

## **WEB APPENDIX: BIOMARKER MEASUREMENT METHOD**

### **Doubly labeled water (DLW)**

Energy intake is estimated as the amount of carbon dioxide produced over a 10- to 14-day period, assuming that the individuals are in energy balance (1). 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  $^2\text{H}_2^{18}\text{O}$ , 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 ( $^2\text{H}$ ) and heavy oxygen ( $^{18}\text{O}$ ) (2–4). The relative decrease in deuterium and oxygen-18 concentrations was used to calculate total daily energy expenditure (TDEE) as described by Schoeller et al. (3, 4).

### **24-hour urine**

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 Human Nutrition Research Center on Aging 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) (5). Urinary values were divided by 0.86 for sodium, and 0.80 for potassium to convert them to the corresponding dietary values (mg/day) (6, 7).

### **Fasting plasma**

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 0.5-mL (one for folate, and one for carotenoids and fatty acids determinations), one 0.2-mL (for lipid 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 (8, 9). 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 (10–12). 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 et al. (13) 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.

## References

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**Web Table 1.** Within-person (CV%w) and between-person (CV%b) coefficients of variation for repeated assessments of energy, protein density, sodium density, potassium density, fatty acids, carotenoids, retinol tocopherols and folate estimated by SFFQs 7DDR<sub>s</sub> ASA24<sub>s</sub> and biomarkers (data provided by 627 U.S. female nurses aged 45–80 years, 2010–2012)

Nutrient <sup>a</sup>	Biomarker			SFFQ			7DDR			ASA24		
	Mean	CV%w	CV%b	Mean	CV%w	CV%b	Mean	CV%w	CV%b	Mean	CV%w	CV%b
<b>Total energy, Kcal/day</b>	2194	9	14	1890	15	23	1738	13	17	1811	28	18
<b>Protein density, %energy</b>	15	23	15	18	10	14	17	12	15	17	26	14
<b>Sodium density, mg/1000 kcal</b>	159	32	18	112	11	17	154	15	15	174	30	14
<b>Potassium density, mg/1000 kcal</b>	143	27	25	180	9	14	155	12	19	158	25	18
<b>Fatty acids, %total fatty acids or fat</b>												
Saturated fatty acids	31	10	3	35	10	14	33	10	12	34	22	11
Monounsaturated fatty acids	22	7	11	38	6	8	36	8	7	36	14	5
Polyunsaturated fatty acids												
Alpha-linolenic acid 18:3n-3c	0.6	23	27	2.3	27	37	2.3	27	38	2.2	68	26
Long Chain n-3 DHA+DPA+EPA	3.5	21	40	0.8	45	79	0.5	79	108	N/A	N/A	N/A
Long Chain n-3 <sup>b</sup>	2.8	23	29	0.4	70	77	0.3	100	104	0.3	288	21
Linoleic acid 18:2n-6cc	30	8	10	18	13	15	20	16	15	19	33	15
Total PUFA	45	7	6	22	12	15	22	15	15	22	33	15
<i>Trans</i> -Fatty acids	1.1	36	14	2.7	14	20	3.3	33	25	N/A	N/A	N/A
<b>Carotenoids, ug/L or ug/day</b>												
Lutein-zeaxanthin	283	18	49	3901	47	59	2701	69	91	2598	133	76
Beta cryptoxanthin	159	42	80	115	42	75	154	149	53	101	206	79
Lycopene	432	24	34	5678	46	59	4988	74	49	5011	147	45
Alpha carotene	101	44	84	888	51	77	613	74	61	501	164	118
Beta carotene	396	31	96	6630	36	52	4293	55	53	N/A	N/A	N/A
Beta carotene <sup>b</sup>	347	24	97	6475	34	49	3988	55	56	3464	112	56
<b>Retinol activity equivalents, ug/L or day</b>												
Retinol activity equivalents	620	12	19	1844	41	61	1481	73	71	N/A	N/A	N/A
Retinol activity equivalents <sup>b</sup>	592	13	18	1016	37	30	788	56	31	759	99	38
<b>Tocopherols, mg/L or mg/day</b>												
Alpha- tocopherol	14131	21	28	43	85	139	37	71	136	N/A	N/A	N/A

Alpha- tocopherol <sup>b</sup>	11418	21	16	11	87	27	9	25	36	8	56	29
Gama - tocopherol	1437	27	48	10	26	34	11	31	27	N/A	N/A	N/A
<b>Folate, ng/ml or ug/day</b>												
Dietary folate equivalents	32	69	47	1148	32	44	1062	30	64	N/A	N/A	N/A
Dietary folate equivalents <sup>b</sup>	19	81	61	622	56	31	518	24	38	484	53	30

Abbreviations: SFFQ, The semi-quantitative food frequency questionnaire; 7DDR, 7 Day Dietary Records; ASA24, Web-based, self-administered 24-hour dietary recall; CV, coefficients of variation;

<sup>a</sup>Two energy biomarker DLW, 9-12 months apart; four urinary biomarkers, approximately every three months over one year; two energy-adjusted urinary biomarkers, 9-12 months apart; two plasma biomarkers, approximately 6 months apart; Two SFFQs, approximately one year apart; Two 7DDRs, approximately 6 months apart; Four ASA24s, approximately every three months over one year.

