.45	0.70	(0.46, 0.85)	0.15
With supplements	0.49	0.55	0.65	(0.52, 0.75)	0.53					
Calcium (mg)	0.57	0.63	0.69	(0.59, 0.77)	0.72	0.49	0.50	0.69	(0.50, 0.81)	0.21
With supplements	0.66	0.66	0.72	(0.62, 0.79)	0.71					
Magnesium (mg)	0.44	0.68	0.72	(0.64, 0.80)	0.77	0.40	0.60	0.72	(0.60, 0.81)	0.37
With supplements	0.58	0.70	0.74	(0.66, 0.81)	0.81					
Iron (mg)	0.32	0.49	0.59	(0.45, 0.69)	0.52	0.34	0.37	0.55	(0.36, 0.70)	0.16
With supplements	0.37	0.48	0.55	(0.42, 0.65)	0.62					

Supplementary Table 1.5 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 226 U.S. female nurses with complete four ASA24, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR				SFFQ2 vs. ASA24			
	Un-adjusted	Energy-adjusted			Un-adjusted	Energy-adjusted		
		Residual method	De-attenuated (ρ & 95%CI)	ICC		Residual method	De-attenuated (ρ & 95%CI)	ICC
Copper (mg)	0.42	0.53	0.64 (0.51, 0.73)	0.53	0.38	0.41	0.54 (0.37, 0.66)	0.25
With supplements	0.56	0.62	0.71 (0.60, 0.79)	0.63				
Zinc (mg)	0.30	0.43	0.52 (0.37, 0.63)	0.51	0.24	0.28	0.52 (0.22, 0.73)	0.09
With supplements	0.65	0.67	0.73 (0.64, 0.80)	0.72				
Phosphorus (mg)	0.37	0.62	0.69 (0.59, 0.77)	0.69	0.39	0.56	0.74 (0.58, 0.85)	0.24
With supplements	0.38	0.60	0.66 (0.56, 0.74)	0.71				
Choline (mg)	0.40	0.60	0.67 (0.57, 0.75)	0.64	0.42	0.45	0.62 (0.44, 0.74)	0.22
With supplements	0.40	0.59	0.67 (0.56, 0.77)	0.62				
Potassium (mg)	0.45	0.64	0.69 (0.60, 0.77)	0.77	0.48	0.57	0.69 (0.56, 0.78)	0.37
With supplements	0.46	0.65	0.69 (0.60, 0.77)	0.78				
Sodium (mg)	0.27	0.46	0.54 (0.41, 0.65)	0.56	0.29	0.32	0.45 (0.27, 0.59)	0.20
Caffeine (mg)	0.76	0.75	0.77 (0.70, 0.83)	0.88	0.71	0.71	0.79 (0.70, 0.85)	0.51

Supplementary Table 1.6: Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted, adjusted and de-attenuated results (data provided by 309 U.S. female nurses aged 45-60 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR				SFFQ2 vs. ASA24			
	Un-adjusted	Energy-adjusted			Un-adjusted	Energy-adjusted		
		Residual method	De-attenuated (ρ & 95%CI)	ICC		Residual method	De-attenuated (ρ & 95%CI)	ICC
Total energy (kcal)	0.31		0.36 (0.24, 0.46)	0.61	0.32		0.40 (0.27, 0.52)	0.32
Total fat (g)	0.36	0.59	0.66 (0.57, 0.73)	0.66	0.33	0.54	0.75 (0.55, 0.87)	0.20
Saturated fat (g)	0.47	0.59	0.66 (0.57, 0.73)	0.65	0.44	0.54	0.76 (0.60, 0.86)	0.23
Polyunsaturated fat (g)	0.24	0.45	0.54 (0.42, 0.64)	0.50	0.20	0.35	0.54 (0.32, 0.71)	0.12
Monounsaturated fat (g)	0.36	0.45	0.52 (0.40, 0.61)	0.61	0.29	0.38	0.58 (0.41, 0.70)	0.20
Arachadonic FA (g)	0.40	0.47	0.56 (0.44, 0.65)	0.53	0.21	0.32	0.44 (0.26, 0.60)	0.18
Lauric FA (g)	0.38	0.38	0.48 (0.35, 0.59)	0.44	0.29	0.30	0.44 (0.27, 0.58)	0.21
Linoleic FA (g)	0.24	0.41	0.50 (0.37, 0.60)	0.50	0.20	0.33	0.52 (0.31, 0.68)	0.13
Linolenic FA (g)	0.36	0.49	0.62 (0.49, 0.72)	0.44	0.25	0.27	0.48 (0.23, 0.67)	0.11
N-3 (DHA+EPA) FA (g)	0.54	0.52	0.68 (0.54, 0.79)	0.39	0.30	0.33	0.61 (0.26, 0.82)	0.09
With supplements	0.62	0.60	0.69 (0.59, 0.77)	0.59				
Oleic FA (g)	0.36	0.45	0.51 (0.40, 0.61)	0.60	0.30	0.38	0.58 (0.41, 0.71)	0.19
Cholesterol (mg)	0.50	0.54	0.62 (0.52, 0.70)	0.60	0.36	0.39	0.61 (0.42, 0.74)	0.18
Protein (g)	0.37	0.53	0.61 (0.50, 0.69)	0.60	0.29	0.39	0.46 (0.29, 0.60)	0.22
Carbohydrate (g)	0.44	0.66	0.71 (0.63, 0.77)	0.74	0.44	0.56	0.68 (0.55, 0.77)	0.34
Total sugar (g)	0.52	0.65	0.70 (0.62, 0.77)	0.73	0.53	0.61	0.72 (0.61, 0.81)	0.36
Fiber (g)	0.54	0.64	0.70 (0.62, 0.77)	0.72	0.44	0.48	0.59 (0.46, 0.70)	0.31
Alcohol (g)	0.85	0.75	0.83 (0.77, 0.88)	0.69	0.73	0.61	0.74 (0.63, 0.82)	0.38
Retinol activity eqvts (mcg)	0.52	0.53	0.75 (0.57, 0.87)	0.33	0.41	0.43	0.62 (0.43, 0.75)	0.18
With supplements	0.60	0.61	0.70 (0.61, 0.78)	0.59				
Alpha carotene (mcg)	0.56	0.50	0.67 (0.52, 0.78)	0.37	0.39	0.33	0.54 (0.26, 0.74)	0.11
Beta carotene (mcg)	0.59	0.56	0.72 (0.59, 0.82)	0.43	0.49	0.45	0.72 (0.52, 0.85)	0.17
With supplements	0.58	0.56	0.71 (0.58, 0.80)	0.44				
Lutein-zeaxanthin (mcg)	0.60	0.60	0.73 (0.62, 0.82)	0.50	0.50	0.52	0.66 (0.45, 0.81)	0.19
Lycopene (mcg)	0.37	0.37	0.70 (0.32, 0.89)	0.15	0.28	0.19	0.49 (0.18, 0.71)	0.08
Beta cryptoxanthin (mcg)	0.28	0.28	0.41 (0.24, 0.55)	0.30	0.37	0.36	0.62 (0.28, 0.83)	0.09
Vitamin B1 (mg)	0.41	0.53	0.64 (0.53, 0.73)	0.51	0.37	0.46	0.69 (0.52, 0.82)	0.19
With supplements	0.65	0.66	0.72 (0.64, 0.79)	0.70				

Supplementary Table 1.6 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted, adjusted and de-attenuated results (data provided by 309 U.S. female nurses aged 45-60 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Vitamin B2 (mg)	0.54	0.61	0.67	(0.58, 0.73)	0.70	0.50	0.67	0.65	(0.51, 0.75)	0.33
With supplements	0.67	0.69	0.75	(0.68, 0.81)	0.75					
Vitamin B3 (mg)	0.40	0.46	0.54	(0.43, 0.63)	0.56	0.32	0.41	0.63	(0.37, 0.8)	0.14
With supplements	0.62	0.66	0.72	(0.64, 0.79)	0.69					
Vitamin B6 (mg)	0.41	0.47	0.55	(0.44, 0.64)	0.57	0.30	0.32	0.47	(0.30, 0.61)	0.19
With supplements	0.60	0.60	0.65	(0.57, 0.72)	0.69					
Vitamin B12 (mg)	0.43	0.44	0.55	(0.42, 0.65)	0.45	0.35	0.34	0.43	(0.14, 0.65)	0.11
With supplements	0.64	0.64	0.70	(0.62, 0.77)	0.68					
Natural folate (mcg)	0.54	0.63	0.72	(0.63, 0.79)	0.61	0.48	0.53	0.67	(0.55, 0.77)	0.32
Folic acid (mcg)	0.52	0.54	0.63	(0.52, 0.71)	0.60	0.44	0.43	0.54	(0.36, 0.68)	0.21
With supplements	0.64	0.65	0.72	(0.63, 0.79)	0.68					
Dietary folate eqvts (mcg)	0.45	0.50	0.58	(0.47, 0.67)	0.57	0.40	0.41	0.56	(0.38, 0.70)	0.20
With supplements	0.64	0.66	0.72	(0.64, 0.79)	0.69					
Vitamin C (mg)	0.47	0.51	0.60	(0.49, 0.69)	0.53	0.54	0.48	0.66	(0.50, 0.78)	0.23
With supplements	0.65	0.66	0.73	(0.64, 0.79)	0.68					
Vitamin D (mg)	0.65	0.65	0.75	(0.66, 0.82)	0.58	0.53	0.54	0.69	(0.47, 0.83)	0.20
With supplements	0.63	0.61	0.65	(0.57, 0.72)	0.77					
Vitamin E (mg)	0.37	0.46	0.55	(0.43, 0.64)	0.52	0.30	0.36	0.49	(0.34, 0.61)	0.25
With supplements	0.71	0.71	0.77	(0.70, 0.83)	0.72					
Vitamin K (mg)	0.57	0.60	0.76	(0.64, 0.85)	0.45	0.44	0.44	0.64	(0.43, 0.78)	0.18
With supplements	0.59	0.61	0.76	(0.64, 0.84)	0.48					
Calcium (mg)	0.60	0.65	0.71	(0.63, 0.78)	0.70	0.51	0.56	0.66	(0.50, 0.78)	0.27
With supplements	0.65	0.66	0.72	(0.64, 0.79)	0.71					
Magnesium (mg)	0.48	0.68	0.74	(0.66, 0.80)	0.73	0.38	0.57	0.71	(0.59, 0.80)	0.37
With supplements	0.56	0.69	0.74	(0.67, 0.80)	0.75					
Iron (mg)	0.40	0.44	0.52	(0.40, 0.62)	0.53	0.32	0.37	0.61	(0.41, 0.75)	0.16
With supplements	0.50	0.53	0.62	(0.51, 0.70)	0.58					

Supplementary Table 1.6 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted, adjusted and de-attenuated results (data provided by 309 U.S. female nurses aged 45-60 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Copper (mg)	0.36	0.47	0.55	(0.43, 0.64)	0.56	0.28	0.34	0.48	(0.33, 0.61)	0.24
With supplements	0.46	0.52	0.58	(0.48, 0.67)	0.62					
Zinc (mg)	0.43	0.46	0.58	(0.45, 0.68)	0.45	0.34	0.28	0.46	(0.27, 0.62)	0.15
With supplements	0.54	0.53	0.59	(0.49, 0.67)	0.66					
Phosphorus (mg)	0.45	0.61	0.67	(0.59, 0.74)	0.67	0.35	0.50	0.61	(0.43, 0.74)	0.23
With supplements	0.45	0.62	0.69	(0.60, 0.75)	0.68					
Choline (mg)	0.49	0.58	0.66	(0.57, 0.73)	0.63	0.35	0.41	0.53	(0.36, 0.66)	0.22
With supplements	0.48	0.59	0.67	(0.58, 0.75)	0.61					
Potassium (mg)	0.51	0.65	0.71	(0.63, 0.78)	0.72	0.46	0.55	0.60	(0.48, 0.70)	0.37
With supplements	0.51	0.65	0.71	(0.63, 0.78)	0.71					
Sodium (mg)	0.28	0.41	0.49	(0.37, 0.59)	0.51	0.24	0.20	0.31	(0.13, 0.46)	0.18
Caffeine (mg)	0.81	0.78	0.81	(0.77, 0.85)	0.87	0.75	0.75	0.80	(0.71, 0.86)	0.49

Supplementary Table 1.7: Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 323 U.S. female nurses aged 61-80 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Total energy (kcal)	0.24		0.27	(0.16, 0.37)	0.65	0.30		0.40	(0.26, 0.53)	0.27
Total fat (g)	0.35	0.59	0.67	(0.57, 0.73)	0.63	0.36	0.62	0.69	(0.49, 0.82)	0.25
Saturated fat (g)	0.41	0.62	0.70	(0.61, 0.77)	0.64	0.43	0.61	0.59	(0.42, 0.72)	0.26
Polyunsaturated fat (g)	0.33	0.49	0.61	(0.49, 0.70)	0.47	0.30	0.52	0.68	(0.14, 0.90)	0.12
Monounsaturated fat (g)	0.37	0.52	0.61	(0.50, 0.69)	0.56	0.31	0.43	0.62	(0.44, 0.74)	0.21
Arachadonic FA (g)	0.32	0.40	0.48	(0.37, 0.58)	0.50	0.30	0.34	0.58	(0.37, 0.73)	0.14
Lauric FA (g)	0.31	0.34	0.43	(0.30, 0.55)	0.41	0.39	0.36	0.51	(0.35, 0.64)	0.22
Linoleic FA (g)	0.35	0.50	0.61	(0.49, 0.70)	0.48	0.32	0.53	0.64	(0.06, 0.90)	0.12
Linolenic FA (g)	0.34	0.41	0.52	(0.39, 0.62)	0.44	0.29	0.42	0.56	(0.34, 0.73)	0.15
N-3 (DHA+EPA) FA (g)	0.52	0.51	0.66	(0.53, 0.75)	0.42	0.25	0.27	0.58	(0.18, 0.82)	0.07
With supplements	0.62	0.60	0.67	(0.58, 0.74)	0.65					
Oleic FA (g)	0.37	0.51	0.60	(0.49, 0.68)	0.55	0.31	0.42	0.61	(0.43, 0.74)	0.20
Cholesterol (mg)	0.49	0.59	0.69	(0.60, 0.78)	0.56	0.44	0.47	0.70	(0.43, 0.86)	0.14
Protein (g)	0.27	0.43	0.49	(0.38, 0.58)	0.63	0.31	0.37	0.57	(0.39, 0.70)	0.20
Carbohydrate (g)	0.38	0.66	0.71	(0.63, 0.77)	0.74	0.45	0.66	0.74	(0.61, 0.83)	0.34
Total sugar (g)	0.54	0.71	0.76	(0.69, 0.82)	0.78	0.56	0.67	0.76	(0.65, 0.84)	0.39
Fiber (g)	0.38	0.59	0.63	(0.55, 0.70)	0.78	0.39	0.56	0.68	(0.56, 0.78)	0.35
Alcohol (g)	0.86	0.77	0.83	(0.78, 0.88)	0.75	0.78	0.70	0.75	(0.67, 0.82)	0.53
Retinol activity eqvts (mcg)	0.38	0.45	0.58	(0.45, 0.68)	0.41	0.38	0.40	0.50	(0.32, 0.65)	0.20
With supplements	0.65	0.67	0.74	(0.66, 0.81)	0.67					
Alpha carotene (mcg)	0.43	0.45	0.61	(0.46, 0.71)	0.37	0.40	0.42	0.61	(0.40, 0.75)	0.18
Beta carotene (mcg)	0.42	0.47	0.59	(0.46, 0.69)	0.45	0.39	0.43	0.60	(0.43, 0.72)	0.23
With supplements	0.40	0.43	0.53	(0.40, 0.63)	0.47					
Lutein-zeaxanthin (mcg)	0.47	0.50	0.61	(0.50, 0.70)	0.50	0.40	0.45	0.65	(0.49, 0.77)	0.23
Lycopene (mcg)	0.42	0.38	0.55	(0.38, 0.68)	0.30	0.18	0.13	0.21	(0.03, 0.38)	0.16
Beta cryptoxanthin (mcg)	0.41	0.43	0.64	(0.46, 0.77)	0.30	0.42	0.45	0.56	(0.38, 0.70)	0.22
Vitamin B1 (mg)	0.31	0.54	0.65	(0.54, 0.73)	0.52	0.28	0.39	0.57	(0.37, 0.71)	0.18
With supplements	0.74	0.75	0.82	(0.75, 0.87)	0.71					

Supplementary Table 1.7 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 323 U.S. female nurses aged 61-80 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Vitamin B2 (mg)	0.49	0.61	0.68	(0.59, 0.74)	0.68	0.45	0.51	0.67	(0.53, 0.78)	0.30
With supplements	0.74	0.76	0.82	(0.75, 0.86)	0.74					
Vitamin B3 (mg)	0.30	0.46	0.55	(0.43, 0.64)	0.52	0.27	0.35	0.52	(0.35, 0.66)	0.20
With supplements	0.72	0.74	0.80	(0.74, 0.85)	0.73					
Vitamin B6 (mg)	0.36	0.49	0.57	(0.46, 0.65)	0.58	0.36	0.45	0.63	(0.43, 0.77)	0.19
With supplements	0.74	0.75	0.81	(0.74, 0.85)	0.74					
Vitamin B12 (mg)	0.44	0.46	0.56	(0.44, 0.65)	0.51	0.31	0.32	0.57	(0.35, 0.73)	0.12
With supplements	0.71	0.69	0.75	(0.67, 0.81)	0.73					
Natural folate (mcg)	0.41	0.55	0.61	(0.51, 0.68)	0.68	0.35	0.46	0.60	(0.47, 0.70)	0.33
Folic acid (mcg)	0.59	0.64	0.74	(0.65, 0.81)	0.59	0.48	0.47	0.56	(0.40, 0.68)	0.26
With supplements	0.72	0.72	0.78	(0.71, 0.83)	0.74					
Dietary folate eqvts (mcg)	0.46	0.60	0.71	(0.61, 0.79)	0.56	0.32	0.39	0.51	(0.34, 0.64)	0.22
With supplements	0.71	0.74	0.79	(0.73, 0.84)	0.78					
Vitamin C (mg)	0.49	0.51	0.59	(0.48, 0.67)	0.60	0.42	0.42	0.56	(0.43, 0.67)	0.32
With supplements	0.75	0.75	0.80	(0.74, 0.85)	0.78					
Vitamin D (mg)	0.55	0.53	0.62	(0.52, 0.70)	0.57	0.54	0.52	0.74	(0.55, 0.85)	0.21
With supplements	0.59	0.55	0.62	(0.52, 0.69)	0.64					
Vitamin E (mg)	0.43	0.47	0.56	(0.45, 0.65)	0.54	0.30	0.35	0.53	(0.37, 0.66)	0.21
With supplements	0.73	0.72	0.77	(0.70, 0.82)	0.79					
Vitamin K (mg)	0.50	0.54	0.65	(0.54, 0.74)	0.51	0.30	0.38	0.60	(0.40, 0.74)	0.17
With supplements	0.47	0.50	0.58	(0.48, 0.67)	0.57					
Calcium (mg)	0.51	0.61	0.68	(0.59, 0.74)	0.69	0.48	0.57	0.71	(0.54, 0.83)	0.25
With supplements	0.65	0.67	0.73	(0.66, 0.80)	0.72					
Magnesium (mg)	0.38	0.64	0.69	(0.61, 0.74)	0.74	0.35	0.58	0.74	(0.62, 0.82)	0.37
With supplements	0.60	0.75	0.79	(0.73, 0.84)	0.82					
Iron (mg)	0.30	0.51	0.60	(0.49, 0.68)	0.56	0.30	0.38	0.49	(0.33, 0.63)	0.22
With supplements	0.30	0.42	0.55	(0.42, 0.66)	0.40					

Supplementary Table 1.7 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 323 U.S. female nurses aged 61-80 years, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Copper (mg)	0.36	0.53	0.60	(0.50, 0.68)	0.64	0.31	0.46	0.55	(0.40, 0.67)	0.26
With supplements	0.53	0.57	0.64	(0.55, 0.71)	0.65					
Zinc (mg)	0.29	0.40	0.47	(0.36, 0.57)	0.53	0.29	0.31	0.53	(0.07, 0.81)	0.07
With supplements	0.66	0.66	0.74	(0.66, 0.81)	0.64					
Phosphorus (mg)	0.37	0.59	0.66	(0.57, 0.72)	0.69	0.39	0.53	0.71	(0.56, 0.82)	0.26
With supplements	0.37	0.60	0.66	(0.57, 0.72)	0.69					
Choline (mg)	0.38	0.51	0.58	(0.48, 0.66)	0.62	0.38	0.40	0.68	(0.46, 0.82)	0.15
With supplements	0.36	0.48	0.54	(0.44, 0.63)	0.63					
Potassium (mg)	0.40	0.54	0.58	(0.49, 0.66)	0.73	0.37	0.52	0.64	(0.52, 0.73)	0.34
With supplements	0.39	0.53	0.58	(0.49, 0.65)	0.74					
Sodium (mg)	0.28	0.47	0.57	(0.45, 0.67)	0.48	0.29	0.40	0.52	0.35, 0.66)	0.21
Caffeine (mg)	0.74	0.73	0.76	(0.70, 0.81)	0.85	0.67	0.67	0.75	(0.67, 0.82)	0.51

Supplementary Table 1.8: Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 298 U.S. female nurses with BMI<25kg/m2 at enrollment, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Total energy (kcal)	0.23		0.26	(0.14, 0.37)	0.62	0.33		0.43	(0.27, 0.56)	0.27
Total fat (g)	0.33	0.63	0.70	(0.61, 0.78)	0.65	0.35	0.58	0.80	(0.61, 0.90)	0.24
Saturated fat (g)	0.43	0.66	0.74	(0.65, 0.80)	0.68	0.43	0.62	0.68	(0.49, 0.81)	0.26
Polyunsaturated fat (g)	0.28	0.51	0.60	(0.49, 0.69)	0.54	0.30	0.47	0.71	(0.45, 0.86)	0.15
Monounsaturated fat (g)	0.37	0.54	0.62	(0.52, 0.70)	0.59	0.33	0.44	0.62	(0.44, 0.74)	0.22
Arachadonic FA (g)	0.35	0.43	0.52	(0.40, 0.62)	0.51	0.29	0.35	0.53	(0.33, 0.69)	0.16
Lauric FA (g)	0.30	0.35	0.44	(0.31, 0.55)	0.47	0.31	0.38	0.48	(0.32, 0.62)	0.24
Linoleic FA (g)	0.29	0.50	0.60	(0.48, 0.69)	0.54	0.31	0.45	0.73	(0.46, 0.88)	0.14
Linolenic FA (g)	0.39	0.51	0.61	(0.49, 0.70)	0.52	0.39	0.44	0.60	(0.42, 0.72)	0.23
N-3 (DHA+EPA) FA (g)	0.55	0.56	0.72	(0.59, 0.82)	0.43	0.29	0.32	0.70	(0.25, 0.90)	0.08
With supplements	0.64	0.63	0.70	(0.61, 0.77)	0.67					
Oleic FA (g)	0.37	0.53	0.62	(0.51, 0.70)	0.59	0.33	0.44	0.62	(0.42, 0.75)	0.20
Cholesterol (mg)	0.48	0.57	0.67	(0.57, 0.75)	0.56	0.44	0.48	0.62	(0.38, 0.78)	0.18
Protein (g)	0.34	0.50	0.57	(0.47, 0.66)	0.62	0.41	0.48	0.56	(0.36, 0.71)	0.20
Carbohydrate (g)	0.41	0.67	0.73	(0.65, 0.79)	0.75	0.54	0.59	0.69	(0.56, 0.79)	0.36
Total sugar (g)	0.56	0.72	0.78	(0.71, 0.83)	0.77	0.61	0.62	0.75	(0.64, 0.84)	0.37
Fiber (g)	0.45	0.62	0.66	(0.58, 0.73)	0.77	0.48	0.51	0.64	(0.52, 0.74)	0.35
Alcohol (g)	0.85	0.77	0.84	(0.78, 0.88)	0.72	0.75	0.66	0.74	(0.65, 0.82)	0.49
Retinol activity eqvts (mcg)	0.44	0.50	0.64	(0.51, 0.73)	0.44	0.42	0.42	0.52	(0.35, 0.66)	0.23
With supplements	0.63	0.66	0.74	(0.65, 0.81)	0.67					
Alpha carotene (mcg)	0.47	0.45	0.60	(0.45, 0.70)	0.39	0.37	0.33	0.48	(0.29, 0.64)	0.18
Beta carotene (mcg)	0.51	0.55	0.71	(0.58, 0.81)	0.42	0.45	0.43	0.58	(0.42, 0.70)	0.25
With supplements	0.47	0.51	0.65	(0.52, 0.74)	0.45					
Lutein-zeaxanthin (mcg)	0.58	0.62	0.74	(0.63, 0.82)	0.51	0.51	0.50	0.69	(0.54, 0.80)	0.27
Lycopene (mcg)	0.38	0.36	0.62	(0.37, 0.79)	0.20	0.24	0.12	0.25	(0.05, 0.42)	0.16
Beta cryptoxanthin (mcg)	0.38	0.42	0.58	(0.42, 0.71)	0.34	0.44	0.44	0.67	(0.40, 0.83)	0.14
Vitamin B1 (mg)	0.43	0.61	0.70	(0.61, 0.78)	0.59	0.44	0.49	0.61	(0.41, 0.75)	0.23
With supplements	0.76	0.76	0.83	(0.77, 0.87)	0.73					

Supplementary Table 1.8 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 298 U.S. female nurses with BMI<25kg/m2 at enrollment, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Vitamin B2 (mg)	0.58	0.65	0.71	(0.63, 0.78)	0.72	0.58	0.57	0.72	(0.59, 0.82)	0.32
With supplements	0.75	0.76	0.82	(0.77, 0.87)	0.75					
Vitamin B3 (mg)	0.36	0.49	0.57	(0.46, 0.66)	0.59	0.37	0.42	0.60	(0.43, 0.73)	0.22
With supplements	0.71	0.73	0.78	(0.71, 0.83)	0.80					
Vitamin B6 (mg)	0.36	0.49	0.55	(0.45, 0.64)	0.62	0.41	0.39	0.51	(0.35, 0.64)	0.25
With supplements	0.67	0.67	0.73	(0.65, 0.80)	0.71					
Vitamin B12 (mg)	0.46	0.49	0.57	(0.46, 0.67)	0.55	0.38	0.36	0.63	(0.37, 0.80)	0.13
With supplements	0.73	0.71	0.77	(0.70, 0.83)	0.72					
Natural folate (mcg)	0.49	0.64	0.72	(0.63, 0.79)	0.64	0.53	0.54	0.67	(0.55, 0.78)	0.35
Folic acid (mcg)	0.58	0.63	0.70	(0.62, 0.78)	0.65	0.50	0.49	0.53	(0.38, 0.65)	0.31
With supplements	0.74	0.73	0.79	(0.72, 0.84)	0.73					
Dietary folate eqvts (mcg)	0.46	0.56	0.64	(0.54, 0.72)	0.63	0.39	0.42	0.51	(0.36, 0.63)	0.29
With supplements	0.73	0.73	0.79	(0.72, 0.84)	0.75					
Vitamin C (mg)	0.51	0.55	0.62	(0.52, 0.70)	0.62	0.54	0.49	0.64	(0.50, 0.75)	0.29
With supplements	0.73	0.74	0.79	(0.73, 0.85)	0.75					
Vitamin D (mg)	0.65	0.65	0.74	(0.65, 0.81)	0.63	0.55	0.57	0.78	(0.60, 0.88)	0.24
With supplements	0.64	0.60	0.64	(0.56, 0.71)	0.74					
Vitamin E (mg)	0.37	0.42	0.50	(0.38, 0.60)	0.53	0.34	0.35	0.47	(0.33, 0.60)	0.28
With supplements	0.70	0.72	0.78	(0.71, 0.83)	0.73					
Vitamin K (mg)	0.56	0.62	0.74	(0.63, 0.82)	0.52	0.44	0.43	0.62	(0.46, 0.73)	0.24
With supplements	0.55	0.60	0.70	(0.60, 0.79)	0.55					
Calcium (mg)	0.59	0.68	0.73	(0.66, 0.80)	0.75	0.56	0.55	0.74	(0.55, 0.86)	0.24
With supplements	0.67	0.68	0.74	(0.66, 0.80)	0.74					
Magnesium (mg)	0.41	0.68	0.75	(0.67, 0.81)	0.71	0.49	0.59	0.74	(0.60, 0.83)	0.33
With supplements	0.57	0.74	0.79	(0.72, 0.84)	0.79					
Iron (mg)	0.32	0.50	0.57	(0.46, 0.65)	0.64	0.35	0.46	0.51	(0.35, 0.64)	0.27
With supplements	0.34	0.43	0.51	(0.39, 0.61)	0.54					

Supplementary Table 1.8 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 298 U.S. female nurses with BMI<25kg/m2 at enrollment, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Copper (mg)	0.35	0.55	0.62	(0.52, 0.70)	0.63	0.35	0.41	0.52	(0.37, 0.64)	0.29
With supplements	0.46	0.51	0.57	(0.47, 0.66)	0.65					
Zinc (mg)	0.34	0.41	0.49	(0.37, 0.59)	0.53	0.39	0.34	0.53	(0.34, 0.68)	0.16
With supplements	0.61	0.61	0.67	(0.59, 0.74)	0.68					
Phosphorus (mg)	0.43	0.67	0.74	(0.66, 0.81)	0.67	0.44	0.55	0.68	(0.48, 0.82)	0.22
With supplements	0.42	0.66	0.73	(0.65, 0.80)	0.68					
Choline (mg)	0.43	0.57	0.63	(0.54, 0.71)	0.67	0.46	0.42	0.62	(0.41, 0.77)	0.18
With supplements	0.40	0.54	0.60	(0.50, 0.68)	0.67					
Potassium (mg)	0.43	0.61	0.66	(0.57, 0.72)	0.73	0.54	0.53	0.66	(0.54, 0.75)	0.36
With supplements	0.43	0.61	0.66	(0.58, 0.73)	0.74					
Sodium (mg)	0.23	0.45	0.56	(0.43, 0.66)	0.47	0.25	0.25	0.39	(0.20, 0.55)	0.17
Caffeine (mg)	0.79	0.75	0.78	(0.72, 0.83)	0.86	0.74	0.73	0.76 v	(0.67, 0.83)	0.53

Supplementary Table 1.9: Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 334 U.S. female nurses with BMI>=25kg/m2 at enrollment, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Total energy (kcal)	0.31		0.34	(0.23, 0.43)	0.64	0.31		0.39	(0.25, 0.50)	0.32
Total fat (g)	0.37	0.55	0.62	(0.53, 0.69)	0.64	0.34	0.51	0.66	(0.45, 0.80)	0.21
Saturated fat (g)	0.44	0.55	0.63	(0.53, 0.70)	0.61	0.46	0.49	0.63	(0.45, 0.75)	0.23
Polyunsaturated fat (g)	0.29	0.42	0.53	(0.41, 0.64)	0.44	0.20	0.38	0.69	(0.39, 0.86)	0.11
Monounsaturated fat (g)	0.36	0.42	0.50	(0.39, 0.59)	0.57	0.29	0.38	0.55	(0.37, 0.68)	0.20
Arachadonic FA (g)	0.38	0.44	0.53	(0.41, 0.62)	0.52	0.21	0.31	0.50	(0.32, 0.65)	0.16
Lauric FA (g)	0.39	0.37	0.50	(0.36, 0.61)	0.38	0.39	0.30	0.46	(0.29, 0.60)	0.19
Linoleic FA (g)	0.29	0.40	0.50	(0.37, 0.61)	0.45	0.21	0.38	0.65	(0.39, 0.82)	0.12
Linolenic FA (g)	0.31	0.39	0.54	(0.39, 0.65)	0.37	0.15	0.22	0.56	(-0.18, 0.90)	0.04
N-3 (DHA+EPA) FA (g)	0.50	0.46	0.61	(0.48, 0.72)	0.39	0.27	0.28	0.52	(0.22, 0.74)	0.08
With supplements	0.61	0.58	0.68	(0.58, 0.75)	0.57					
Oleic FA (g)	0.36	0.42	0.49	(0.38, 0.58)	0.57	0.29	0.37	0.54	(0.37, 0.68)	0.19
Cholesterol (mg)	0.50	0.54	0.62	(0.53, 0.70)	0.58	0.35	0.38	0.58	(0.36, 0.74)	0.14
Protein (g)	0.32	0.46	0.52	(0.41, 0.61)	0.61	0.23	0.31	0.45	(0.30, 0.58)	0.23
Carbohydrate (g)	0.41	0.63	0.67	(0.59, 0.73)	0.73	0.45	0.62	0.71	(0.58, 0.81)	0.33
Total sugar (g)	0.51	0.64	0.70	(0.63, 0.77)	0.74	0.55	0.67	0.73	(0.62, 0.82)	0.37
Fiber (g)	0.47	0.62	0.66	(0.58, 0.72)	0.72	0.35	0.54	0.61	(0.47, 0.71)	0.30
Alcohol (g)	0.86	0.75	0.82	(0.77, 0.87)	0.73	0.74	0.64	0.73	(0.64, 0.81)	0.43
Retinol activity eqvts (mcg)	0.45	0.48	0.69	(0.52, 0.81)	0.32	0.37	0.42	0.65	(0.43, 0.80)	0.16
With supplements	0.65	0.65	0.74	(0.65, 0.80)	0.63					
Alpha carotene (mcg)	0.52	0.49	0.68	(0.53, 0.79)	0.36	0.41	0.42	0.75	(0.42, 0.90)	0.11
Beta carotene (mcg)	0.51	0.49	0.62	(0.50, 0.71)	0.45	0.42	0.45	0.76	(0.53, 0.89)	0.15
With supplements	0.52	0.48	0.60	(0.48, 0.69)	0.47					
Lutein-zeaxanthin (mcg)	0.49	0.49	0.61	(0.49, 0.70)	0.48	0.42	0.45	0.68	v(0.39, 0.85)	0.13
Lycopene (mcg)	0.42	0.38	0.61	(0.40, 0.75)	0.25	0.26	0.19	0.40	(0.13, 0.61)	0.08
Beta cryptoxanthin (mcg)	0.33	0.34	0.52	(0.35, 0.66)	0.27	0.36	0.42	0.54	(0.34, 0.69)	0.16
Vitamin B1 (mg)	0.30	0.47	0.59	(0.47, 0.69)	0.44	0.28	0.39	0.62	(0.42, 0.77)	0.15
With supplements	0.64	0.66	0.73	(0.65, 0.79)	0.69					