<sup>b</sup>Subgroups of women who didn't take supplements for this nutrient ( $N = 363$  for long-chain n-3 fatty acids, 335 for beta-carotene, 207 for retinol, 148 for alpha-tocopherol, and 134 for folate).

**Web Table 2.** Spearman correlation coefficients between energy, protein, potassium sodium and Na/K ratio estimated from ASA24, SFFQ, WebFFQ, 7DDR and urinary recovery biomarkers (data provided by 624 U.S. female nurses aged 45–80 years, 2010–2012)

Nutrient	Unadjusted <i>r</i>	Deattenuated <i>r</i>	95% CI	Energy- Adjusted <sup>a</sup> <i>r</i>	Deattenuated <i>r</i>	95% CI
<b>Energy</b>						
Single ASA24	0.14	0.16	(0.06, 0.26)	N/A	N/A	N/A
Averaged ASA24	0.18	0.20	(0.11, 0.29)	N/A	N/A	N/A
SFFQ2	0.09	0.11	(0.02, 0.19)	N/A	N/A	N/A
SFFQ1 and 2	0.10	0.12	(0.03, 0.20)	N/A	N/A	N/A
WebFFQ	0.12	0.14	(0.05, 0.22)	N/A	N/A	N/A
Single 7DDR	0.38	0.43	(0.35, 0.50)	N/A	N/A	N/A
Averaged 7DDR	0.40	0.46	(0.37, 0.53)	N/A	N/A	N/A
<b>Protein</b>						
Single ASA24	0.31	0.32	(0.23, 0.40)	0.25	0.37	(0.19, 0.52)
Averaged ASA24	0.39	0.42	(0.35, 0.49)	0.33	0.54	(0.34, 0.70)
SFFQ2	0.25	0.27	(0.19, 0.35)	0.29	0.46	(0.27, 0.62)
SFFQ1 and 2	0.29	0.32	(0.25, 0.39)	0.33	0.52	(0.31, 0.68)
WebFFQ	0.26	0.29	(0.21, 0.37)	0.28	0.46	(0.27, 0.61)
Single 7DDR	0.67	0.64	(0.58, 0.69)	0.38	0.59	(0.36, 0.76)
Averaged 7DDR	0.65	0.71	(0.65, 0.75)	0.42	0.67	(0.41, 0.83)
<b>Potassium</b>						
Single ASA24	0.40	0.43	(0.35, 0.51)	0.33	0.41	(0.29, 0.51)
Averaged ASA24	0.48	0.52	(0.44, 0.58)	0.43	0.52	(0.41, 0.61)
SFFQ2	0.32	0.36	(0.29, 0.43)	0.38	0.49	(0.38, 0.58)
SFFQ1 and 2	0.34	0.40	(0.33, 0.47)	0.39	0.49	(0.38, 0.58)
WebFFQ	0.32	0.35	(0.28, 0.42)	0.32	0.42	(0.32, 0.51)
Single 7DDR	0.65	0.68	(0.62, 0.73)	0.46	0.59	(0.48, 0.68)
Averaged 7DDR	0.65	0.73	(0.67, 0.78)	0.50	0.64	(0.52, 0.72)
<b>Sodium</b>						
Single ASA24	0.24	0.24	(0.14, 0.34)	0.15	0.25	(0.08, 0.40)
Averaged ASA24	0.29	0.35	(0.26, 0.42)	0.19	0.36	(0.17, 0.53)
SFFQ2	0.16	0.18	(0.10, 0.27)	0.25	0.48	(0.25, 0.65)
SFFQ1 and 2	0.18	0.21	(0.11, 0.29)	0.26	0.49	(0.25, 0.67)
WebFFQ	0.15	0.18	(0.09, 0.27)	0.26	0.46	(0.23, 0.65)
Single 7DDR	0.43	0.52	(0.43, 0.58)	0.29	0.49	(0.23, 0.67)
Averaged 7DDR	0.48	0.57	(0.49, 0.64)	0.32	0.54	(0.26, 0.74)
<b>Na/K<sup>b</sup></b>						
Single ASA24	0.36	0.40	(0.32, 0.48)	N/A	N/A	N/A
Averaged ASA24	0.39	0.53	(0.46, 0.60)	N/A	N/A	N/A
SFFQ2	0.45	0.53	(0.46, 0.60)	N/A	N/A	N/A
SFFQ1 and 2	0.51	0.57	(0.50, 0.63)	N/A	N/A	N/A
WebFFQ	0.43	0.49	(0.41, 0.55)	N/A	N/A	N/A
Single 7DDR	0.59	0.66	(0.60, 0.71)	N/A	N/A	N/A
Averaged 7DDR	0.65	0.73	(0.67, 0.78)	N/A	N/A	N/A

Abbreviations: SFFQ, The semi-quantitative food frequency questionnaire; WebFFQ: web-based version of the SFFQ; 7DDR, 7 Day Dietary Records; ASA24, Web-based, self-administered 24-hour dietary recall.

<sup>a</sup> Protein, sodium and potassium from self-reported measures and recovery biomarker were adjusted for total energy intake using energy density method.

<sup>b</sup> Sodium/potassium (Na/K) ratio was calculated from sodium value divided by potassium value.



**Web Table 3.** *P* values for pairwise comparisons among deattenuated and energy-adjusted Spearman correlation coefficients with recovery biomarkers (data provided by 624 U.S. female nurses aged 45–80 years, 2010–2012)

<b>Comparison</b>	<b>Protein<sup>a</sup> (% energy)</b>	<b>Sodium<sup>a</sup> (mg/1000 kcal)</b>	<b>Potassium<sup>a</sup> (mg/1000 kcal)</b>	<b>Na/K<sup>b</sup></b>	<b>Energy (kcal/day)</b>
<b>SFFQ2</b>					
vs SFFQ1 and 2	0.61	0.76	0.64	0.70	0.87
vs Single ASA24	0.20	0.01	0.07	0.04	0.25
vs Averaged ASA24	0.20	0.02	0.07	0.07	0.22
vs Single 7DDR	0.03	0.28	0.02	0.001	<0.001
vs Averaged 7DDR	0.03	0.27	0.02	0.003	<0.001
<b>SFFQ1 and 2</b>					
vs Single ASA24	0.09	0.01	0.21	0.02	0.18
vs Averaged ASA24	0.10	0.01	0.20	0.04	0.16
vs Single 7DDR	0.03	0.20	0.01	0.001	<0.001
vs Averaged 7DDR	0.03	0.19	0.01	0.003	<0.001
<b>Single ASA24</b>					
vs Averaged ASA24	0.97	0.88	0.93	0.65	0.93
vs Single 7DDR	<0.001	<0.001	<0.001	<0.001	<0.001
vs Averaged 7DDR	<0.001	<0.001	<0.001	<0.001	<0.001
<b>Averaged ASA24</b>					
vs Single 7DDR	<0.001	<0.001	<0.001	<0.001	0.37
vs Averaged 7DDR	<0.001	<0.001	<0.001	<0.001	<0.001
<b>Single 7DDR</b>					
vs Averaged 7DDR	0.99	0.99	0.99	0.99	1.00

Abbreviation: SFFQ, The semi-quantitative food frequency questionnaire; 7DDR, 7 Day Dietary Records; ASA24, Web-based, automated-self-administered 24-hour dietary recall.

<sup>a</sup>Protein, sodium and potassium from self-reported measures and recovery biomarker were adjusted for total energy intake using energy density method;

<sup>b</sup>Sodium/potassium (Na/K) ratio was calculated from sodium value divided by potassium value.

**Web Table 4.** Spearman correlation coefficients between specific fatty acids ( of total fatty acids) estimated from ASA24, SFFQ, WebFFQ, 7DDR and plasma fatty acids biomarkers (data provided by 627 U.S. female nurses aged 45–80 years, 2010–2012)

<b>Nutrient</b>	<b>Unadjusted <i>r</i></b>	<b>Adjusted<sup>a</sup> <i>r</i></b>	<b>Deattenuated <i>r</i></b>	<b>95% CI</b>
<b>Saturated fatty acids</b>				
Single ASA24	0.04	0.03	0.06	(-0.12, 0.24)
Averaged ASA24	0.07	0.05	0.10	(-0.05, 0.24)
SFFQ2	0.09	0.08	0.15	(0.00, 0.29)
SFFQ1 and 2	0.09	0.08	0.15	(0.00, 0.29)
WebFFQ	0.12	0.10	0.19	(0.03, 0.33)
Single 7DDR	0.16	0.16	0.30	(0.14, 0.43)
Averaged 7DDR	0.18	0.17	0.33	(0.17, 0.47)
<b>Monounsaturated fatty acids</b>				
Single ASA24	0.02	0.04	0.05	(-0.06, 0.15)
Averaged ASA24	0.02	0.06	0.07	(-0.02, 0.15)
SFFQ2	0.04	0.07	0.08	(-0.01, 0.17)
SFFQ1 and 2	0.05	0.08	0.09	(0.00, 0.17)
WebFFQ	0.04	0.06	0.07	(-0.03, 0.15)
Single 7DDR	0.04	0.03	0.04	(-0.06, 0.12)
Averaged 7DDR	0.04	0.03	0.04	(-0.05, 0.12)
<b>Alpha-linolenic acid (18:3n-3c)</b>				
Single ASA24	0.18	0.17	0.21	(0.10, 0.31)
Averaged ASA24	0.22	0.22	0.27	(0.18, 0.36)
SFFQ2	0.28	0.28	0.34	(0.25, 0.42)
SFFQ1 and 2	0.34	0.34	0.41	(0.32, 0.49)
WebFFQ	0.30	0.30	0.36	(0.28, 0.44)
Single 7DDR	0.34	0.34	0.42	(0.33, 0.50)
Averaged 7DDR	0.39	0.39	0.48	(0.39, 0.56)
<b>Long Chain n-3 (DHA+DPA+EPA)</b>				
SFFQ2	0.57	0.56	0.62	(0.56, 0.68)
SFFQ1 and 2	0.61	0.61	0.66	(0.60, 0.71)
WebFFQ	0.56	0.57	0.61	(0.55, 0.66)
Single 7DDR	0.61	0.60	0.66	(0.60, 0.71)
Averaged 7DDR	0.67	0.66	0.74	(0.68, 0.79)
<b>Long Chain n-3<sup>b</sup></b>				
Single ASA24	0.19	0.19	0.23	(0.09, 0.36)
Averaged ASA24	0.31	0.30	0.36	(0.25, 0.46)
SFFQ2	0.57	0.53	0.58	(0.47, 0.66)
SFFQ1 and 2	0.61	0.56	0.64	(0.54, 0.72)
WebFFQ	0.49	0.44	0.52	(0.41, 0.61)
Single 7DDR	0.47	0.46	0.53	(0.42, 0.62)
Averaged 7DDR	0.55	0.52	0.61	(0.51, 0.69)
<b>Linoleic acid (18:2n-6cc)</b>				
Single ASA24	0.06	0.06	0.08	(-0.04, 0.19)