Supplementary Table 1.9 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 334 U.S. female nurses with BMI>=25kg/m2 at enrollment, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Vitamin B2 (mg)	0.47	0.57	0.64	(0.55, 0.71)	0.67	0.44	0.53	0.67	(0.54, 0.78)	0.30
With supplements	0.67	0.70	0.76	(0.69, 0.81)	0.74					
Vitamin B3 (mg)	0.35	0.43	0.52	(0.41, 0.62)	0.50	0.24	0.35	0.53	(0.30, 0.71)	0.13
With supplements	0.64	0.68	0.77	(0.69, 0.83)	0.64					
Vitamin B6 (mg)	0.40	0.48	0.57	(0.46, 0.66)	0.54	0.28	0.40	0.65	(0.40, 0.82)	0.13
With supplements	0.68	0.68	0.75	(0.68, 0.81)	0.74					
Vitamin B12 (mg)	0.42	0.43	0.56	(0.43, 0.66)	0.42	0.29	0.30	0.43	(0.17, 0.63)	0.11
With supplements	0.67	0.65	0.72	(0.64, 0.78)	0.71					
Natural folate (mcg)	0.46	0.56	0.63	(0.54, 0.70)	0.63	0.36	0.50	0.60	(0.46, 0.71)	0.27
Folic acid (mcg)	0.53	0.56	0.67	(0.57, 0.75)	0.52	0.43	0.41	0.63	(0.44, 0.77)	0.17
With supplements	0.64	0.66	0.72	(0.65, 0.79)	0.71					
Dietary folate eqvts (mcg)	0.45	0.54	0.65	(0.54, 0.73)	0.50	0.34	0.39	0.68	(0.43, 0.84)	0.12
With supplements	0.64	0.68	0.75	(0.67, 0.81)	0.72					
Vitamin C (mg)	0.47	0.50	0.60	(0.49, 0.69)	0.53	0.43	0.47	0.61	(0.46, 0.72)	0.25
With supplements	0.67	0.68	0.74	(0.67, 0.80)	0.71					
Vitamin D (mg)	0.55	0.51	0.62	(0.51, 0.71)	0.51	0.47	0.46	0.73	(0.52, 0.86)	0.17
With supplements	0.62	0.61	0.67	(0.59, 0.73)	0.70					
Vitamin E (mg)	0.43	0.50	0.61	(0.50, 0.69)	0.53	0.27	0.37	0.54	(0.37, 0.68)	0.18
With supplements	0.74	0.73	0.78	(0.72, 0.83)	0.80					
Vitamin K (mg)	0.51	0.53	0.67	(0.55, 0.77)	0.44	0.32	0.38	0.64	(0.29, 0.85)	0.11
With supplements	0.50	0.52	0.63	(0.52, 0.72)	0.49					
Calcium (mg)	0.52	0.57	0.64	(0.54, 0.71)	0.63	0.49	0.54	0.62	(0.46, 0.74)	0.26
With supplements	0.64	0.66	0.73	(0.65, 0.79)	0.71					
Magnesium (mg)	0.44	0.62	0.68	(0.61, 0.74)	0.74	0.31	0.56	0.70	(0.59, 0.79)	0.37
With supplements	0.59	0.71	0.74	(0.67, 0.80)	0.79					
Iron (mg)	0.37	0.44	0.56	(0.44, 0.66)	0.44	0.26	0.34	0.64	(0.40, 0.80)	0.11
With supplements	0.46	0.52	0.65	(0.54, 0.74)	0.46					

Supplementary Table 1.9 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 334 U.S. female nurses with BMI>=25kg/m2 at enrollment, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Copper (mg)	0.37	0.47	0.55	(0.45, 0.64)	0.57	0.25	0.38	0.56	(0.39, 0.69)	0.20
With supplements	0.55	0.60	0.68	(0.59, 0.75)	0.63					
Zinc (mg)	0.37	0.44	0.55	(0.43, 0.65)	0.46	0.26	0.26	0.53	(0.15, 0.78)	0.07
With supplements	0.61	0.61	0.69	(0.60, 0.75)	0.64					
Phosphorus (mg)	0.39	0.54	0.60	(0.51, 0.67)	0.68	0.33	0.49	0.66	(0.52, 0.77)	0.26
With supplements	0.41	0.55	0.61	(0.53, 0.69)	0.69					
Choline (mg)	0.44	0.53	0.61	(0.51, 0.69)	0.60	0.29	0.39	0.58	(0.40, 0.71)	0.19
With supplements	0.45	0.53	0.62	(0.52, 0.70)	0.58					
Potassium (mg)	0.47	0.58	0.64	(0.56, 0.71)	0.72	0.36	0.54	0.60	(0.48, 0.70)	0.34
With supplements	0.47	0.58	0.64	(0.56, 0.71)	0.72					
Sodium (mg)	0.30	0.40	0.48	(0.37, 0.58)	0.51	0.26	0.32	0.41	(0.24, 0.55)	0.21
Caffeine (mg)	0.76	0.76	0.81	(0.75, 0.85)	0.86	0.72	0.71	0.75	(0.66, 0.82)	0.48

Supplementary Table 1.10: Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 127 U.S. female nurses with BMI>=30 kg/m2 at enrollment, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Total energy (kcal)	0.25		0.29	(0.11, 0.45)	0.62	0.38		0.46	(0.25, 0.63)	0.36
Total fat (g)	0.30	0.55	0.62	(0.46, 0.74)	0.63	0.37	0.43	0.55	(0.02, 0.84)	0.11
Saturated fat (g)	0.40	0.53	0.64	(0.45, 0.77)	0.52	0.48	0.39	0.68	(0.27, 0.88)	0.14
Polyunsaturated fat (g)	0.20	0.42	0.56	(0.33, 0.73)	0.37	0.23	0.35	0.57	(0.00, 0.87)	0.10
Monounsaturated fat (g)	0.29	0.46	0.54	(0.37, 0.67)	0.58	0.34	0.38	0.71	(-0.16, 0.97)	0.07
Arachadonic FA (g)	0.31	0.40	0.48	(0.30, 0.64)	0.52	0.09	0.31	0.44	(0.12, 0.67)	0.17
Lauric FA (g)	0.42	0.43	0.59	(0.35, 0.75)	0.37	0.47	0.34	0.53	(0.04, 0.82)	0.11
Linoleic FA (g)	0.22	0.39	0.53	(0.29, 0.70)	0.37	0.26	0.39	0.46	(-0.1, 0.80)	0.10
Linolenic FA (g)	0.20	0.35	0.51	(0.25, 0.71)	0.31	0.14	0.17	0.46	(-0.07, 0.79)	0.08
N-3 (DHA+EPA) FA (g)	0.43	0.40	0.50	(0.30, 0.66)	0.44	0.17	0.25	0.39	(0.10, 0.62)	0.18
With supplements	0.58	0.53	0.62	(0.44, 0.75)	0.54					
Oleic FA (g)	0.29	0.46	0.53	(0.36, 0.67)	0.58	0.35	0.38	0.79	(-0.69, 1.00)	0.05
Cholesterol (mg)	0.44	0.44	0.52	(0.34, 0.66)	0.57	0.27	0.38	0.57	(0.24, 0.79)	0.16
Protein (g)	0.24	0.42	0.48	(0.31, 0.62)	0.61	0.23	0.39	0.46	(0.16, 0.68)	0.21
Carbohydrate (g)	0.42	0.65	0.71	(0.58, 0.80)	0.73	0.55	0.53	0.68	(0.45, 0.83)	0.29
Total sugar (g)	0.57	0.70	0.75	(0.64, 0.83)	0.78	0.74	0.68	0.72	(0.48, 0.86)	0.35
Fiber (g)	0.43	0.52	0.59	(0.43, 0.72)	0.63	0.41	0.51	0.61	(0.38, 0.77)	0.33
Alcohol (g)	0.85	0.70	0.77	(0.65, 0.86)	0.70	0.75	0.55	0.66	(0.46, 0.80)	0.36
Retinol activity eqvts (mcg)*	0.41	0.41		N/A	-0.02	0.44	0.44	0.84	(-0.44, 1.00)	0.09
With supplements	0.62	0.60	0.72	(0.55, 0.83)	0.54					
Alpha carotene (mcg)	0.48	0.45	0.66	(0.36, 0.84)	0.31	0.38	0.40	0.71	(-0.14, 0.96)	0.09
Beta carotene (mcg)	0.48	0.48	0.71	(0.37, 0.88)	0.29	0.48	0.54	0.78	(-0.25, 0.98)	0.11
With supplements	0.48	0.44	0.61	(0.36, 0.78)	0.36					
Lutein-zeaxanthin (mcg)	0.47	0.51	0.76	(0.38, 0.92)	0.29	0.49	0.54	0.47	(-0.11, 0.81)	0.16
Lycopene (mcg)	0.39	0.38	0.69	(0.11, 0.92)	0.17	0.21	0.19	0.4	(-0.19, 0.79)	0.06
Beta cryptoxanthin (mcg)	0.23	0.28	0.53	(0.07, 0.81)	0.16	0.28	0.41	0.45	(0.07, 0.72)	0.13
Vitamin B1 (mg)	0.25	0.40	0.58	(0.29, 0.78)	0.29	0.29	0.34	0.60	(0.14, 0.85)	0.11
With supplements	0.60	0.60	0.67	(0.53, 0.78)	0.66					

Supplementary Table 1.10 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 127 U.S. female nurses with BMI>=30 kg/m2 at enrollment, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Vitamin B2 (mg)	0.40	0.48	0.58	(0.39, 0.72)	0.52	0.42	0.48	0.58	(0.23, 0.80)	0.20
With supplements	0.64	0.67	0.72	(0.60, 0.81)	0.77					
Vitamin B3 (mg)	0.30	0.36	0.46	(0.25, 0.63)	0.42	0.21	0.38	0.34	(-0.26, 0.75)	0.09
With supplements	0.60	0.62	0.74	(0.57, 0.85)	0.53					
Vitamin B6 (mg)	0.35	0.46	0.58	(0.37, 0.73)	0.43	0.27	0.41	0.59	(0.25, 0.80)	0.18
With supplements	0.59	0.58	0.63	(0.49, 0.74)	0.70					
Vitamin B12 (mg)	0.44	0.41	0.52	(0.31, 0.69)	0.41	0.33	0.33	0.58	(0.10, 0.85)	0.11
With supplements	0.56	0.51	0.55	(0.40, 0.67)	0.74					
Natural folate (mcg)	0.38	0.54	0.63	(0.46, 0.75)	0.57	0.38	0.52	0.79	(0.38, 0.93)	0.19
Folic acid (mcg)	0.53	0.56	0.65	(0.48, 0.77)	0.59	0.47	0.37	0.59	(0.23, 0.81)	0.15
With supplements	0.61	0.63	0.69	(0.55, 0.79)	0.70					
Dietary folate eqvts (mcg)	0.39	0.47	0.59	(0.38, 0.74)	0.45	0.34	0.42	0.55	(0.21, 0.78)	0.17
With supplements	0.60	0.63	0.70	(0.56, 0.81)	0.66					
Vitamin C (mg)	0.39	0.42	0.51	(0.32, 0.67)	0.47	0.31	0.46	0.43	(0.14, 0.65)	0.24
With supplements	0.61	0.59	0.65	(0.51, 0.77)	0.69					
Vitamin D (mg)	0.50	0.44	0.52	(0.34, 0.67)	0.53	0.57	0.56	0.67	(0.23, 0.88)	0.18
With supplements	0.64	0.61	0.67	(0.53, 0.78)	0.69					
Vitamin E (mg)	0.43	0.54	0.69	(0.48, 0.83)	0.44	0.31	0.37	0.43	(0.16, 0.64)	0.23
With supplements	0.78	0.76	0.80	(0.70, 0.87)	0.79					
Vitamin K (mg)	0.43	0.52	0.77	(0.41, 0.92)	0.31	0.43	0.56	0.39	(-0.39, 0.85)	0.11
With supplements	0.45	0.50	0.67	(0.43, 0.82)	0.38					
Calcium (mg)	0.49	0.52	0.59	(0.43, 0.71)	0.62	0.50	0.50	0.56	(0.25, 0.77)	0.21
With supplements	0.64	0.65	0.71	(0.58, 0.81)	0.71					
Magnesium (mg)	0.39	0.51	0.56	(0.40, 0.68)	0.67	0.36	0.48	0.48	(0.26, 0.66)	0.37
With supplements	0.51	0.56	0.60	(0.46, 0.71)	0.72					
Iron (mg)	0.29	0.33	0.45	(0.21, 0.64)	0.35	0.23	0.27	0.48	(-0.44, 0.91)	0.05
With supplements	0.40	0.49	0.60	(0.40, 0.74)	0.48					

Supplementary Table 1.10 (continued): Spearman correlation coefficient for comparison of daily nutrient intake from SFFQ2 with the means of two 7DDRs, and four ASA24s, unadjusted and energy-adjusted and de-attenuated results (data provided by 127 U.S. female nurses with BMI>=30 kg/m2 at enrollment, 2010-2012)

Nutrient	SFFQ2 vs. 7DDR					SFFQ2 vs. ASA24				
	Un-adjusted	Energy-adjusted				Un-adjusted	Energy-adjusted			
		Residual method	De-attenuated (ρ & 95%CI)		ICC		Residual method	De-attenuated (ρ & 95%CI)		ICC
Copper (mg)	0.30	0.38	0.48	(0.28, 0.65)	0.45	0.27	0.41	0.49	(0.23, 0.68)	0.25
With supplements	0.57	0.58	0.68	(0.52, 0.80)	0.58					
Zinc (mg)	0.44	0.48	0.60	(0.40, 0.73)	0.48	0.31	0.25	0.68	(-0.67, 0.99)	0.04
With supplements	0.62	0.58	0.66	(0.51, 0.78)	0.63					
Phosphorus (mg)	0.36	0.50	0.55	(0.39, 0.68)	0.67	0.38	0.53	0.65	(0.34, 0.84)	0.22
With supplements	0.39	0.53	0.58	(0.43, 0.70)	0.70					
Choline (mg)	0.42	0.51	0.58	(0.41, 0.70)	0.63	0.26	0.39	0.48	(0.16, 0.70)	0.20
With supplements	0.42	0.51	0.58	(0.41, 0.71)	0.61					
Potassium (mg)	0.44	0.51	0.57	(0.40, 0.69)	0.65	0.38	0.43	0.43	(0.19, 0.62)	0.32
With supplements	0.46	0.53	0.59	(0.43, 0.71)	0.65					
Sodium (mg)	0.16	0.27	0.34	(0.12, 0.52)	0.43	0.31	0.29	0.39	(0.13, 0.60)	0.24
Caffeine (mg)	0.73	0.75	0.78	(0.68, 0.85)	0.86	0.75	0.72	0.76	(0.55, 0.87)	0.40

* The ICC for Retinol activity equivalent in 7DDR was -0.02, thus de-attenuated Spearman correlation coefficient was not available.

Supplementary Table 1.11: Regression coefficients of predicting the average daily nutrient intakes of two 7DDRs, and four ASA24s using daily nutrient intakes from each SFFQ, unadjusted and energy-adjusted using energy density and residual method (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ2 vs.				SFFQ1 vs.				WebFFQ vs.			
	7DDR		ASA24		7DDR		ASA24		7DDR		ASA24	
	Un-adjusted	Residual method	Un-adjusted	Residual method	Un-adjusted	Residual method	Un-adjusted	Residual method	Un-adjusted	Residual method	Un-adjusted	Residual method
Total energy (kcal)	0.20		0.27		0.21		0.30		0.19		0.25	
Total fat (g)	0.29	0.55	0.35	0.57	0.31	0.53	0.36	0.55	0.25	0.51	0.31	0.53
Saturated fat (g)	0.39	0.58	0.47	0.57	0.39	0.59	0.45	0.57	0.34	0.61	0.40	0.58
Polyunsaturated fat (g)	0.24	0.45	0.27	0.52	0.30	0.46	0.35	0.53	0.23	0.46	0.29	0.50
Monounsaturated fat (g)	0.29	0.41	0.32	0.41	0.31	0.40	0.33	0.41	0.26	0.39	0.30	0.38
Arachadonic FA (g)	0.34	0.40	0.44	0.54	0.37	0.56	0.33	0.57	0.34	0.44	0.34	0.53
Lauric FA (g)	0.34	0.32	0.37	0.33	0.41	0.33	0.40	0.38	0.36	0.33	0.35	0.38
Linoleic FA (g)	0.25	0.44	0.28	0.53	0.50	0.47	0.78	0.53	0.46	0.46	0.79	0.50
Linolenic FA (g)	0.29	0.42	0.26	0.33	0.56	0.41	0.71	0.32	0.52	0.43	0.65	0.36
N-3 (DHA+EPA) FA (g)	0.50	0.54	0.33	0.41	0.49	0.52	0.57	0.39	0.48	0.42	0.58	0.30
With supplements	0.66	0.69	0.65	0.65	0.53	0.56						
Oleic FA (g)	0.29	0.40	0.31	0.40	0.45	0.39	0.39	0.40	0.48	0.38	0.60	0.37
Cholesterol (mg)	0.44	0.54	0.50	0.57	0.51	0.62	0.44	0.45	0.42	0.54	0.43	0.52
Protein (g)	0.25	0.48	0.29	0.48	0.28	0.47	0.35	0.46	0.24	0.48	0.29	0.49
Carbohydrates (g)	0.34	0.75	0.43	0.78	0.35	0.76	0.44	0.72	0.31	0.70	0.38	0.73
Total sugar (g)	0.48	0.73	0.57	0.76	0.50	0.75	0.58	0.74	0.46	0.73	0.52	0.76
Fiber (g)	0.41	0.70	0.45	0.67	0.46	0.74	0.52	0.72	0.40	0.70	0.45	0.70
Alcohol (g)	0.49	0.48	0.58	0.57	0.28	0.48	0.30	0.57	0.27	0.47	0.31	0.57
Retinol activity eqvts (mcg)	0.43	0.49	0.50	0.54	0.34	0.48	0.37	0.54	0.30	0.46	0.27	0.48
With supplements	0.63	0.67	0.62	0.64	0.60	0.65						
Alpha carotene (mcg)	0.53	0.55	0.85	0.87	0.45	0.49	0.60	0.78	0.41	0.47	0.53	0.86
Beta carotene (mcg)	0.57	0.63	0.71	0.78	0.46	0.60	0.66	0.78	0.47	0.58	0.61	0.72
With supplements	0.58	0.63	0.55	0.57	0.52	0.58						
Lutein-zeaxanthin (mcg)	0.54	0.60	0.64	0.72	0.43	0.57	0.60	0.69	0.38	0.55	0.54	0.64
Lycopene (mcg)	0.48	0.48	0.66	0.49	0.20	0.44	0.18	0.29	0.18	0.49	0.20	0.47
Beta cryptoxanthin (mcg)	0.41	0.48	0.90	1.03	0.31	0.46	0.35	0.94	0.23	0.44	0.29	0.93
Vitamin B1 (mg)	0.29	0.47	0.33	0.50	0.33	0.42	0.28	0.36	0.29	0.57	0.28	0.53
With supplements	0.72	0.73	0.68	0.68	0.68	0.70						