Averaged ASA24	0.10	0.10	0.13	(0.03, 0.22)
SFFQ2	0.16	0.20	0.25	(0.16, 0.34)
SFFQ1 and 2	0.17	0.20	0.25	(0.16, 0.35)
WebFFQ	0.18	0.19	0.23	(0.14, 0.33)
Single 7DDR	0.21	0.20	0.25	(0.15, 0.34)
Averaged 7DDR	0.25	0.23	0.29	(0.20, 0.38)
<b>Polyunsaturated fatty acids</b>				
Single ASA24	0.08	0.06	0.09	(-0.04, 0.22)
Averaged ASA24	0.11	0.09	0.14	(0.03, 0.24)
SFFQ2	0.16	0.18	0.27	(0.15, 0.37)
SFFQ1 and 2	0.18	0.19	0.27	(0.16, 0.37)
WebFFQ	0.19	0.17	0.25	(0.13, 0.36)
Single 7DDR	0.21	0.19	0.28	(0.16, 0.38)
Averaged 7DDR	0.24	0.22	0.32	(0.21, 0.42)
<b>Trans-Fatty Acids</b>				
SFFQ2	0.29	0.27	0.33	(0.16, 0.48)
SFFQ1 and 2	0.32	0.30	0.42	(0.25, 0.57)
WebFFQ	0.30	0.27	0.38	(0.19, 0.57)
Single 7DDR	0.28	0.27	0.46	(0.30, 0.59)
Averaged 7DDR	0.33	0.30	0.53	(0.36, 0.66)

Abbreviations: SFFQ, The semi-quantitative food frequency questionnaire; WebFFQ: web-based version of the SFFQ; 7DDR, 7 Day Dietary Records; ASA24, Web-based, self-administered 24-hour dietary recall; BMI, body mass index.

<sup>a</sup> Fatty acid biomarkers and associated self-reported fatty acid composition were adjusted for age and BMI at enrollment, current weight and smoking status at each measurement, fatty acid biomarkers were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing.

<sup>b</sup> Analyses for long-chain n-3 fatty acids was performed among subgroups of women ( $N = 363$ ) not taking fatty acid supplement.

**Web Table 5.** Spearman correlation coefficients between carotenoids, retinol, tocopherol and folate intakes estimated from ASA24, SFFQ, WebFFQ, 7DDR and plasma concentration biomarkers (data provided by 627 U.S. female nurses aged 45–80 years, 2010–2012)

<b>Nutrient</b>	<b>Unadjusted <i>r</i></b>	<b>Adjusted<sup>a</sup> <i>r</i></b>	<b>Deattenuated <i>r</i></b>	<b>95% CI</b>
<b>Lycopene</b>				
Single ASA24	0.17	0.14	0.16	(0.06, 0.28)
Averaged ASA24	0.22	0.20	0.25	(0.15, 0.34)
SFFQ2	0.26	0.28	0.33	(0.24, 0.41)
SFFQ1 and 2	0.26	0.30	0.32	(0.22, 0.40)
WebFFQ	0.27	0.30	0.37	(0.28, 0.45)
Single 7DDR	0.28	0.32	0.37	(0.28, 0.45)
Averaged 7DDR	0.35	0.40	0.47	(0.38, 0.55)
<b>Alpha-carotene</b>				
Single ASA24	0.29	0.25	0.27	(0.18, 0.36)
Averaged ASA24	0.37	0.34	0.37	(0.30, 0.44)
SFFQ2	0.47	0.47	0.51	(0.44, 0.57)
SFFQ1 and 2	0.49	0.51	0.56	(0.49, 0.62)
WebFFQ	0.42	0.42	0.46	(0.39, 0.53)
Single 7DDR	0.46	0.47	0.51	(0.44, 0.57)
Averaged 7DDR	0.55	0.55	0.60	(0.53, 0.65)
<b>Beta-carotene</b>				
SFFQ2	0.39	0.40	0.41	(0.35, 0.48)
SFFQ1 and 2	0.43	0.44	0.47	(0.40, 0.53)
WebFFQ	0.35	0.36	0.39	(0.32, 0.45)
Single 7DDR	0.45	0.45	0.48	(0.41, 0.54)
Averaged 7DDR	0.52	0.53	0.56	(0.50, 0.62)
<b>Beta-carotene<sup>b</sup></b>				
Single ASA24	0.31	0.22	0.24	(0.11, 0.36)
Averaged ASA24	0.42	0.33	0.36	(0.25, 0.45)
SFFQ2	0.46	0.48	0.47	(0.37, 0.56)
SFFQ1 and 2	0.47	0.50	0.50	(0.40, 0.59)
WebFFQ	0.40	0.39	0.41	(0.32, 0.51)
Single 7DDR	0.48	0.46	0.50	(0.40, 0.58)
Averaged 7DDR	0.54	0.53	0.58	(0.49, 0.66)
<b>Lutein-zeaxanthin</b>				
Single ASA24	0.27	0.23	0.25	(0.16, 0.34)
Averaged ASA24	0.33	0.29	0.32	(0.24, 0.39)
SFFQ2	0.33	0.32	0.35	(0.28, 0.42)
SFFQ1 and 2	0.35	0.35	0.38	(0.31, 0.45)
WebFFQ	0.29	0.30	0.33	(0.26, 0.40)
Single 7DDR	0.40	0.39	0.42	(0.34, 0.49)
Averaged 7DDR	0.46	0.45	0.48	(0.41, 0.55)
<b>Beta-cryptoxanthin</b>				
Single ASA24	0.32	0.29	0.33	(0.24, 0.42)

Averaged ASA24	0.42	0.40	0.45	(0.37, 0.52)
SFFQ2	0.40	0.41	0.46	(0.38, 0.53)
SFFQ1 and 2	0.43	0.44	0.50	(0.42, 0.56)
WebFFQ	0.40	0.41	0.45	(0.37, 0.52)
Single 7DDR	0.42	0.39	0.44	(0.36, 0.50)
Averaged 7DDR	0.50	0.47	0.53	(0.46, 0.60)
<b>Retinol activity equivalents</b>				
SFFQ2	0.14	0.09	0.10	(0.01, 0.18)
SFFQ1 and 2	0.15	0.10	0.11	(0.03, 0.20)
WebFFQ	0.18	0.11	0.13	(0.05, 0.22)
Single 7DDR	0.17	0.13	0.14	(0.06, 0.23)
Averaged 7DDR	0.18	0.13	0.15	(0.07, 0.24)
<b>Retinol activity equivalents<sup>b</sup></b>				
Single ASA24	0.02	0.03	0.04	(-0.16, 0.23)
Averaged ASA24	0.03	0.02	0.03	(-0.14, 0.20)
SFFQ2	0.03	0.13	0.17	(-0.01, 0.34)
SFFQ1 and 2	0.06	0.12	0.15	(-0.02, 0.32)
WebFFQ	0.07	0.11	0.14	(-0.04, 0.30)
Single 7DDR	0.09	0.13	0.17	(-0.01, 0.33)
Averaged 7DDR	0.10	0.15	0.20	(0.03, 0.37)
<b>Alpha-tocopherol</b>				
SFFQ2	0.49	0.52	0.65	(0.57, 0.72)
SFFQ1 and 2	0.51	0.54	0.67	(0.59, 0.73)
WebFFQ	0.50	0.54	0.67	(0.59, 0.74)
Single 7DDR	0.54	0.59	0.74	(0.66, 0.81)
Averaged 7DDR	0.57	0.64	0.79	(0.71, 0.85)
<b>Alpha-tocopherol<sup>b</sup></b>				
Single ASA24	0.09	0.08	0.08	(-0.20, 0.34)
Averaged ASA24	0.13	0.07	0.08	(-0.13, 0.29)
SFFQ2	0.11	0.22	0.29	(0.08, 0.48)
SFFQ1 and 2	0.13	0.27	0.37	(0.15, 0.55)
WebFFQ	0.16	0.27	0.38	(0.16, 0.56)
Single 7DDR	0.13	0.36	0.49	(0.28, 0.67)
Averaged 7DDR	0.15	0.40	0.54	(0.33, 0.71)
<b>Gamma-tocopherol</b>				
SFFQ2	0.00	0.02	0.02	(-0.07, 0.11)
SFFQ1 and 2	-0.01	0.03	0.03	(-0.06, 0.11)
WebFFQ	0.02	0.03	0.03	(-0.06, 0.11)
Single 7DDR	0.11	0.11	0.12	(0.03, 0.20)
Averaged 7DDR	0.12	0.12	0.13	(0.05, 0.22)
<b>Dietary folate equivalents<sup>c</sup></b>				
SFFQ2	0.47	0.49	0.54	(0.46, 0.61)
SFFQ1 and 2	0.50	0.50	0.55	(0.47, 0.62)
WebFFQ	0.47	0.46	0.51	(0.42, 0.58)

Single 7DDR	0.57	0.57	0.63	(0.56, 0.69)
Averaged 7DDR	0.61	0.61	0.68	(0.60, 0.73)
<b>Dietary folate equivalents<sup>b</sup></b>				
Single ASA24	0.31	0.29	0.31	(0.13, 0.47)
Averaged ASA24	0.43	0.40	0.43	(0.27, 0.57)
SFFQ2	0.37	0.43	0.47	(0.32, 0.60)
SFFQ1 and 2	0.40	0.44	0.47	(0.32, 0.60)
WebFFQ	0.29	0.33	0.36	(0.19, 0.50)
Single 7DDR	0.56	0.58	0.62	(0.49, 0.73)
Averaged 7DDR	0.60	0.62	0.68	(0.55, 0.78)

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Abbreviations: SFFQ, The semi-quantitative food frequency questionnaire; WebFFQ: web-based version of the SFFQ; 7DDR, 7 Day Dietary Records; ASA24, Web-based, self-administered 24-hour dietary recall; BMI, body mass index.