Supplementary Table 1.11 (continued): Regression coefficients of predicting the average daily nutrient intakes of two 7DDRs, and four ASA24s using daily nutrient intakes from each SFFQ, unadjusted and energy-adjusted (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ2 vs.				SFFQ1 vs.				WebFFQ vs.			
	7DDR		ASA24		7DDR		ASA24		7DDR		ASA24	
	Un-	Residual method	Un-	Residual method	Un-	Residual method	Un-	Residual method	Un-	Residual method	Un-	Residual method
Vitamin B2 (mg)	0.44	0.55	0.45	0.54	0.43	0.54	0.53	0.52	0.41	0.58	0.49	0.58
With supplements	0.60	0.60	0.56	0.56	0.56	0.57						
Vitamin B3 (mg)	0.31	0.52	0.32	0.52	0.31	0.49	0.33	0.48	0.26	0.52	0.29	0.41
With supplements	0.77	0.83	0.77	0.80	0.74	0.80						
Vitamin B6 (mg)	0.36	0.59	0.37	0.54	0.36	0.53	0.42	0.46	0.31	0.63	0.37	0.60
With supplements	0.55	0.55	0.52	0.52	0.51	0.51						
Vitamin B12 (mg)	0.43	0.49	0.39	0.39	0.40	0.44	0.39	0.36	0.42	0.52	0.37	0.38
With supplements	0.70	0.70	0.69	0.69	0.68	0.68						
Natural folate (mcg)	0.42	0.61	0.39	0.60	0.42	0.56	0.40	0.55	0.38	0.58	0.37	0.55
Folic acid (mcg)	0.79	0.84	0.81	0.80	0.44	0.76	0.45	0.88	0.40	0.88	0.44	0.88
With supplements	0.66	0.67	0.62	0.63	0.71	0.73						
Dietary folate eqvts (mcg)	0.42	0.57	0.36	0.43	0.35	0.49	0.38	0.37	0.32	0.53	0.36	0.46
With supplements	0.71	0.75	0.68	0.70	0.69	0.76						
Vitamin C (mg)	0.50	0.61	0.67	0.80	0.49	0.56	0.67	0.78	0.45	0.55	0.64	0.76
With supplements	0.81	0.85	0.79	0.82	0.78	0.81						
Vitamin D (mg)	0.57	0.61	0.66	0.72	0.54	0.56	0.68	0.69	0.52	0.58	0.61	0.66
With supplements	0.89	0.90	0.82	0.81	0.83	0.84						
Vitamin E (mg)	0.36	0.48	0.31	0.37	0.39	0.48	0.33	0.35	0.29	0.43	0.26	0.33
With supplements	0.71	0.73	0.72	0.74	0.66	0.68						
Vitamin K (mg)	0.47	0.53	0.44	0.52	0.47	0.53	0.45	0.53	0.44	0.50	0.39	0.47
With supplements	0.49	0.56	0.50	0.56	0.45	0.51						
Calcium (mg)	0.45	0.58	0.54	0.65	0.34	0.55	0.30	0.65	0.34	0.54	0.29	0.58
With supplements	0.66	0.71	0.66	0.71	0.61	0.68						
Magnesium (mg)	0.37	0.84	0.39	0.86	0.50	0.77	0.38	0.83	0.39	0.78	0.28	0.76
With supplements	0.63	0.96	0.61	0.90	0.57	0.90						
Iron (mg)	0.33	0.58	0.31	0.46	0.36	0.55	0.41	0.39	0.32	0.67	0.38	0.49
With supplements	0.41	0.51	0.44	0.50	0.36	0.43						

Supplementary Table 1.11 (continued): Regression coefficients of predicting the average daily nutrient intakes of two 7DDRs, and four ASA24s using daily nutrient intakes from each SFFQ, unadjusted and energy-adjusted (data provided by 632 U.S. female nurses aged 45-80 years, 2010-2012)

Nutrient	SFFQ2 vs.				SFFQ1 vs.				WebFFQ vs.			
	7DDR		ASA24		7DDR		ASA24		7DDR		ASA24	
	Un-	Residual method	Un-	Residual method	Un-	Residual method	Un-	Residual method	Un-	Residual method	Un-	Residual method
Copper (mg)	0.33	0.55	0.30	0.47	0.41	0.54	0.45	0.45	0.37	0.56	0.42	0.44
With supplements	0.49	0.54	0.49	0.55	0.43	0.49						
Zinc (mg)	0.34	0.52	0.34	0.39	0.33	0.45	0.35	0.38	0.30	0.53	0.29	0.38
With supplements	0.62	0.65	0.58	0.62	0.54	0.59						
Phosphorus (mg)	0.32	0.62	0.35	0.61	0.48	0.53	0.51	0.58	0.37	0.57	0.45	0.58
With supplements	0.32	0.62	0.33	0.54	0.28	0.56						
Choline (mg)	0.37	0.60	0.36	0.54	0.45	0.61	0.67	0.48	0.43	0.59	0.60	0.48
With supplements	0.36	0.56	0.42	0.62	0.36	0.59						
Potassium (mg)	0.39	0.77	0.42	0.75	0.33	0.68	0.42	0.68	0.32	0.66	0.39	0.66
With supplements	0.39	0.77	0.42	0.69	0.35	0.67						
Sodium (mg)	0.22	0.42	0.24	0.30	0.71	0.37	0.83	0.25	0.78	0.40	0.84	0.33
Caffeine (mg)	0.80	0.81	1.46	1.46	0.35	0.90	0.34	1.58	0.31	0.62	0.32	1.12

Supplementary Table 2.1: Spearman correlation coefficients for energy intakes estimated from FFQs, two 7DDRs, four ASA24s and four repeated biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

Methods	N	DLW measurements										
		Day 1	Day 2	Day 3	Day 4	Day 1 & 2	Day 3 & 4	Day 1 & 3	Day 2 & 4	Day 1 & 4	Day 2 & 3	Days, all
N		164	162	181	193	326	374	317	355	309	343	624
SFFQ2	624	0.03	0.11	0.11	0.14	0.08	0.13	0.09	0.14	0.07	0.12	0.11
SFFQ1	624	0.00	0.15	0.17	0.05	0.08	0.10	0.10	0.10	0.01	0.16	0.10
SFFQ1&2	624	0.02	0.14	0.16	0.10	0.09	0.13	0.11	0.13	0.05	0.15	0.11
WebFFQ	624	0.03	0.16	0.14	0.12	0.11	0.12	0.09	0.15	0.06	0.16	0.12
7DDR												
Week 1	623	0.42	0.44	0.47	0.29	0.43	0.38	0.44	0.37	0.33	0.46	0.39
Week 2	623	0.31	0.47	0.40	0.30	0.39	0.35	0.37	0.37	0.31	0.43	0.38
Week 1 & 2	624	0.39	0.51	0.45	0.32	0.45	0.39	0.43	0.41	0.34	0.48	0.42
ASA24												
Day 1	462	0.08	0.36	0.09	0.10	0.21	0.08	0.08	0.22	0.08	0.21	0.15
Day 2	454	0.17	0.28	0.09	0.12	0.22	0.11	0.16	0.19	0.15	0.18	0.18
Day 3	452	0.09	0.29	0.03	0.10	0.17	0.06	0.06	0.18	0.09	0.16	0.12
Day 4	452	-0.01	0.24	0.20	0.15	0.13	0.17	0.09	0.21	0.10	0.23	0.17
Day 1 & 2	564	0.14	0.37	0.13	0.13	0.24	0.12	0.13	0.24	0.13	0.23	0.19
Day 3 & 4	547	0.10	0.30	0.13	0.10	0.20	0.11	0.10	0.19	0.10	0.22	0.16
Day 1 & 3	578	0.09	0.30	0.04	0.17	0.19	0.10	0.06	0.22	0.13	0.17	0.15
Day 2 & 4	564	0.13	0.31	0.19	0.11	0.23	0.14	0.16	0.20	0.14	0.25	0.20
Day 1 & 4	577	0.04	0.36	0.16	0.18	0.22	0.16	0.09	0.26	0.12	0.25	0.19
Day 2 & 3	559	0.16	0.36	0.06	0.12	0.25	0.09	0.12	0.22	0.12	0.20	0.17
Days, all	624	0.12	0.38	0.15	0.17	0.26	0.15	0.13	0.26	0.14	0.26	0.21

Supplementary Table 2.2a: Spearman correlation coefficients for absolute protein intakes estimated from FFQs, 7DDRs, ASA24s and biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

Methods		Urinary biomarkers for protein (g/day)										
	N	Day 1	Day 2	Day 3	Day 4	Day 1 & 2	Day 3 & 4	Day 1 & 3	Day 2 & 4	Day 1 & 4	Day 2 & 3	Days, all
N		607	600	605	609	622	624	624	624	623	623	624
SFFQ2	624	0.19	0.23	0.19	0.24	0.23	0.24	0.21	0.26	0.24	0.23	0.25
SFFQ1	624	0.25	0.23	0.22	0.27	0.27	0.26	0.26	0.27	0.28	0.25	0.29
SFFQ1&2	624	0.24	0.25	0.22	0.28	0.27	0.28	0.26	0.29	0.29	0.26	0.30
WebFFQ	624	0.18	0.22	0.21	0.22	0.22	0.24	0.22	0.24	0.22	0.24	0.24
7DDR												
Week 1	623	0.51	0.52	0.50	0.48	0.57	0.54	0.57	0.54	0.55	0.57	0.60
Week 2	623	0.46	0.51	0.50	0.50	0.54	0.55	0.55	0.55	0.54	0.56	0.59
Week 1 & 2	624	0.53	0.56	0.55	0.53	0.61	0.60	0.61	0.60	0.59	0.62	0.65
ASA24												
Day 1	462	0.29	0.31	0.31	0.27	0.33	0.32	0.34	0.31	0.31	0.34	0.34
Day 2	454	0.22	0.27	0.23	0.26	0.26	0.27	0.25	0.27	0.27	0.26	0.28
Day 3	452	0.19	0.22	0.27	0.23	0.24	0.27	0.26	0.24	0.23	0.27	0.27
Day 4	452	0.23	0.23	0.24	0.26	0.25	0.27	0.26	0.27	0.27	0.26	0.28
Day 1 & 2	564	0.29	0.32	0.30	0.29	0.34	0.32	0.33	0.32	0.31	0.34	0.35
Day 3 & 4	547	0.23	0.29	0.30	0.30	0.29	0.33	0.30	0.32	0.29	0.32	0.33
Day 1 & 3	578	0.28	0.30	0.33	0.28	0.33	0.34	0.35	0.31	0.31	0.35	0.35
Day 2 & 4	564	0.24	0.28	0.26	0.30	0.28	0.30	0.28	0.31	0.30	0.29	0.31
Day 1 & 4	577	0.28	0.30	0.28	0.29	0.32	0.31	0.32	0.32	0.31	0.32	0.34
Day 2 & 3	559	0.23	0.29	0.28	0.28	0.29	0.30	0.29	0.30	0.28	0.30	0.31
Days, all	624	0.30	0.34	0.33	0.33	0.36	0.37	0.36	0.36	0.35	0.37	0.38

Supplementary Table 2.2b: Spearman correlation coefficients for protein nutrient density (using concurrent DLW assessment) estimated from FFQs, 7DDRs, ASA24s and biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

		Urinary biomarkers for protein (% energy)										
Methods	**N**	**Day 1**	**Day 2**	**Day 3**	**Day 4**	**Day 1 & 2**	**Day 3 & 4**	**Day 1 & 3**	**Day 2 & 4**	**Day 1 & 4**	**Day 2 & 3**	**Days, all**
N		156	157	177	189	313	366	307	346	301	334	609
SFFQ2	624	0.21	0.36	0.19	0.34	0.28	0.28	0.20	0.35	0.31	0.28	0.30
SFFQ1	624	0.25	0.37	0.22	0.31	0.32	0.27	0.25	0.34	0.30	0.30	0.31
SFFQ1&2	624	0.25	0.41	0.23	0.35	0.32	0.30	0.25	0.37	0.33	0.32	0.33
WebFFQ	624	0.24	0.26	0.20	0.35	0.25	0.28	0.23	0.32	0.32	0.24	0.28
7DDR												
Week 1	623	0.39	0.42	0.32	0.35	0.40	0.34	0.36	0.38	0.39	0.37	0.38
Week 2	623	0.34	0.41	0.26	0.43	0.37	0.35	0.32	0.42	0.41	0.33	0.38
Week 1 & 2	624	0.40	0.45	0.32	0.43	0.43	0.38	0.38	0.44	0.43	0.39	0.42
ASA24												
Day 1	462	0.20	0.12	0.23	0.24	0.16	0.24	0.24	0.19	0.25	0.18	0.23
Day 2	454	0.38	0.23	0.23	0.17	0.31	0.19	0.33	0.20	0.28	0.23	0.26
Day 3	452	0.27	0.37	0.16	0.33	0.31	0.24	0.24	0.35	0.30	0.27	0.29
Day 4	452	0.27	0.22	0.00	0.33	0.26	0.17	0.12	0.28	0.33	0.10	0.21
Day 1 & 2	564	0.36	0.21	0.27	0.17	0.30	0.22	0.33	0.20	0.27	0.24	0.27
Day 3 & 4	547	0.32	0.33	0.04	0.38	0.33	0.23	0.19	0.36	0.37	0.19	0.28
Day 1 & 3	578	0.33	0.29	0.21	0.32	0.30	0.27	0.30	0.31	0.33	0.26	0.31
Day 2 & 4	564	0.45	0.27	0.10	0.23	0.36	0.17	0.27	0.25	0.34	0.18	0.26
Day 1 & 4	577	0.29	0.21	0.12	0.32	0.26	0.23	0.21	0.27	0.33	0.16	0.25
Day 2 & 3	559	0.39	0.36	0.20	0.24	0.37	0.21	0.32	0.29	0.31	0.28	0.30
Days, all	624	0.45	0.35	0.17	0.29	0.39	0.24	0.33	0.32	0.37	0.26	0.32

Supplementary Table 2.2c: Spearman correlation coefficients for absolute sodium intakes estimated from FFQs, 7DDRs, ASA24s and biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

		Urinary biomarkers for sodium (mg/day)										
Methods	**N**	**Day 1**	**Day 2**	**Day 3**	**Day 4**	**Day 1 & 2**	**Day 3 & 4**	**Day 1 & 3**	**Day 2 & 4**	**Day 1 & 4**	**Day 2 & 3**	**Days, all**
N		607	603	606	605	623	623	624	624	623	623	624
SFFQ2	624	0.09	0.10	0.11	0.16	0.12	0.15	0.12	0.17	0.15	0.12	0.15
SFFQ1	624	0.14	0.10	0.15	0.16	0.15	0.17	0.17	0.16	0.18	0.14	0.18
SFFQ1&2	624	0.13	0.11	0.14	0.17	0.15	0.18	0.16	0.18	0.18	0.14	0.18
WebFFQ	624	0.08	0.09	0.13	0.16	0.11	0.16	0.12	0.16	0.14	0.12	0.15
7DDR												
Week 1	623	0.29	0.37	0.33	0.36	0.40	0.41	0.37	0.43	0.39	0.41	0.46
Week 2	623	0.28	0.34	0.28	0.32	0.38	0.36	0.33	0.38	0.35	0.36	0.41
Week 1 & 2	624	0.32	0.39	0.33	0.38	0.43	0.42	0.39	0.45	0.41	0.43	0.48
ASA24												
Day 1	462	0.12	0.19	0.14	0.19	0.18	0.19	0.13	0.22	0.18	0.19	0.20
Day 2	454	0.13	0.15	0.12	0.18	0.16	0.17	0.13	0.18	0.18	0.15	0.17
Day 3	452	0.08	0.14	0.15	0.13	0.13	0.17	0.13	0.16	0.12	0.16	0.17
Day 4	452	0.23	0.20	0.19	0.15	0.25	0.21	0.23	0.20	0.22	0.23	0.26
Day 1 & 2	564	0.17	0.20	0.14	0.24	0.21	0.21	0.15	0.24	0.23	0.19	0.22
Day 3 & 4	547	0.18	0.20	0.22	0.18	0.22	0.24	0.22	0.21	0.20	0.24	0.26
Day 1 & 3	578	0.13	0.20	0.16	0.20	0.19	0.22	0.15	0.23	0.19	0.21	0.22
Day 2 & 4	564	0.20	0.21	0.16	0.21	0.24	0.21	0.19	0.23	0.24	0.21	0.25
Day 1 & 4	577	0.20	0.22	0.18	0.20	0.25	0.22	0.21	0.24	0.23	0.24	0.26
Day 2 & 3	559	0.12	0.15	0.16	0.18	0.16	0.20	0.15	0.18	0.17	0.17	0.19
Days, all	624	0.21	0.23	0.20	0.25	0.26	0.27	0.22	0.28	0.27	0.25	0.29

Supplementary Table 2.2d: Spearman correlation coefficients for sodium density (using concurrent DLW assessment) estimated from FFQs, 7DDRs, ASA24s and biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

		Urinary biomarkers for sodium (mg/1000 kcal)										
Methods	**N**	**Day 1**	**Day 2**	**Day 3**	**Day 4**	**Day 1 & 2**	**Day 3 & 4**	**Day 1 & 3**	**Day 2 & 4**	**Day 1 & 4**	**Day 2 & 3**	**Days, all**
N		157	157	176	187	314	363	307	344	302	333	609
SFFQ2	624	0.16	0.18	0.23	0.33	0.17	0.27	0.19	0.27	0.25	0.21	0.23
SFFQ1	624	0.21	0.20	0.20	0.26	0.20	0.22	0.20	0.23	0.26	0.20	0.22
SFFQ1&2	624	0.21	0.21	0.21	0.32	0.22	0.25	0.20	0.27	0.28	0.21	0.24
WebFFQ	624	0.16	0.22	0.21	0.34	0.19	0.27	0.18	0.29	0.28	0.21	0.25
7DDR												
Week 1	623	0.13	0.26	0.23	0.33	0.20	0.27	0.17	0.29	0.27	0.24	0.25
Week 2	623	0.33	0.28	0.24	0.37	0.30	0.30	0.28	0.32	0.36	0.26	0.31
Week 1 & 2	624	0.26	0.34	0.26	0.39	0.29	0.32	0.25	0.36	0.35	0.30	0.32
ASA24												
Day 1	462	0.13	0.09	0.04	0.07	0.12	0.05	0.08	0.08	0.11	0.07	0.09
Day 2	454	0.15	0.16	0.15	0.10	0.16	0.12	0.14	0.13	0.12	0.15	0.14
Day 3	452	0.24	0.22	0.07	0.22	0.23	0.13	0.17	0.22	0.20	0.15	0.18
Day 4	452	0.30	0.19	0.13	0.19	0.24	0.13	0.20	0.18	0.24	0.15	0.18
Day 1 & 2	564	0.21	0.13	0.12	0.11	0.17	0.11	0.15	0.11	0.17	0.12	0.14
Day 3 & 4	547	0.31	0.25	0.10	0.21	0.28	0.14	0.21	0.23	0.24	0.17	0.20
Day 1 & 3	578	0.23	0.16	0.07	0.19	0.20	0.11	0.14	0.18	0.20	0.12	0.16
Day 2 & 4	564	0.30	0.19	0.13	0.12	0.24	0.12	0.20	0.15	0.19	0.15	0.17
Day 1 & 4	577	0.22	0.10	0.11	0.14	0.16	0.11	0.16	0.12	0.18	0.11	0.14
Day 2 & 3	559	0.25	0.20	0.14	0.17	0.22	0.15	0.21	0.18	0.18	0.17	0.18
Days, all	624	0.32	0.19	0.11	0.15	0.25	0.13	0.21	0.16	0.22	0.14	0.18

Supplementary Table 2.2e: Spearman correlation coefficients for absolute potassium intakes estimated from FFQs, 7DDRs, ASA24s and biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

Methods	N	Urinary biomarkers for potassium (mg/day)										
		Day 1	Day 2	Day 3	Day 4	Day 1 & 2	Day 3 & 4	Day 1 & 3	Day 2 & 4	Day 1 & 4	Day 2 & 3	Days, all
N		607	603	606	605	623	623	624	624	623	623	624
SFFQ2	624	0.26	0.30	0.23	0.26	0.32	0.27	0.27	0.30	0.29	0.29	0.31
SFFQ1	624	0.30	0.29	0.24	0.24	0.33	0.26	0.30	0.29	0.30	0.29	0.32
SFFQ1&2	624	0.30	0.32	0.25	0.27	0.35	0.28	0.31	0.31	0.32	0.31	0.34
WebFFQ	624	0.26	0.28	0.25	0.23	0.29	0.26	0.28	0.27	0.26	0.28	0.29
7DDR												
Week 1	623	0.50	0.51	0.45	0.48	0.58	0.53	0.53	0.55	0.55	0.54	0.60
Week 2	623	0.44	0.47	0.47	0.46	0.52	0.53	0.52	0.52	0.51	0.53	0.57
Week 1 & 2	624	0.50	0.52	0.49	0.50	0.58	0.56	0.56	0.56	0.56	0.56	0.62
ASA24												
Day 1	462	0.30	0.27	0.24	0.28	0.32	0.29	0.30	0.31	0.32	0.28	0.33
Day 2	454	0.27	0.36	0.27	0.30	0.36	0.32	0.30	0.37	0.33	0.34	0.36
Day 3	452	0.32	0.33	0.38	0.34	0.36	0.42	0.40	0.38	0.38	0.41	0.43
Day 4	452	0.33	0.28	0.29	0.33	0.35	0.35	0.35	0.34	0.37	0.32	0.38
Day 1 & 2	564	0.33	0.34	0.27	0.31	0.38	0.32	0.33	0.37	0.36	0.33	0.37
Day 3 & 4	547	0.36	0.34	0.37	0.36	0.39	0.42	0.42	0.39	0.41	0.41	0.44
Day 1 & 3	578	0.33	0.32	0.33	0.33	0.37	0.37	0.37	0.37	0.37	0.37	0.40
Day 2 & 4	564	0.33	0.34	0.30	0.36	0.37	0.36	0.34	0.39	0.39	0.34	0.40
Day 1 & 4	577	0.34	0.31	0.27	0.33	0.36	0.33	0.33	0.36	0.38	0.32	0.38
Day 2 & 3	559	0.32	0.36	0.35	0.35	0.38	0.40	0.37	0.40	0.38	0.40	0.42
Days, all	624	0.37	0.37	0.34	0.38	0.42	0.41	0.40	0.42	0.42	0.39	0.44

Supplementary Table 2.2f: Spearman correlation coefficients for potassium density (using concurrent DLW assessment) estimated from FFQs, 7DDRs, ASA24s and biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

		Urinary biomarkers for potassium (mg/1000 kcal)										
Methods	**N**	**Day 1**	**Day 2**	**Day 3**	**Day 4**	**Day 1 & 2**	**Day 3 & 4**	**Day 1 & 3**	**Day 2 & 4**	**Day 1 & 4**	**Day 2 & 3**	**Days, all**
N		157	157	176	187	314	363	307	344	302	333	609
SFFQ2	624	0.40	0.41	0.23	0.39	0.40	0.31	0.32	0.40	0.38	0.32	0.35
SFFQ1	624	0.39	0.38	0.27	0.25	0.37	0.26	0.33	0.31	0.30	0.32	0.31
SFFQ1&2	624	0.42	0.43	0.28	0.34	0.41	0.31	0.35	0.38	0.36	0.35	0.36
WebFFQ	624	0.31	0.40	0.26	0.33	0.35	0.30	0.30	0.36	0.30	0.33	0.32
7DDR												
Week 1	623	0.48	0.53	0.50	0.36	0.50	0.43	0.49	0.44	0.41	0.51	0.46
Week 2	623	0.37	0.43	0.44	0.40	0.40	0.42	0.42	0.42	0.38	0.44	0.41
Week 1 & 2	624	0.45	0.51	0.51	0.40	0.48	0.45	0.49	0.45	0.42	0.50	0.47
ASA24												
Day 1	462	0.34	0.21	0.40	0.27	0.28	0.33	0.36	0.25	0.29	0.30	0.29
Day 2	454	0.39	0.38	0.27	0.23	0.38	0.25	0.34	0.30	0.31	0.33	0.32
Day 3	452	0.26	0.47	0.31	0.35	0.36	0.33	0.29	0.41	0.29	0.39	0.34
Day 4	452	0.29	0.32	0.14	0.27	0.30	0.19	0.20	0.29	0.29	0.23	0.25
Day 1 & 2	564	0.42	0.33	0.38	0.21	0.37	0.29	0.40	0.26	0.29	0.36	0.32
Day 3 & 4	547	0.33	0.45	0.25	0.35	0.39	0.30	0.29	0.40	0.33	0.35	0.34
Day 1 & 3	578	0.39	0.35	0.39	0.35	0.37	0.36	0.39	0.35	0.35	0.37	0.36
Day 2 & 4	564	0.41	0.42	0.23	0.29	0.41	0.26	0.31	0.35	0.34	0.32	0.33
Day 1 & 4	577	0.40	0.29	0.29	0.34	0.34	0.31	0.34	0.32	0.35	0.29	0.32
Day 2 & 3	559	0.33	0.50	0.32	0.29	0.42	0.30	0.33	0.39	0.30	0.41	0.36
Days, all	624	0.44	0.44	0.38	0.31	0.44	0.34	0.41	0.37	0.36	0.41	0.38

Supplementary Table 2.3: Mean daily nutrient intakes and reproducibility for energy, protein, sodium and potassium estimated by recovery biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012)

Methods	Absolute level										Energy-adjusted level §					
	Total Energy (kcal)			Protein (g/day)			Sodium (mg/day)		Potassium (mg/day)		Protein (% energy)		Sodium (mg/1000 kcal)		Potassium (mg/1000 kcal)	
	N	Mean	(SD)	N	Mean	(SD)	Mean	(SD)	Mean	(SD)	Mean	(SD)	Mean	(SD)	Mean	(SD)
Biomarker																
Day 1 M1	164	2111	(338)	607	80.2	(20.8)	3546	(1319)	3068	(1047)	14.8	(3.7)	1629	(581)	1426	(509)
Day 2 M1	162	2232	(363)	600	80.8	(24.7)	3486	(1484)	3064	(1061)	14.9	(4.1)	1598	(642)	1420	(494)
Day 3 M1	153	2254	(380)	605	78.8	(22.1)	3220	(1319)	3021	(1052)	14.5	(3.5)	1477	(574)	1405	(505)
Day 4 M1	145	2183	(381)	609	79.2	(24.2)	3298	(1285)	3011	(1120)	14.5	(4.8)	1509	(546)	1396	(524)
M1, all	624	2194	(369)	624	79.8	(18.9)	3388	(1004)	3043	(866)	14.7	(3.2)	1553	(412)	1412	(415)
ICC*	0.71			0.56			0.37		0.49		0.51		0.30		0.52	

§Nutrient density was calculated using the first DLW measurement (randomly assessed every three months over one-year period).
* ICC: rank intraclass correlation

Supplementary Table 2.4: Spearman Correlation coefficients for protein, sodium, potassium intakes estimated from FFQs, 7DDRs, ASA24s and biomarkers, absolute intake and nutrient density using the first DLW measurements (randomly assessed every three months over one-year period) (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient (N=624)	Biomarker (Reference 1)					7DDR (Reference 2)				
	Absolute intake	Nutrient density	De-attenuated (ρ & 95%CI)		ICC	Absolute intake	Nutrient density	De-attenuated (ρ & 95%CI)		ICC
Protein										
SFFQ2	0.25	0.34	0.39	(0.31, 0.45)	0.51	0.32	0.48	0.55	(0.47, 0.61)	0.60
SFFQ1	0.29	0.36	0.39	(0.32, 0.46)	0.51	0.36	0.48	0.54	(0.47, 0.61)	0.60
SFFQ1&2	0.29	0.38	0.43	(0.36, 0.50)	0.51	0.37	0.53	0.60	(0.53, 0.66)	0.60
WebFFQ	0.26	0.33	0.37	(0.29, 0.44)	0.51	0.34	0.47	0.54	(0.46, 0.60)	0.60
ASA24*	0.39	0.36	0.43	(0.36, 0.49)	0.51	0.47	0.52	0.60	(0.53, 0.66)	0.60
7DDR	0.65	0.57	0.62	(0.55, 0.67)	0.51					
Sodium										
SFFQ2	0.16	0.32	0.40	(0.31, 0.48)	0.30	0.28	0.45	0.56	(0.47, 0.63)	0.50
SFFQ1	0.18	0.29	0.36	(0.27, 0.44)	0.30	0.31	0.42	0.50	(0.42, 0.58)	0.50
SFFQ1&2	0.18	0.32	0.40	(0.32, 0.48)	0.30	0.32	0.47	0.58	(0.50, 0.65)	0.50
WebFFQ	0.15	0.28	0.36	(0.27, 0.44)	0.30	0.27	0.44	0.53	(0.44, 0.60)	0.50
ASA24*	0.29	0.26	0.33	(0.24, 0.41)	0.30	0.45	0.41	0.50	(0.41, 0.57)	0.50
7DDR	0.48	0.47	0.59	(0.51, 0.66)	0.30					
Potassium										
SFFQ2	0.32	0.50	0.52	(0.45, 0.58)	0.52	0.45	0.57	0.63	(0.56, 0.68)	0.70
SFFQ1	0.33	0.42	0.45	(0.38, 0.52)	0.52	0.48	0.52	0.56	(0.49, 0.61)	0.70
SFFQ1&2	0.34	0.49	0.53	(0.45, 0.59)	0.52	0.50	0.59	0.63	(0.57, 0.68)	0.70
WebFFQ	0.32	0.42	0.45	(0.37, 0.51)	0.52	0.42	0.50	0.56	(0.49, 0.62)	0.70
ASA24*	0.48	0.49	0.54	(0.47, 0.60)	0.52	0.60	0.62	0.67	(0.61, 0.72)	0.70
7DDR	0.65	0.62	0.68	(0.62, 0.73)	0.52					

*ASA24: averaged ASA24 (mean=2.9 days).

Supplementary Table 2.5: Validity coefficients for protein, sodium, and potassium intakes estimated from FFQs, 7DDRs, ASA24s and four repeated biomarkers, unadjusted and energy adjusted using the first DLW measurement (randomly assessed every three months over one-year period) (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient (N=624)	Validity coefficient (Method of triads)								
	Surrogate vs true			Biomarker vs true			7DDR vs true		
	Absolute intake	Nutrient density	De-attenuated*	Absolute intake	Nutrient density	De-attenuated*	Absolute intake	Nutrient density	De-attenuated*
Surrogate									
Protein									
SFFQ2	0.35	0.54	0.59	0.71	0.64	0.66	0.91	0.90	0.94
SFFQ1	0.40	0.55	0.58	0.72	0.66	0.67	0.90	0.87	0.93
SFFQ1&2	0.41	0.59	0.65	0.71	0.64	0.67	0.91	0.89	0.93
WebFFQ	0.37	0.52	0.57	0.71	0.63	0.65	0.92	0.90	0.95
ASA24**	0.53	0.57	0.65	0.73	0.63	0.67	0.89	0.91	0.93
Sodium									
SFFQ2	0.31	0.55	0.62	0.52	0.58	0.65	0.92	0.81	0.91
SFFQ1	0.34	0.51	0.55	0.53	0.57	0.65	0.91	0.83	0.91
SFFQ1&2	0.35	0.57	0.63	0.52	0.57	0.64	0.92	0.83	0.92
WebFFQ	0.29	0.51	0.57	0.52	0.55	0.63	0.93	0.86	0.93
ASA24**	0.52	0.48	0.53	0.56	0.55	0.62	0.86	0.86	0.95
Potassium									
SFFQ2	0.47	0.68	0.69	0.68	0.74	0.75	0.96	0.84	0.91
SFFQ1	0.49	0.59	0.61	0.67	0.71	0.74	0.97	0.88	0.92
SFFQ1&2	0.51	0.68	0.70	0.66	0.72	0.76	0.98	0.86	0.90
WebFFQ	0.45	0.58	0.61	0.70	0.72	0.74	0.92	0.86	0.92
ASA24**	0.67	0.70	0.73	0.72	0.70	0.74	0.90	0.89	0.92

* This de-attenuated validity coefficient was calculated based on the correlation coefficient de-attenuated for the variations in each reference method (biomarkers and 7DDR), as shown in Supplementary Table 2.4. **ASA24: averaged ASA24 (mean=2.9 days).