<sup>a</sup> Carotenoids, retinol, tocopherol and folate biomarkers and associated self-reported 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, biomarkers were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing; plasma carotenoids, retinol and tocopherols was additionally adjusted for plasma lipids.

<sup>b</sup> Analyses for beta-carotene ( $N = 335$ ), retinol ( $N = 207$ ), alpha-tocopherol ( $N = 148$ ) and folate ( $N = 134$ ) were performed among subgroups of women not taking a supplement containing the corresponding nutrient.

<sup>c</sup> Plasma folate biomarker was measured among 456 participants.

**Web Table 6.** *P* values for pairwise comparisons among deattenuated and adjusted Spearman correlation coefficients with plasma fatty acids (% total fatty acids)<sup>a</sup> (data provided by 627 U.S. female nurses aged 45–80 years, 2010–2012)

Comparison	Saturated Fatty Acids	MUFA	ALA	Long Chain Fatty acids	Linoleic acid	PUFA	Trans-fatty acids
SFFQ2							
vs SFFQ1 and 2	0.63	0.99	0.18	0.58	0.23	0.53	0.99
vs Single ASA24	0.14	0.29	0.005	<0.001	<0.001	0.002	N/A
vs Averaged ASA24	0.14	0.30	0.003	<0.001	0.001	0.002	N/A
vs Single 7DDR	0.06	0.23	0.17	0.41	0.95	0.85	0.89
vs Averaged 7DDR	0.06	0.23	0.18	0.41	0.99	0.77	0.90
SFFQ1 and 2							
vs Single ASA24	0.14	0.24	<0.001	<0.001	0.001	0.002	N/A
vs Averaged ASA24	0.14	0.27	<0.001	<0.001	0.002	0.002	N/A
vs Single 7DDR	0.02	0.20	0.39	0.26	0.54	0.55	0.87
vs Averaged 7DDR	0.13	0.19	0.41	0.26	0.50	0.51	0.88
Single ASA24							
vs Averaged ASA24	0.91	0.99	0.84	0.65	0.77	0.97	N/A
vs Single 7DDR	0.001	0.95	<0.001	<0.001	<0.001	<0.001	N/A
vs Averaged 7DDR	<0.001	0.94	<0.001	<0.001	<0.001	<0.001	N/A
Averaged ASA24							
vs Single 7DDR	0.001	0.91	<0.001	<0.001	<0.001	<0.001	N/A
vs Averaged 7DDR	0.001	0.91	<0.001	<0.001	<0.001	<0.001	N/A
Single 7DDR							
vs Averaged 7DDR	1.00	1.00	0.98	1.00	1.00	0.99	0.99

Abbreviations: SFFQ, The semi-quantitative food frequency questionnaire; 7DDR, 7 Day Dietary Records; ASA24, Web-based, self-administered 24-hour dietary recall; MUFA, Monounsaturated Fatty Acids; ALA, alpha-linolenic acid; PUFA, Polyunsaturated Fatty Acids; BMI, body mass index.

<sup>a</sup>Fatty acid biomarkers and associated self-reported fatty acid composition were adjusted for age and BMI at enrollment, current weight and smoking status at each measurement, fatty acid biomarkers were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing;

<sup>b</sup>Analyses for long-chain n-3 fatty acids ( $N = 363$ ) were performed among subgroups of women not taking fatty acid supplement.

**Web Table 7.** *P* values for pairwise comparisons among deattenuated and adjusted Spearman correlation coefficients with plasma carotenoids, retinol, tocopherol and folate<sup>a</sup> (data provided by 627 U.S. female nurses aged 45–80 years, 2010–2012)

Comparison	Alpha carotene	Beta carotene <sup>b</sup>	Lutein-zeaxanthin	Beta cryptoxanthin	Lycopene	Retinol <sup>b</sup>	Alpha tocopherol <sup>b</sup>	Folate <sup>b</sup>
SFFQ2								
vs SFFQ1 and 2	0.89	0.63	0.86	0.61	0.24	0.61	0.19	0.52
vs Single ASA24	<0.001	<0.001	0.004	0.002	<0.001	0.28	0.62	0.47
vs Averaged ASA24	<0.001	<0.001	0.002	0.002	<0.001	0.19	0.95	0.15
vs Single 7DDR	0.80	0.77	0.10	0.61	0.71	0.98	0.02	0.40
vs Averaged 7DDR	0.85	0.76	0.09	0.54	0.68	0.88	0.02	0.40
SFFQ1 and 2								
vs Single ASA24	<0.001	<0.001	0.003	<0.001	<0.001	0.30	0.83	0.84
vs Averaged ASA24	<0.001	<0.001	0.001	<0.001	0.002	0.25	0.55	0.31
vs Single 7DDR	0.84	0.61	0.06	0.48	0.33	0.76	<0.001	0.05
vs Averaged 7DDR	0.89	0.57	0.06	0.39	0.28	0.65	<0.001	0.05
Single ASA24								
vs Averaged ASA24	0.79	0.91	0.71	0.99	0.88	0.88	0.92	0.65
vs Single 7DDR	<0.001	<0.001	<0.001	0.003	<0.001	0.12	0.01	0.01
vs Averaged 7DDR	<0.001	<0.001	<0.001	0.007	<0.001	0.14	0.01	0.01
Averaged ASA24								
vs Single 7DDR	<0.001	<0.001	<0.001	0.004	<0.001	0.09	0.01	0.04
vs Averaged 7DDR	<0.001	<0.001	<0.001	0.006	<0.001	0.09	0.01	0.03
Single 7DDR								
vs Averaged 7DDR	1.00	0.98	0.99	0.99	0.99	0.99	1.00	1.00

Abbreviations: SFFQ, The semi-quantitative food frequency questionnaire; 7DDR, 7 Day Dietary Records; ASA24, Web-based, self-administered 24-hour dietary recall; BMI, body mass index.