Supplementary Table 2.6a: Spearman correlation coefficients for protein density (using first DLW assessment) estimated from FFQs, 7DDRs, ASA24s and biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

		Urinary biomarkers for protein (% energy)										
Methods	**N**	**Day 1**	**Day 2**	**Day 3**	**Day 4**	**Day 1 & 2**	**Day 3 & 4**	**Day 1 & 3**	**Day 2 & 4**	**Day 1 & 4**	**Day 2 & 3**	**Days, all**
N		607	600	605	609	622	624	624	624	623	623	624
SFFQ2	624	0.28	0.24	0.24	0.31	0.30	0.31	0.30	0.32	0.35	0.27	0.34
SFFQ1	624	0.30	0.26	0.22	0.34	0.33	0.32	0.30	0.35	0.38	0.27	0.36
SFFQ1&2	624	0.32	0.28	0.25	0.35	0.35	0.34	0.33	0.36	0.40	0.29	0.38
WebFFQ	624	0.26	0.24	0.25	0.30	0.29	0.30	0.30	0.31	0.33	0.27	0.33
7DDR												
Week 1	623	0.42	0.41	0.40	0.41	0.46	0.45	0.47	0.46	0.47	0.46	0.50
Week 2	623	0.40	0.40	0.42	0.43	0.45	0.47	0.46	0.47	0.46	0.45	0.50
Week 1 & 2	624	0.44	0.45	0.45	0.46	0.50	0.51	0.51	0.51	0.52	0.50	0.56
ASA24												
Day 1	462	0.21	0.24	0.26	0.24	0.26	0.28	0.28	0.27	0.27	0.29	0.30
Day 2	454	0.25	0.29	0.25	0.23	0.30	0.26	0.28	0.29	0.28	0.30	0.31
Day 3	452	0.17	0.24	0.28	0.25	0.24	0.30	0.26	0.30	0.25	0.29	0.30
Day 4	452	0.18	0.21	0.19	0.24	0.21	0.23	0.21	0.25	0.24	0.22	0.25
Day 1 & 2	564	0.26	0.29	0.28	0.26	0.30	0.30	0.31	0.30	0.30	0.32	0.33
Day 3 & 4	547	0.19	0.27	0.27	0.29	0.26	0.30	0.26	0.33	0.28	0.30	0.32
Day 1 & 3	578	0.23	0.28	0.32	0.29	0.30	0.33	0.32	0.33	0.30	0.34	0.35
Day 2 & 4	564	0.24	0.27	0.22	0.25	0.28	0.25	0.26	0.29	0.29	0.27	0.30
Day 1 & 4	577	0.22	0.25	0.26	0.26	0.27	0.28	0.28	0.28	0.29	0.28	0.31
Day 2 & 3	559	0.24	0.29	0.29	0.26	0.29	0.30	0.30	0.32	0.29	0.33	0.34
Days, all	624	0.28	0.33	0.32	0.31	0.34	0.34	0.35	0.36	0.35	0.36	0.38

Supplementary Table 2.6b: Spearman correlation coefficients for sodium density (using first DLW assessment) estimated from FFQs, 7DDRs, ASA24s and biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

		Urinary biomarkers for sodium (mg/1000 kcal)										
Methods	**N**	**Day 1**	**Day 2**	**Day 3**	**Day 4**	**Day 1 & 2**	**Day 3 & 4**	**Day 1 & 3**	**Day 2 & 4**	**Day 1 & 4**	**Day 2 & 3**	**Days, all**
N		607	600	605	609	622	624	624	624	623	623	624
SFFQ2	624	0.22	0.20	0.22	0.26	0.25	0.30	0.28	0.29	0.30	0.24	0.32
SFFQ1	624	0.21	0.17	0.20	0.22	0.22	0.26	0.26	0.24	0.27	0.22	0.29
SFFQ1&2	624	0.23	0.19	0.23	0.26	0.25	0.30	0.29	0.28	0.31	0.24	0.33
WebFFQ	624	0.19	0.20	0.20	0.24	0.24	0.27	0.25	0.28	0.26	0.23	0.30
7DDR												
Week 1	623	0.24	0.30	0.26	0.30	0.33	0.35	0.32	0.36	0.34	0.33	0.39
Week 2	623	0.31	0.33	0.27	0.30	0.39	0.35	0.36	0.38	0.36	0.35	0.42
Week 1 & 2	624	0.31	0.37	0.30	0.35	0.42	0.40	0.39	0.43	0.40	0.39	0.47
ASA24												
Day 1	462	0.10	0.16	0.06	0.17	0.14	0.13	0.08	0.19	0.16	0.13	0.16
Day 2	454	0.11	0.19	0.12	0.11	0.19	0.14	0.14	0.18	0.13	0.18	0.18
Day 3	452	0.14	0.18	0.12	0.17	0.21	0.18	0.17	0.20	0.19	0.16	0.22
Day 4	452	0.19	0.22	0.21	0.22	0.23	0.26	0.23	0.25	0.23	0.25	0.27
Day 1 & 2	564	0.13	0.18	0.10	0.18	0.18	0.15	0.13	0.21	0.18	0.16	0.19
Day 3 & 4	547	0.16	0.20	0.16	0.20	0.21	0.23	0.20	0.22	0.21	0.20	0.24
Day 1 & 3	578	0.14	0.21	0.11	0.22	0.21	0.20	0.15	0.25	0.22	0.19	0.23
Day 2 & 4	564	0.17	0.21	0.17	0.19	0.23	0.21	0.21	0.23	0.21	0.22	0.25
Day 1 & 4	577	0.13	0.18	0.14	0.21	0.17	0.21	0.15	0.22	0.19	0.18	0.21
Day 2 & 3	559	0.16	0.20	0.16	0.17	0.23	0.21	0.21	0.22	0.21	0.20	0.24
Days, all	624	0.17	0.22	0.16	0.22	0.23	0.23	0.20	0.25	0.24	0.21	0.26

Supplementary Table 2.6c: Spearman correlation coefficients for potassium density (using first DLW assessment) estimated from FFQs, 7DDRs, ASA24s and biomarkers (data provided by 624 U.S. female nurses aged 45-80 years, 2010-2012).

		Urinary biomarkers for potassium (mg/1000 kcal)										
Methods	**N**	**Day 1**	**Day 2**	**Day 3**	**Day 4**	**Day 1 & 2**	**Day 3 & 4**	**Day 1 & 3**	**Day 2 & 4**	**Day 1 & 4**	**Day 2 & 3**	**Days, all**
N		607	600	605	609	622	624	624	624	623	623	624
SFFQ2	624	0.38	0.35	0.38	0.37	0.41	0.43	0.43	0.41	0.43	0.42	0.46
SFFQ1	624	0.33	0.32	0.33	0.35	0.37	0.39	0.38	0.39	0.40	0.38	0.43
SFFQ1&2	624	0.38	0.36	0.38	0.39	0.42	0.45	0.44	0.43	0.45	0.44	0.48
WebFFQ	624	0.33	0.33	0.35	0.34	0.36	0.39	0.38	0.38	0.37	0.39	0.41
7DDR												
Week 1	623	0.49	0.48	0.48	0.46	0.55	0.54	0.56	0.52	0.54	0.55	0.60
Week 2	623	0.47	0.43	0.45	0.44	0.51	0.51	0.52	0.48	0.51	0.51	0.55
Week 1 & 2	624	0.52	0.49	0.50	0.48	0.57	0.56	0.58	0.54	0.56	0.57	0.62
ASA24												
Day 1	462	0.30	0.28	0.30	0.31	0.33	0.34	0.34	0.32	0.34	0.33	0.37
Day 2	454	0.35	0.38	0.37	0.32	0.41	0.39	0.41	0.40	0.40	0.42	0.44
Day 3	452	0.30	0.29	0.34	0.33	0.33	0.38	0.36	0.34	0.35	0.36	0.39
Day 4	452	0.30	0.31	0.26	0.36	0.35	0.35	0.32	0.37	0.38	0.32	0.38
Day 1 & 2	564	0.36	0.35	0.36	0.33	0.40	0.39	0.41	0.38	0.39	0.41	0.44
Day 3 & 4	547	0.34	0.35	0.34	0.38	0.39	0.42	0.39	0.41	0.42	0.40	0.45
Day 1 & 3	578	0.33	0.32	0.35	0.35	0.37	0.40	0.39	0.38	0.39	0.39	0.42
Day 2 & 4	564	0.37	0.40	0.37	0.39	0.44	0.42	0.42	0.44	0.44	0.43	0.47
Day 1 & 4	577	0.34	0.32	0.32	0.38	0.38	0.39	0.38	0.38	0.41	0.36	0.42
Day 2 & 3	559	0.36	0.36	0.39	0.35	0.40	0.42	0.42	0.40	0.40	0.43	0.45
Days, all	624	0.41	0.40	0.41	0.40	0.46	0.46	0.46	0.45	0.47	0.47	0.51

Supplementary Table 3.1: Spearman correlation coefficients for specific fatty acid intakes estimated from SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient	Biomarker (reference 1)				7DDR (reference 2)			
	Un-adjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC	Un-adjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC
Saturated fatty acids								
SFFQ2	0.09	0.08	0.15 (0.00, 0.29)	0.20	0.53	0.53	0.62 (0.54, 0.67)	0.58
Single ASA24	0.04	0.03	0.06 (-0.12, 0.24)	0.20	0.33	0.32	0.37 (0.28, 0.46)	0.58
Averaged ASA24	0.07	0.05	0.10 (-0.05, 0.24)	0.20	0.43	0.43	0.50 (0.42, 0.57)	0.58
Averaged 7DDR	0.18	0.17	0.33 (0.17, 0.47)	0.20				
Monounsaturated fatty acids								
SFFQ2	0.04	0.07	0.08 (-0.01, 0.17)	0.64	0.39	0.39	0.51 (0.42, 0.59)	0.41
Single ASA24	0.02	0.04	0.04 (-0.06, 0.15)	0.64	0.16	0.17	0.22 (0.10, 0.33)	0.41
Averaged ASA24	0.00	0.05	0.05 (-0.04, 0.14)	0.64	0.24	0.26	0.33 (0.24, 0.42)	0.41
Averaged 7DDR	0.04	0.04	0.04 (-0.05, 0.12)	0.64				
Alpha-linolenic acid (18:3n-3c) **								
SFFQ2	0.28	0.28	0.34 (0.25, 0.42)	0.57	0.45	0.45	0.55 (0.47, 0.63)	0.47
Single ASA24	0.18	0.17	0.21 (0.10, 0.31)	0.57	0.27	0.25	0.31 (0.21, 0.41)	0.47
Averaged ASA24	0.22	0.22	0.27 (0.18, 0.36)	0.57	0.37	0.36	0.44 (0.36, 0.52)	0.47
Averaged 7DDR	0.39	0.39	0.48 (0.39, 0.56)	0.57				
Long Chain N-3 (DHA+DPA+EPA)								
SFFQ2	0.57	0.56	0.62 (0.56, 0.68)	0.75	0.62	0.61	0.71 (0.65, 0.77)	0.59
Averaged 7DDR	0.67	0.66	0.74 (0.68, 0.79)	0.75				
Long Chain N-3* (N=363)**								
SFFQ2	0.57	0.53	0.58 (0.47, 0.66)	0.61	0.56	0.55	0.67 (0.56, 0.74)	0.51
Single ASA24	0.19	0.19	0.23 (0.09, 0.36)	0.61	0.23	0.22	0.26 (0.13, 0.39)	0.51
Averaged ASA24	0.31	0.30	0.36 (0.25, 0.46)	0.61	0.38	0.31	0.37 (0.25, 0.47)	0.51
Averaged 7DDR	0.55	0.52	0.61 (0.51, 0.69)	0.61				

Supplementary Table 3.1 (continued): Spearman correlation coefficients for specific fatty acid intakes estimated from SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient	Biomarker (reference 1)				7DDR (reference 2)			
	Un-adjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC	Un-adjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC
Linoleic acid (18:2n-6cc)								
SFFQ2	0.16	0.20	0.25 (0.16, 0.34)	0.53	0.38	0.40	0.50 (0.41, 0.58)	0.44
Single ASA24	0.06	0.06	0.08 (-0.04, 0.19)	0.53	0.26	0.26	0.33 (0.21, 0.42)	0.44
Averaged ASA24	0.10	0.10	0.13 (0.03, 0.22)	0.53	0.35	0.35	0.45 (0.36, 0.53)	0.44
Averaged 7DDR	0.25	0.23	0.29 (0.20, 0.38)	0.53				
Polyunsaturated fatty acids								
SFFQ2	0.16	0.18	0.27 (0.15, 0.37)	0.38	0.44	0.45	0.56 (0.48, 0.64)	0.46
Single ASA24	0.08	0.06	0.09 (-0.04, 0.22)	0.38	0.27	0.27	0.33 (0.22, 0.43)	0.46
Averaged ASA24	0.11	0.09	0.14 (0.03, 0.24)	0.38	0.37	0.37	0.46 (0.37, 0.54)	0.46
Averaged 7DDR	0.24	0.22	0.32 (0.21, 0.42)	0.38				
Trans Fatty acids								
SFFQ2	0.29	0.27	0.33 (0.16, 0.48)	0.21	0.41	0.41	0.53 (0.44, 0.61)	0.40
Averaged 7DDR	0.33	0.30	0.53 (0.36, 0.66)	0.21				

* Nutrients were adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing;
**Linolenic acid from 7DDR and ASA24 included alpha- linolenic acid and gama- linolenic acid;
***Subgroups of women who didn't take supplements for this nutrient.

Supplementary Table 3.2: Validity coefficients for fatty acids estimated from SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient	Validity coefficient (Method of triads)								
	Surrogate vs true			Averaged biomarker vs true			Averaged 7DDR vs true		
	Un-adjusted	Adjusted[a]	De-attenuated[b]	Un-adjusted	Adjusted[a]	De-attenuated[b]	Un-adjusted	Adjusted[a]	De-attenuated[b]
Surrogate									
Saturated fatty acids									
SFFQ2	0.51	0.50	0.53	0.17	0.16	0.28	1.00[d]	1.00[d]	1.00[d]
Single ASA24	0.27	0.24	0.26	0.15	0.13	0.23	1.00[d]	1.00[d]	1.00[d]
Averaged ASA24	0.41	0.36	0.39	0.17	0.14	0.26	1.00[d]	1.00[d]	1.00[d]
Monounsaturated fatty acids									
SFFQ2	0.62	0.83	1.00[d]	0.06	0.08	0.08	0.62	0.47	0.50
Single ASA24	0.28	0.41	0.47	0.07	0.10	0.09	0.57	0.41	0.47
Averaged ASA24	0.00	0.57	0.64	0.00	0.09	0.08	N/A	0.46	0.51
Alpha-linolenic acid (18:3n-3c)									
SFFQ2	0.57	0.57	0.62	0.49	0.49	0.54	0.79	0.79	0.88
Single ASA24	0.35	0.33	0.37	0.51	0.51	0.57	0.76	0.76	0.84
Averaged ASA24	0.46	0.45	0.50	0.48	0.49	0.54	0.81	0.80	0.88
Long Chain N-3 (DHA+DPA+EPA)									
SFFQ2	0.73	0.72	0.77	0.78	0.78	0.80	0.85	0.85	0.92
Long Chain N-3[c] (N=363)									
SFFQ2	0.76	0.75	0.80	0.75	0.71	0.73	0.74	0.73	0.84
Single ASA24	0.28	0.28	0.31	0.67	0.67	0.73	0.82	0.78	0.83
Averaged ASA24	0.46	0.42	0.47	0.67	0.71	0.77	0.82	0.73	0.79
Linoleic acid (18:2n-6cc)									
SFFQ2	0.49	0.59	0.66	0.32	0.34	0.38	0.77	0.68	0.76
Single ASA24	0.25	0.26	0.30	0.24	0.23	0.27	1.00[d]	1.00[d]	1.00[d]
Averaged ASA24	0.37	0.39	0.45	0.27	0.26	0.29	0.94	0.90	1.00[d]

Supplementary Table 3.2 (continued): Validity coefficients for fatty acids estimated from SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient	Validity coefficient (Method of triads)								
	Surrogate vs true			Averaged biomarker vs true			Averaged 7DDR vs true		
	Un-adjusted	Adjusted[a]	De-attenuated[b]	Un-adjusted	Adjusted[a]	De-attenuated[b]	Un-adjusted	Adjusted[a]	De-attenuated[b]
Polyunsaturated fatty acids									
SFFQ2	0.54	0.61	0.69	0.30	0.30	0.39	0.81	0.74	0.81
Single ASA24	0.30	0.27	0.30	0.27	0.22	0.30	0.90	0.99	1.00[d]
Averaged ASA24	0.41	0.39	0.45	0.27	0.23	0.31	0.90	0.95	1.00[d]
Trans Fatty acids									
SFFQ2	0.60	0.61	0.57	0.48	0.44	0.57	0.68	0.67	0.92

a. Nutrients adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing;
b. This de-attenuated validity coefficient was calculated based on the correlation de-attenuated for the variations in each reference method (biomarkers and 7DDR), as shown in supplementary Table 3.1.
c. Subgroups of women who didn't take supplements for this nutrient.
d. Validity coefficients >1 (Heywood cases) were set to 1.00.