<sup>a</sup>Carotenoids, retinol, tocopherol and folate biomarkers and associated self-reported 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, biomarkers were further adjusted for postmenopausal status, hormone use, and fasting status at blood drawing; Plasma carotenoids, retinol and tocopherols was additionally adjusted for plasma lipids.

<sup>b</sup>Analyses for beta-carotene ( $N = 335$ ), retinol ( $N = 207$ ), alpha-tocopherol ( $N = 148$ ) and folate ( $N = 134$ ) were performed among subgroups of women not taking a supplement containing the corresponding nutrient.



**Web Table 8.** Distribution of fatty acids ( of total fatty acids) estimated by SFFQ, 7DDR, ASA24, and biomarkers (data provided by 627 U.S. female nurses aged 45–80 years, 2010–2012)

Fatty Acid	Biomarker	Paper SFFQ2		7DDR	ASA24
	Blood 1 and 2 average	Harvard <sup>a</sup> nutrient base	Standard <sup>a</sup> nutrient base	Week 1 and 2 average	Day1-4 average
<b>Saturated fatty acids</b>					
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
<b>Monounsaturated fatty acids</b>					
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 <sup>b</sup>	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
<b>Polyunsaturated fatty acids</b>					
n-3					
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 <sup>c</sup>	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 <sup>d</sup>	3.5 ± 1.6	0.8 ± 0.7	0.8 ± 0.7	0.5 ± 0.7	N/A
n-6					
18:2n-6c	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	N/A	N/A	N/A
20:2n-6c	0.22 ± 0.04	0.04 ± 0.02	N/A	N/A	N/A
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	N/A	N/A	N/A

Total PUFA	44.8 ± 4.0	22.2 ± 4.6	21.5 ± 4.6	22.4 ± 4.2	21.9 ± 5.6
<b>Trans-Fatty acids</b>					
t-16:1n-7	0.13 ± 0.03	0.11 ± 0.04	N/A	0.06 ± 0.03	N/A
Total 18:1 <i>trans</i> -	0.8 ± 0.3	2.2 ± 0.6	N/A	2.6 ± 1.0	N/A
Total 18:2 <i>trans</i> -	0.2 ± 0.1	0.4 ± 0.1	N/A	0.5 ± 0.1	N/A
Total trans	1.1 ± 0.4	2.7 ± 0.7	N/A	3.3 ± 1.1	N/A
Believed to be trans	0.2 ± 0.1	0.5 ± 0.2	N/A	N/A	N/A
<b>Dairy fat</b>					
Derived dairy fat	N/A	N/A	19.0 ± 9.1	N/A	N/A
15:00	0.15 ± 0.04	N/A	N/A	N/A	N/A
17:00	0.33 ± 0.07	N/A	N/A	0.12 ± 0.05	N/A
t-16:1n-7	0.13 ± 0.03	N/A	N/A	N/A	N/A

Abbreviations: SFFQ, The semi-quantitative food frequency questionnaire; 7DDR, 7 Day Dietary Records; ASA24, Web-based, self-administered 24-hour dietary recall; BMI, body mass index.

<sup>a</sup> In our SFFQ2, two sets of fatty acid variables were available. One type was derived based primarily on USDA sources (Standard) while the other one (Harvard) 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.

<sup>b</sup> Total 18:1 was the sum of 18:1n-9c and 18:1n-7c;

<sup>c</sup> Total 18:3 was the sum of alpha-linolenic (18:3n-3c) and gamma-linolenic (18:3n-6c).

<sup>d</sup> Long Chain n-3 fatty acid was the sum of 20:5n-3c, 22:5n-3c and 22:6n-3c.

**Web Table 9.** Reproducibility (rank intraclass correlation)<sup>a</sup> of fatty acids ( of total fatty acids) estimated by biomarkers, SFFQ, and 7DDR (data provided by 627 U.S. female nurses aged 45–80 years, 2010–2012)

Variable	Biomarker		7DDR		SFFQ (Harvard <sup>b</sup> )		SFFQ (standard <sup>b</sup> )	
	Unadjusted	Adjusted	Unadjusted	Adjusted	Unadjusted	Adjusted	Unadjusted	Adjusted
<b>Saturated fatty acids</b>								
12:00	0.13	0.14	0.40	0.40	0.62	0.63	0.64	0.61
14:00	0.32	0.30	0.52	0.52	0.62	0.61	0.62	0.61
16:00	0.52	0.47	0.54	0.54	0.63	0.63	0.63	0.63
18:00	0.17	0.15	0.54	0.53	0.68	0.67	0.67	0.67
Total	0.21	0.20	0.58	0.58	0.65	0.64	0.64	0.64
<b>Monounsaturated fatty acids</b>								
16:1n-7c	0.77	0.73	0.50	0.50	0.57	0.54	0.55	0.56
18:1n-9c	0.69	0.65	0.42	0.42	0.63	0.61	0.62	0.63
Total 18:1 <sup>c</sup>	0.68	0.64	0.42	0.42	0.61	0.61	0.62	0.61
20:1n-9c	0.41	0.39	0.34	0.34	0.63	0.58	0.59	0.61
Total	0.69	0.64	0.41	0.41	0.62	0.62	0.62	0.62
<b>Polyunsaturated fatty acids</b>								
n-3								
18:3n-3c	0.59	0.57	0.48	0.47	0.59	0.60	0.60	0.58
Total 18:3 <sup>d</sup>	0.55	0.54	0.48	0.47	0.59	0.60	0.60	0.58
20:5n-3c	0.77	0.74	0.62	0.60	0.75	0.72	0.75	0.72
22:5n-3c	0.63	0.60	0.43	0.43	0.72	0.70	0.72	0.67
22:6n-3c	0.78	0.74	0.57	0.55	0.74	0.73	0.74	0.72
Long Chain n-3 <sup>e</sup>	0.78	0.75	0.61	0.59	0.75	0.74	0.75	0.73
n-6								
18:2n-6cc	0.58	0.53	0.44	0.44	0.54	0.54	0.55	0.53
18:3n-6c	0.66	0.64	0.48	0.47	N/A	N/A	N/A	N/A
20:2n-6c	0.49	0.48	0.57	0.57	N/A	N/A	N/A	N/A
20:4n-6c	0.77	0.73	0.50	0.50	0.63	0.62	0.63	0.62

Total n-6	0.48	0.44	0.57	0.57	N/A	N/A	N/A	N/A
Total PUFA	0.43	0.38	0.46	0.46	0.58	0.59	0.60	0.58
<b>Trans-fat</b>								
t-16:1n-7	0.39	0.39	0.41	0.41	0.66	0.54	N/A	N/A
Total 18:1 <i>trans</i> -	0.22	0.20	0.40	0.39	0.64	0.61	N/A	N/A
total 18:2 <i>trans</i> -isomers	0.33	0.26	0.41	0.40	0.63	0.54	N/A	N/A
Total 18:2 <i>trans</i> -	0.33	0.26	0.41	0.40	0.63	0.54	N/A	N/A
Total trans	0.25	0.23	0.41	0.40	0.65	0.63	N/A	N/A
Believed to be trans	0.47	0.44	N/A	N/A	0.63	0.63	N/A	N/A
<b>Dairy related fat</b>								
Derived dairy fat	N/A	N/A	N/A	N/A	N/A	N/A	0.59	0.59
15:00	0.33	0.34	N/A	N/A	N/A	N/A	N/A	N/A
17:00	0.31	0.32	0.41	0.40	N/A	N/A	N/A	N/A
t-16:1n-7	0.39	0.39	0.41	0.41	0.66	0.54	N/A	N/A

Abbreviations: SFFQ, The semi-quantitative food frequency questionnaire; 7DDR, 7 Day Dietary Records; ASA24, Web-based, self-administered 24-hour dietary recall; BMI, body mass index.

<sup>a</sup> Two plasma biomarkers, approximately 6 months apart; Two SFFQs, approximately one year apart; Two 7DDRs, approximately 6 months apart; Four ASA24s, approximately every three months over one year. Plasma fatty acid biomarker was adjusted for age and BMI at enrollment, current weight, smoking status postmenopausal status, hormone use, and fasting status at blood drawing at each measurement. Self-reported fat composition was adjusted for age and BMI at enrollment, current weight and smoking status at each measurement.

<sup>b</sup> In our SFFQ2, two sets of fatty acid variables were available. One type was derived based primarily on USDA sources (Standard) while the other one (Harvard) 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.

<sup>d</sup> Total 18:1 was the sum of 18:1n-9c and 18:1n-7c;

<sup>d</sup> Total 18:3 was the sum of alpha-linolenic (18:3n-3c) and gamma-linolenic (18:3n-6c).



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
<b>Trans-Fatty acids</b>												
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 <i>trans</i> -	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 <i>trans</i> -	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
<b>Dairy fat</b>												
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

Abbreviations: SFFQ, The semi-quantitative food frequency questionnaire; 7DDR, 7 Day Dietary Records; ASA24, Web-based, self-administered 24-hour dietary recall; BMI, body mass index.

<sup>a</sup> In our SFFQ2, two sets of fatty acid variables were available. One type was derived based primarily on USDA sources (Standard) while the other one (Harvard) 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.

<sup>b</sup> Plasma fatty acid biomarker was adjusted for age and BMI at enrollment, current weight, smoking status postmenopausal status, hormone use, and fasting status at blood drawing at each measurement. Self-reported fat composition was adjusted for age and BMI at enrollment, current weight and smoking status at each measurement.

<sup>c</sup> Total 18:1 was the sum of 18:1n-9c and 18:1n-7c;

<sup>d</sup> Total 18:3 was the sum of alpha-linolenic (18:3n-3c) and gamma-linolenic (18:3n-6c).

<sup>e</sup> Long Chain n-3 fatty acid was the sum of 20:5n-3c, 22:5n-3c and 22:6n-3c.