Supplementary Table 3.3: Spearman correlation coefficients for carotenoids, retinol, tocopherols, and folate intakes estimated from SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient	Biomarker (reference 1)				7DDR (reference 2)			
	Unadjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC	Unadjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC
Alpha carotene								
SFFQ2	0.47	0.47	0.51 (0.44, 0.57)	0.78	0.50	0.47	0.65 (0.54, 0.73)	0.36
Single ASA24	0.29	0.25	0.27 (0.18, 0.36)	0.78	0.25	0.21	0.29 (0.17, 0.40)	0.36
Averaged ASA24	0.37	0.34	0.37 (0.30, 0.44)	0.78	0.31	0.26	0.36 (0.26, 0.45)	0.36
Averaged 7DDR	0.55	0.55	0.60 (0.53, 0.65)	0.78				
Beta carotene								
SFFQ2	0.39	0.40	0.41 (0.35, 0.48)	0.81	0.50	0.48	0.62 (0.53, 0.69)	0.44
Averaged 7DDR	0.52	0.53	0.56 (0.50, 0.62)	0.81				
Beta carotene (N=335)**								
SFFQ2	0.46	0.45	0.47 (0.37, 0.56)	0.78	0.59	0.60	0.74 (0.63, 0.82)	0.48
Single ASA24	0.31	0.22	0.24 (0.11, 0.36)	0.78	0.38	0.31	0.38 (0.24, 0.50)	0.48
Averaged ASA24	0.42	0.33	0.36 (0.25, 0.45)	0.78	0.47	0.41	0.51 (0.39, 0.61)	0.48
Averaged 7DDR	0.54	0.53	0.58 (0.49, 0.66)	0.78				
Lutein-zeaxanthin								
SFFQ2	0.33	0.32	0.35 (0.28, 0.42)	0.75	0.51	0.52	0.63 (0.55, 0.69)	0.51
Single ASA24	0.27	0.23	0.25 (0.16, 0.34)	0.75	0.33	0.30	0.36 (0.26, 0.45)	0.51
Averaged ASA24	0.33	0.29	0.32 (0.24, 0.39)	0.75	0.44	0.41	0.50 (0.42, 0.57)	0.51
Averaged 7DDR	0.46	0.45	0.48 (0.41, 0.55)	0.75				
Beta cryptoxanthin								
SFFQ2	0.40	0.41	0.46 (0.38, 0.53)	0.71	0.36	0.35	0.51 (0.39, 0.61)	0.29
Single ASA24	0.32	0.29	0.33 (0.24, 0.42)	0.71	0.30	0.25	0.36 (0.23, 0.48)	0.29
Averaged ASA24	0.42	0.40	0.45 (0.37, 0.52)	0.71	0.40	0.34	0.51 (0.39, 0.61)	0.29
Averaged 7DDR	0.50	0.47	0.53 (0.46, 0.60)	0.71				

Supplementary Table 3.3 (continued): Spearman correlation coefficients for carotenoids, retinol, tocopherols, and folate intakes estimated from SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient	Biomarker (reference 1)				7DDR (reference 2)			
	Unadjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC	Unadjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC
Lycopene								
SFFQ2	0.26	0.28	0.33 (0.24, 0.41)	0.56	0.40	0.37	0.62 (0.46, 0.73)	0.22
Single ASA24	0.17	0.14	0.16 (0.06, 0.28)	0.56	0.17	0.14	0.22 (0.07, 0.36)	0.22
Averaged ASA24	0.22	0.20	0.25 (0.15, 0.34)	0.56	0.24	0.20	0.32 (0.19, 0.44)	0.22
Averaged 7DDR	0.35	0.40	0.47 (0.38, 0.55)	0.56				
Retinol activity equivalents								
SFFQ2	0.14	0.09	0.10 (0.01, 0.18)	0.64	0.64	0.65	0.74 (0.68, 0.79)	0.64
Averaged 7DDR	0.18	0.13	0.15 (0.07, 0.24)	0.64				
Retinol activity equivalents (N=207)**								
SFFQ2	0.03	0.13	0.17 (-0.01, 0.34)	0.48	0.54	0.58	0.83 (0.55, 0.93)	0.32
Single ASA24	0.02	0.03	0.04 (-0.16, 0.23)	0.48	0.35	0.29	0.41 (0.18, 0.59)	0.32
Averaged ASA24	0.03	0.02	0.03 (-0.14, 0.20)	0.48	0.42	0.39	0.55 (0.35, 0.70)	0.32
Averaged 7DDR	0.10	0.15	0.20 (0.03, 0.37)	0.48				
Alpha-tocopherol								
SFFQ2	0.49	0.52	0.65 (0.57, 0.72)	0.53	0.72	0.73	0.79 (0.74, 0.83)	0.77
Averaged 7DDR	0.57	0.64	0.79 (0.71, 0.85)	0.53				
Alpha-tocopherol ** (N=145)								
SFFQ2	0.10	0.10	0.14 (-0.08, 0.34)	0.44	0.40	0.33	0.42 (0.22, 0.58)	0.56
Single ASA24	0.09	0.09	0.11 (-0.17, 0.37)	0.44	0.31	0.26	0.34 (0.09, 0.54)	0.56
Averaged ASA24	0.13	0.16	0.15 (-0.08, 0.37)	0.44	0.41	0.38	0.48 (0.29, 0.64)	
Averaged 7DDR	0.14	0.33	0.44 (0.22, 0.62)	0.44				

Supplementary Table 3.3 (continued): Spearman correlation coefficients for carotenoids, retinol, tocopherols, and folate intakes estimated from SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient	Biomarker (reference 1)				7DDR (reference 2)			
	Unadjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC	Unadjusted	Adjusted*	De-attenuated (ρ & 95%CI)	ICC
Gama-tocopherol								
SFFQ2	0.00	0.02	0.02 (-0.07, 0.11)	0.67	0.24	0.38	0.50 (0.40, 0.58)	0.38
Averaged 7DDR	0.12	0.12	0.13 (0.05, 0.22)	0.67				
Dietary folate equivalents								
SFFQ2	0.47	0.49	0.54 (0.46, 0.61)	0.68	0.69	0.72	0.78 (0.72, 0.82)	0.77
Averaged 7DDR	0.61	0.61	0.68 (0.60, 0.73)	0.68				
Dietary folate equivalents ** (N=135)								
SFFQ2	0.38	0.40	0.44 (0.28, 0.58)	0.69	0.56	0.62	0.71 (0.56, 0.81)	0.61
Single ASA24	0.31	0.30	0.33 (0.14, 0.49)	0.69	0.39	0.37	0.43 (0.23, 0.59)	0.61
Averaged ASA24	0.42	0.40	0.44 (0.28, 0.58)	0.69	0.56	0.52	0.59 (0.43, 0.71)	0.61
Averaged 7DDR	0.60	0.50	0.55 (0.39, 0.67)	0.69				

* Nutrients were adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; self-reported nutrients were further adjusted for total energy intake; plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing; Plasma carotenoids, retinol and was further adjusted for plasma lipids.

**Subgroups of women who didn't take supplements for this nutrient.

Supplementary Table 3.4: Validity coefficients for fatty acids estimated from SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient	Validity coefficient (Method of triads)								
	Surrogate vs true			Biomarker vs true			7DDR vs true		
	Un-adjusted	Adjusted[a]	De-attenuated[b]	Un-adjusted	Adjusted[a]	De-attenuated[b]	Un-adjusted	Adjusted[a]	De-attenuated[b]
Surrogate									
Alpha carotene									
SFFQ2	0.65	0.63	0.74	0.72	0.74	0.69	0.76	0.74	0.87
Single ASA24	0.36	0.31	0.36	0.80	0.81	0.75	0.69	0.68	0.80
Averaged ASA24	0.46	0.40	0.47	0.81	0.85	0.79	0.68	0.65	0.76
Beta carotene									
SFFQ2	0.61	0.60	0.67	0.64	0.66	0.61	0.82	0.80	0.92
Beta carotene[c] (N=335)									
SFFQ2	0.71	0.71	0.77	0.65	0.63	0.61	0.83	0.84	0.96
Single ASA24	0.47	0.36	0.40	0.66	0.61	0.61	0.81	0.86	0.96
Averaged ASA24	0.60	0.51	0.56	0.69	0.65	0.64	0.78	0.81	0.91
Lutein-zeaxanthin									
SFFQ2	0.60	0.61	0.68	0.55	0.53	0.52	0.84	0.86	0.93
Single ASA24	0.44	0.39	0.43	0.61	0.59	0.58	0.75	0.77	0.83
Averaged ASA24	0.56	0.51	0.58	0.59	0.56	0.55	0.78	0.80	0.87
Beta cryptoxanthin									
SFFQ2	0.54	0.55	0.67	0.75	0.74	0.69	0.67	0.63	0.77
Single ASA24	0.44	0.39	0.47	0.73	0.74	0.70	0.68	0.64	0.76
Averaged ASA24	0.58	0.54	0.66	0.72	0.74	0.68	0.69	0.63	0.78
Lycopene									
SFFQ2	0.55	0.51	0.66	0.48	0.55	0.50	0.73	0.73	0.94
Single ASA24	0.29	0.22	0.27	0.59	0.63	0.58	0.59	0.63	0.80
Averaged ASA24	0.39	0.32	0.41	0.57	0.63	0.61	0.62	0.63	0.78

Supplementary Table 3.4 (continued): Validity coefficients for fatty acids estimated from SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Nutrient	Validity coefficient (Method of triads)								
	Surrogate vs true			Biomarker vs true			7DDR vs true		
	Un-adjusted	Adjusted[a]	De-attenuated[b]	Un-adjusted	Adjusted[a]	De-attenuated[b]	Un-adjusted	Adjusted[a]	De-attenuated[b]
Retinol activity equivalents									
SFFQ2	0.71	0.67	0.70	0.20	0.13	0.14	0.91	0.97	1.00[d]
Retinol activity equivalents[c] (N=237)									
SFFQ2	0.40	0.71	0.84	0.07	0.18	0.20	1.00[d]	1.00[d]	0.99
Single ASA24	0.26	0.24	0.29	0.08	0.12	0.14	1.00[d]	1.00[d]	1.00[d]
Averaged ASA24	0.35	0.23	0.29	0.08	0.09	0.10	1.00[d]	1.00[d]	1.00[d]
Alpha-tocopherol									
SFFQ2	0.79	0.77	0.81	0.62	0.68	0.81	0.92	0.95	0.98
Alpha-tocopherol[c] (N=145)									
SFFQ2	0.53	0.32	0.37	0.19	0.32	0.38	0.75	1.00[d]	1.00[d]
Single ASA24	0.45	0.27	0.29	0.20	0.34	0.38	0.69	0.98	1.00[d]
Averaged ASA24	0.62	0.43	0.40	0.21	0.37	0.37	0.66	0.89	1.00[d]
Gama - tocopherol									
SFFQ2	0.00	0.25	0.28	0.00	0.08	0.07	N/A	1.00[d]	1.00[d]
Dietary folate equivalents									
SFFQ2	0.73	0.76	0.79	0.64	0.64	0.69	0.95	0.95	0.99
Dietary folate equivalents[c] (N=135)									
SFFQ2	0.60	0.70	0.75	0.64	0.57	0.58	0.94	0.88	0.94
Single ASA24	0.45	0.47	0.51	0.69	0.64	0.65	0.87	0.79	0.85
Averaged ASA24	0.63	0.64	0.69	0.67	0.62	0.64	0.89	0.81	0.86

a. Nutrients were adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; self-reported nutrients were further adjusted for total energy intake; plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing; Plasma carotenoids, retinol and was further adjusted for plasma lipids. b. This de-attenuated validity coefficient was calculated based on the correlation de-attenuated for the variations in each reference method (biomarkers and 7DDR), as shown in supplementary Table 3.3.
c. Subgroups of women who didn't take supplements for this nutrient. d. Validity coefficients >1 (Heywood cases) were set to 1.00.

Supplementary Table 3.5: Distribution of fatty acids (% of total fatty acids) estimated by SFFQ, 7DDR, ASA24, and biochemical indicator (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012)

Fatty acid	Biomarker	SFFQ Sacks	SFFQ original	7DDR	ASA24
	Blood 1&2	FFQ2s	FFQ2	week1&2	days, all
Saturated fatty acids					
12:00	0.02 ± 0.03	1.6 ± 1.4	0.9 ± 0.9	1.5 ± 1.1	1.2 ± 1.1
14:00	0.5 ± 0.2	3.4 ± 1.3	2.5 ± 0.9	3.0 ± 0.9	3.1 ± 1.3
16:00	19.3 ± 2.1	19.7 ± 2.6	17.5 ± 2.1	17.2 ± 1.8	17.6 ± 2.4
18:00	9.4 ± 1.7	8.2 ± 1.9	7.9 ± 1.8	7.9 ± 1.2	8.4 ± 2.1
Total	31.4 ± 2.7	35.6 ± 6.2	31.7 ± 5.5	33.1 ± 4.7	33.8 ± 6.2
MUFA					
16:1n-7c	1.5 ± 0.6	1.5 ± 0.3	1.5 ± 0.3	1.5 ± 0.3	1.5 ± 0.5
18:1n-9c	18.3 ± 2.4	32.4 ± 4.3	35.3 ± 4.6	33.9 ± 3.1	33.3 ± 3.8
Total 18:1*	20.6 ± 2.5	35.7 ± 4.2	35.3 ± 4.6	33.9 ± 3.1	33.3 ± 3.8
20:1n-9c	0.13 ± 0.03	0.3 ± 0.2	0.4 ± 0.2	0.3 ± 0.1	0.28 ± 0.13
Total	22.2 ± 2.8	38.0 ± 4.2	37.6 ± 4.6	36.1 ± 3.1	35.6 ± 3.8
PUFA					
18:3n-3c	0.6 ± 0.2	2.3 ± 1.1	2.2 ± 1.0	2.3 ± 1.0	2.2 ± 1.0
Total 18:3**	1.1 ± 0.3	2.3 ± 1.1	2.2 ± 1.0	2.3 ± 1.0	2.2 ± 1.0
20:5n-3c	1.0 ± 0.8	0.3 ± 0.3	0.3 ± 0.3	0.2 ± 0.4	0.1 ± 0.2
22:5n-3c	0.6 ± 0.2	0.04 ± 0.04	0.05 ± 0.04	0.03 ± 0.04	0.04 ± 0.06
22:6n-3c	1.9 ± 0.8	0.4 ± 0.3	0.4 ± 0.3	0.3 ± 0.3	0.2 ± 0.3
Long Chain N-3***	3.5 ± 1.6	0.8 ± 0.7	0.8 ± 0.7	0.5 ± 0.7	N/A
18:2n-6cc	30.0 ± 3.7	18.1 ± 3.7	17.9 ± 3.8	19.5 ± 3.7	19.0 ± 4.9
18:3n-6c	0.5 ± 0.2	0.02 ± 0.01			
20:2n-6c	0.22 ± 0.04	0.04 ± 0.02			
20:4n-6c	8.2 ± 1.9	0.2 ± 0.1	0.3 ± 0.1	0.2 ± 0.1	0.2 ± 0.1
Total N-6	40.7 ± 3.9	18.5 ± 3.7			
Total PUFA	44.8 ± 4.0	22.2 ± 4.6	21.5 ± 4.6	22.4 ± 4.2	21.9 ± 5.6
Trans Fatty acids					
t-16:1n-7	0.13 ± 0.03	0.11 ± 0.04		0.06 ± 0.03	
Total 18:1 trans	0.8 ± 0.3	2.2 ± 0.6		2.6 ± 1.0	
Total 18:2 trans	0.2 ± 0.1	0.4 ± 0.1		0.5 ± 0.1	
Total trans	1.1 ± 0.4	2.7 ± 0.7		3.3 ± 1.1	
Believed to be trans	0.2 ± 0.1	0.5 ± 0.2			
Dairy fat					
15:00	0.15 ± 0.04		19.0 ± 9.1		
17:00	0.33 ± 0.07		19.0 ± 9.1	0.12 ± 0.05	
t-16:1n-7	0.13 ± 0.03		19.0 ± 9.1		

Note: MUFA: monounsaturated fatty acids; PUFA: polyunsaturated fatty acids
*Total 18:1 was the sum of 18:1n-9c and 18:1n-7c; **Total 18:3 was the sum of alpha-linolenic (18:3n-3c) and grama-linolenic (18:3n-6c). ***Long Chain N-3 fatty acid was the sum of 20:5n-3c, 22:5n-3c and 22:6n-3c.

Supplementary Table 3.6: Spearman correlation coefficient between fatty acids (% of total fatty acids) estimated by SFFQ, 7DDR, ASA24, and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012)

Fatty acid	FFQ2 Sacks vs biomarker			FFQ2 original vs biomarker			Averaged ASA24 vs biomarker			7DDR vs biomarker		
	Un-adjusted	Adjust-ed[a]	De-attenuated	Un-adjusted	Adjust-ed[a]	De-attenuated	Un-adjusted	Adjust-ed[a]	De-attenuated	Un-adjusted	Adjust-ed[a]	De-attenuated
Saturated fatty acids												
12:00	0.05	0.04	0.11	0.05	0.04	0.10	0.02	0.02	0.05	0.17	0.16	0.32
14:00	0.19	0.18	0.29	0.07	0.18	0.30	0.12	0.12	0.18	0.25	0.25	0.40
16:00	0.12	0.13	0.17	0.07	0.09	0.13	0.06	0.03	0.04	0.19	0.19	0.25
18:00	-0.03	-0.03	-0.08	0.04	-0.04	-0.10	0.00	0.02	0.04	-0.03	-0.01	-0.03
Total	0.09	0.08	0.15	0.05	0.08	0.13	0.07	0.05	0.10	0.18	0.17	0.33
Monounsaturated fatty acids												
16:1n-7c	0.13	0.16	0.18	0.15	0.17	0.19	0.09	0.08	0.09	0.16	0.14	0.14
18:1n-9c	0.07	0.10	0.11	0.05	0.08	0.09	0.04	0.08	0.09	0.08	0.08	0.09
Total 18:1*	0.04	0.07	0.08	0.03	0.06	0.07	0.04	0.08	0.09	0.06	0.06	0.07
20:1n-9c	0.01	0.00	0.00	0.04	0.03	0.05	0.01	0.02	0.03	0.04	0.05	0.06
Total	0.04	0.07	0.08	0.04	0.06	0.07	0.00	0.05	0.05	0.04	0.04	0.04
Polyunsaturated fatty acids												
18:3n-3c	0.28	0.28	0.34	0.28	0.27	0.33	0.22	0.22	0.27	0.39	0.39	0.48
Total 18:3**	0.16	0.16	0.18	0.15	0.15	0.18	0.16	0.17	0.22	0.23	0.26	0.33
20:5n-3c	0.55	0.54	0.59	0.55	0.53	0.59	0.15	0.16	0.16	0.65	0.63	0.69
22:5n-3c	0.35	0.33	0.40	0.31	0.31	0.37	0.03	0.00	0.00	0.10	0.08	0.09
22:6n-3c	0.56	0.55	0.60	0.55	0.55	0.61	0.24	0.27	0.28	0.63	0.62	0.69
Long Chain N-3	0.57	0.56	0.62	0.57	0.57	0.63	N/A	N/A	N/A	0.67	0.66	0.74
18:2n-6cc	0.16	0.20	0.25	0.19	0.22	0.28	0.10	0.10	0.13	0.25	0.23	0.29
18:3n-6c	-0.03	-0.01	-0.01									
20:2n-6c	0.00	-0.01	-0.01									
20:4n-6c	0.04	0.04	0.04	0.07	0.08	0.09	0.14	0.13	0.15	0.20	0.21	0.23
Total N-6	0.13	0.16	0.23									
Total PUFA	0.16	0.18	0.27	0.17	0.19	0.27	0.11	0.09	0.14	0.24	0.22	0.32

Supplementary Table 3.6 (continued): Spearman correlation coefficient between fatty acids (% of total fatty acids) estimated by SFFQ, 7DDR, ASA24, and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012)

Fatty acid	FFQ2 Sacks vs biomarker			FFQ2 original vs biomarker			Averaged ASA24 vs biomarker			7DDR vs biomarker		
	Un-adjusted	Adjust-ed[a]	De-attenuated	Un-adjusted	Adjust-ed[a]	De-attenuated	Un-adjusted	Adjust-ed[a]	De-attenuated	Un-adjusted	Adjust-ed[a]	De-attenuated
Trans Fatty acids												
t-16:1n-7	0.29	0.31	0.39	N/A	N/A	N/A	N/A	N/A	N/A	0.26	0.26	0.36
Total 18:1 trans	0.27	0.25	0.31	N/A	N/A	N/A	N/A	N/A	N/A	0.30	0.28	0.52
Total 18:2 trans	0.28	0.27	0.42	N/A	N/A	N/A	N/A	N/A	N/A	0.30	0.29	0.48
Total trans	0.29	0.27	0.33	N/A	N/A	N/A	N/A	N/A	N/A	0.33	0.30	0.53
Believed to be trans	0.36	0.35	0.38	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
Dairy fat												
15:00	N/A	N/A	N/A	0.26	0.25	0.36	N/A	N/A	N/A	N/A	N/A	N/A
17:00	N/A	N/A	N/A	0.06	0.06	0.07	N/A	N/A	N/A	0.04	0.04	0.05
t-16:1n-7	N/A	N/A	N/A	0.29	0.29	0.38	N/A	N/A	N/A	N/A	N/A	N/A

a: Nutrients adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing;

[*] Total 18:1 was the sum of 18:1n-9c and 18:1n-7c;

[**]Total 18:3 was the sum of alpha-linolenic (18:3n-3c) and grama-linolenic (18:3n-6c).

Supplementary Table 3.7: Spearman correlation coefficients for fatty acids estimated from each SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Methods		Saturated fatty acids			Monounsaturated fatty acids			Long Chain N-3*			Linoleic Acid			Trans Fatty acid		
	N	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2
N		615	306	627	615	306	627	356	183	363	615	306	627	615	306	627
SFFQ1	627	0.09	0.05	0.07	0.05	-0.04	0.03	0.51	0.57	0.54	0.12	0.19	0.13	0.29	0.15	0.29
SFFQ2	627	0.09	0.07	0.08	0.06	-0.04	0.04	0.53	0.60	0.56	0.14	0.19	0.15	0.27	0.10	0.27
SFFQ1&2	627	0.09	0.07	0.08	0.05	-0.04	0.03	0.55	0.63	0.58	0.15	0.21	0.16	0.31	0.15	0.31
WebFFQ	627	0.12	0.08	0.12	0.04	-0.02	0.03	0.49	0.52	0.49	0.15	0.18	0.17	0.31	0.10	0.30
7DDR																
Week 1	626	0.12	0.14	0.13	0.01	-0.02	0.01	0.45	0.43	0.46	0.17	0.19	0.18	0.28	0.10	0.26
Week 2	626	0.12	0.21	0.16	0.07	0.00	0.05	0.44	0.53	0.46	0.21	0.26	0.24	0.28	0.21	0.29
Week 1 & 2	627	0.13	0.20	0.16	0.05	0.00	0.04	0.50	0.56	0.52	0.23	0.26	0.25	0.32	0.17	0.31
ASA24																
Day 1	464	0.05	0.02	0.04	-0.03	0.03	-0.02	0.27	0.21	0.26	0.04	0.03	0.06	N/A	N/A	N/A
Day 2	457	0.08	0.15	0.10	0.02	0.05	0.04	0.21	0.21	0.19	0.07	0.14	0.08	N/A	N/A	N/A
Day 3	456	-0.03	0.01	-0.03	0.02	0.07	0.05	0.15	0.29	0.20	0.05	0.10	0.06	N/A	N/A	N/A
Day 4	450	0.02	-0.05	0.01	0.01	-0.01	-0.02	0.08	0.23	0.11	0.06	0.06	0.06	N/A	N/A	N/A
Day 1 & 2	567	0.08	0.09	0.08	-0.01	0.03	0.01	0.32	0.27	0.31	0.05	0.11	0.07	N/A	N/A	N/A
Day 3 & 4	549	-0.01	-0.01	-0.02	0.00	0.00	0.00	0.20	0.35	0.23	0.08	0.11	0.08	N/A	N/A	N/A
Day 1 & 3	581	0.01	0.03	0.01	-0.03	0.04	0.00	0.24	0.32	0.28	0.05	0.08	0.07	N/A	N/A	N/A
Day 2 & 4	565	0.07	0.07	0.08	0.02	0.05	0.02	0.23	0.31	0.22	0.07	0.11	0.08	N/A	N/A	N/A
Day 1 & 4	578	0.06	-0.02	0.04	-0.02	-0.01	-0.03	0.20	0.21	0.19	0.05	0.05	0.05	N/A	N/A	N/A
Day 2 & 3	563	0.04	0.09	0.05	0.00	0.05	0.03	0.24	0.31	0.25	0.08	0.15	0.09	N/A	N/A	N/A
Days, all	627	0.05	0.06	0.05	-0.03	0.01	-0.01	0.29	0.38	0.31	0.07	0.13	0.08	N/A	N/A	N/A

* Subgroups of women who didn't take supplements for this nutrient.

Supplementary Table 3.8: Adjusted* Spearman correlation coefficients for fatty acids estimated from each SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Methods		Saturated fatty acids			Monounsaturated fatty acids			Long Chain N-3**			Linoleic Acid			Trans Fatty acid		
	N	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2
N		614	305	626	614	305	626	355	182	362	614	305	626	614	305	626
SFFQ1	626	0.09	0.01	0.06	0.07	0.00	0.05	0.48	0.52	0.52	0.14	0.17	0.15	0.27	0.11	0.26
SFFQ2	626	0.09	0.03	0.07	0.08	0.00	0.06	0.47	0.58	0.51	0.19	0.21	0.21	0.24	0.09	0.24
SFFQ1&2	626	0.09	0.03	0.07	0.07	-0.01	0.06	0.52	0.58	0.56	0.19	0.21	0.20	0.28	0.12	0.28
WebFFQ	626	0.11	0.05	0.10	0.06	0.00	0.05	0.43	0.48	0.46	0.17	0.16	0.19	0.29	0.08	0.28
7DDR																
Week 1	625	0.13	0.10	0.14	0.01	-0.01	0.02	0.40	0.40	0.43	0.17	0.16	0.18	0.26	0.10	0.25
Week 2	625	0.13	0.19	0.16	0.06	0.02	0.04	0.40	0.52	0.44	0.21	0.23	0.23	0.27	0.19	0.27
Week 1 & 2	626	0.14	0.16	0.16	0.05	0.01	0.05	0.45	0.53	0.49	0.23	0.23	0.24	0.30	0.16	0.29
ASA24																
Day 1	463	0.03	-0.01	0.02	-0.01	0.05	0.01	0.25	0.19	0.25	0.01	0.03	0.03	N/A	N/A	N/A
Day 2	456	0.08	0.14	0.09	0.04	0.06	0.07	0.22	0.19	0.22	0.07	0.12	0.08	N/A	N/A	N/A
Day 3	455	-0.04	-0.03	-0.04	0.02	0.08	0.06	0.14	0.29	0.20	0.06	0.10	0.07	N/A	N/A	N/A
Day 4	449	0.01	-0.08	-0.01	0.02	0.02	0.01	0.11	0.18	0.12	0.08	0.05	0.07	N/A	N/A	N/A
Day 1 & 2	566	0.07	0.06	0.06	0.03	0.07	0.05	0.29	0.19	0.28	0.05	0.10	0.06	N/A	N/A	N/A
Day 3 & 4	548	-0.01	-0.06	-0.03	0.02	0.03	0.03	0.17	0.27	0.21	0.09	0.10	0.09	N/A	N/A	N/A
Day 1 & 3	580	0.00	0.00	-0.01	0.00	0.07	0.03	0.23	0.25	0.26	0.04	0.10	0.07	N/A	N/A	N/A
Day 2 & 4	564	0.08	0.02	0.06	0.05	0.07	0.06	0.20	0.19	0.20	0.10	0.11	0.10	N/A	N/A	N/A
Day 1 & 4	577	0.05	-0.05	0.02	0.02	0.02	0.01	0.22	0.19	0.21	0.04	0.04	0.05	N/A	N/A	N/A
Day 2 & 3	562	0.03	0.05	0.03	0.02	0.08	0.06	0.23	0.30	0.27	0.09	0.15	0.10	N/A	N/A	N/A
Days, all	626	0.05	0.01	0.03	0.02	0.06	0.04	0.29	0.27	0.31	0.08	0.12	0.09	N/A	N/A	N/A

* Nutrients adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing;

** Subgroups of women who didn't take supplements for this nutrient.

Supplementary Table 3.9: Spearman correlation coefficients for carotenoids, retinol, tocopherol and folate estimated from each SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Methods		Alpha carotene			Beta carotene*			Retinol*			Alpha-tocopherol*			Dietary folate equivalents*		
	N	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2
N		615	306	627	325	161	335	199	103	207	138	75	145	135	135	135
SFFQ1	627	0.41	0.38	0.41	0.40	0.32	0.41	0.09	-0.07	0.04	0.11	-0.07	0.10	0.30	0.22	0.28
SFFQ2	627	0.44	0.45	0.45	0.43	0.39	0.44	0.01	0.05	0.00	0.05	0.22	0.09	0.39	0.33	0.38
SFFQ1&2	627	0.45	0.45	0.46	0.44	0.38	0.45	0.07	-0.03	0.03	0.11	0.09	0.14	0.39	0.30	0.37
WebFFQ	627	0.38	0.41	0.40	0.39	0.37	0.40	0.03	0.06	0.05	0.19	0.15	0.17	0.30	0.32	0.32
7DDR														0.58	0.53	0.60
Week 1	626	0.47	0.42	0.46	0.49	0.43	0.48	0.03	0.06	0.03	0.12	-0.09	0.08	0.58	0.53	0.60
Week 2	626	0.42	0.44	0.43	0.45	0.45	0.47	0.13	0.10	0.12	0.13	0.18	0.14	0.49	0.58	0.58
Week 1 & 2	627	0.53	0.51	0.53	0.53	0.51	0.53	0.10	0.11	0.08	0.12	0.06	0.11	0.58	0.60	0.64
ASA24																
Day 1	464	0.19	0.14	0.19	0.26	0.21	0.27	-0.04	-0.03	-0.06	0.21	0.01	0.19	0.31	0.37	0.36
Day 2	457	0.25	0.25	0.25	0.29	0.34	0.31	0.06	0.01	0.04	0.00	0.06	0.03	0.29	0.26	0.30
Day 3	456	0.31	0.32	0.32	0.33	0.33	0.36	0.07	-0.05	0.02	0.14	0.12	0.08	0.30	0.32	0.32
Day 4	450	0.30	0.33	0.31	0.23	0.24	0.24	0.08	-0.14	0.04	0.09	-0.21	0.02	0.23	0.24	0.24
Day 1 & 2	567	0.24	0.20	0.24	0.34	0.33	0.36	0.01	0.01	-0.03	0.08	0.00	0.09	0.32	0.37	0.37
Day 3 & 4	549	0.37	0.38	0.38	0.35	0.36	0.37	0.05	-0.06	0.02	0.17	-0.02	0.10	0.32	0.33	0.33
Day 1 & 3	581	0.28	0.27	0.28	0.35	0.35	0.37	0.00	-0.06	-0.04	0.20	0.09	0.17	0.37	0.37	0.39
Day 2 & 4	565	0.30	0.29	0.31	0.25	0.24	0.26	0.03	-0.06	0.00	0.03	-0.08	-0.01	0.24	0.27	0.26
Day 1 & 4	578	0.30	0.27	0.31	0.32	0.33	0.34	0.01	-0.10	-0.03	0.16	-0.12	0.12	0.31	0.35	0.34
Day 2 & 3	563	0.29	0.31	0.29	0.35	0.39	0.37	0.08	-0.01	0.03	0.13	0.15	0.09	0.33	0.34	0.36
Days, all	627	0.34	0.32	0.34	0.39	0.41	0.41	0.03	-0.03	-0.01	0.17	0.02	0.12	0.36	0.41	0.40

* Subgroups of women who didn't take supplements for this nutrient.

Supplementary Table 3.10: Adjusted* Spearman correlation coefficients for carotenoids, tocopherol and folate estimated from each SFFQ, 7DDR, ASA24 and biomarker (data provided by 627 U.S. female nurses aged 45-80 years, 2010-2012).

Methods		Alpha carotene			Beta carotene**			Retinol**			Alpha-tocopherol **			Dietary folate equivalents**		
	N	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2	Blood 1	Blood 2	Blood 1 & 2
N		613	305	625	323	160	333	197	102	205	137	74	144	135	135	135
SFFQ1	627	0.44	0.43	0.44	0.40	0.35	0.42	0.16	-0.08	0.09	0.21	0.00	0.18	0.34	0.24	0.31
SFFQ2	627	0.44	0.45	0.46	0.42	0.32	0.44	0.11	0.09	0.09	0.13	0.03	0.12	0.38	0.39	0.41
SFFQ1&2	627	0.49	0.48	0.50	0.45	0.38	0.47	0.16	-0.02	0.10	0.24	0.06	0.23	0.42	0.35	0.41
WebFFQ	627	0.39	0.41	0.40	0.37	0.31	0.38	0.09	0.09	0.10	0.30	0.02	0.23	0.29	0.38	0.35
7DDR																
Week 1	626	0.47	0.39	0.45	0.51	0.46	0.50	0.02	0.08	0.04	0.38	0.03	0.29	0.48	0.37	0.45
Week 2	626	0.42	0.44	0.44	0.41	0.38	0.43	0.22	0.09	0.18	0.28	0.09	0.29	0.37	0.46	0.45
Week 1 & 2	627	0.52	0.49	0.52	0.53	0.49	0.54	0.15	0.09	0.13	0.38	0.08	0.33	0.45	0.45	0.49
ASA24																
Day 1	464	0.19	0.16	0.19	0.17	0.05	0.17	-0.01	-0.07	-0.05	0.12	-0.08	0.10	0.24	0.33	0.29
Day 2	457	0.21	0.21	0.21	0.19	0.14	0.19	0.08	-0.04	0.06	0.01	0.09	0.08	0.36	0.33	0.37
Day 3	456	0.26	0.23	0.26	0.28	0.23	0.29	0.13	0.08	0.09	0.22	0.09	0.16	0.24	0.32	0.31
Day 4	450	0.29	0.31	0.31	0.19	0.18	0.19	0.00	-0.20	-0.02	0.14	-0.16	0.06	0.18	0.20	0.19
Day 1 & 2	567	0.23	0.21	0.23	0.26	0.19	0.26	0.01	-0.06	-0.03	0.11	0.00	0.15	0.36	0.40	0.40
Day 3 & 4	549	0.34	0.32	0.36	0.29	0.27	0.30	0.08	-0.03	0.04	0.18	-0.02	0.11	0.27	0.30	0.30
Day 1 & 3	581	0.26	0.23	0.26	0.28	0.22	0.28	0.03	-0.04	-0.03	0.24	0.00	0.18	0.29	0.35	0.34
Day 2 & 4	565	0.28	0.29	0.29	0.18	0.12	0.18	0.00	-0.15	-0.03	0.08	-0.05	0.05	0.28	0.29	0.30
Day 1 & 4	578	0.29	0.28	0.30	0.23	0.19	0.23	0.00	-0.11	-0.05	0.16	-0.14	0.09	0.21	0.26	0.24
Day 2 & 3	563	0.25	0.25	0.25	0.26	0.23	0.27	0.09	0.00	0.04	0.15	0.10	0.12	0.32	0.37	0.37
Days, all	627	0.33	0.30	0.33	0.31	0.26	0.31	0.03	0.00	-0.02	0.21	-0.22	0.16	0.37	0.39	0.40

* Nutrients were adjusted for age and BMI at enrollment, current weight and smoking status at each measurement; self-reported nutrients were further adjusted for total energy intake; plasma levels were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing; Plasma carotenoids, retinol and was further adjusted for plasma lipids.

** Subgroups of women who didn't take supplements for this nutrient.