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Dietary Intake and Major Food Sources of Polyphenols in Finnish Adults^{1–3}

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Abstract

Phenolic acids, flavonoids, proanthocyanidins, and ellagitannins are polyphenols that may have beneficial effects on human health and provide protection against chronic diseases. To date, limited data exist on quantitative intake of polyphenols. The aims of this study were to estimate the quantitative intakes of polyphenols by using analyzed concentrations together with individual food consumption records and to determine major dietary sources. Analyzed concentrations of phenolic acids, anthocyanidins, and other flavonoids, proanthocyanidins, and ellagitannins (44 total polyphenol compounds) were entered into the national food composition database, Fineli. The absolute intakes of the polyphenols and the corresponding food sources were calculated on the basis of 48-h dietary recalls of 2007 Finnish adults. The mean total intake of polyphenols was 863 ± 415 mg/d. Phenolic acids comprised the dominant group of polyphenols (75% of total intake) followed by proanthocyanidins (14%) and anthocyanidins and other flavonoids (10%). Due to their high consumption and high concentrations of phenolic acids, coffee and cereals were the main contributors to total polyphenol intake. Berries and berry products were the main source for anthocyanidins, ellagitannins, and proanthocyanidins, and fruits were the main source for flavonols, flavones, and flavanones. The results give additional support to the recommendations for a varied diet with fruits, berries, cereals, and vegetables. J. Nutr. 138: 562–566, 2008.

Introduction

Phenolic compounds are widely distributed in all plants, the most common polyphenol classes being phenolic acids, flavonoids, and tannins (1–3). These compounds constitute a very diverse group of secondary plant metabolites and they are further divided into subclasses according to their chemical structure. Phenolic acids have a simple structure with a single aromatic ring and they comprise hydroxycinnamic acids and hydroxybenzoic acids. Flavonoids include anthocyanidins, flavonols, flavones, flavanones, catechins (monomeric flavan-3-ols), and isoflavonoids (1,4,5). Tannins can be divided into condensed tannins, i.e. proanthocyanidins (PA) and hydrolysable tannins (gallo- and ellagitannins) (1,6).

Polyphenols are an integral part of the human diet. Antioxidant, antiinflammatory, anticarcinogenic, and other bioactivities demonstrated for various polyphenols suggest that they may have beneficial effects on human health and provide protection against such chronic diseases as cardiovascular diseases, neurodegenerative disorders, and cancers (3–7). Scientific evidence of the role of polyphenol consumption in disease prevention is promising, but not conclusive, and more clinical trials and epidemiologic studies are needed. Intake estimations of specific polyphenol groups have been published for phenolic acids (8,9), flavonoids (3-5,9-11), anthocyanidins (4,9,11,12), and PA (11,13). In some studies (14,15), the intake estimations of total polyphenols have been based on results obtained using unspecific spectrophotometric methods. Only 2 studies (4,11) have reported the intakes of various polyphenol groups calculated from individual food consumption data, but neither of these includes phenolic acids or ellagitannins.

In our study, concentrations of phenolic acids (16–18), anthocyanidins (19), and other flavonoids (20), PA (21), and ellagitannins (19), derived from analyses of Finnish foods, were incorporated into the food composition database. We estimated the intake of these polyphenols and determined the dominating types of polyphenols and individual compounds and their major dietary sources. Correlations between the intakes of polyphenols and carotenoids, fiber, sterols, and some vitamins were also determined. To our knowledge, this is the first report providing intake estimation for such a wide range of dietary polyphenols.

Methods

Sampling and food analyses. The selection of food samples collected for the analyses included most of the key vegetable foods consumed by

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adults in the FINDIET 2002 Study (22). The food samples were collected in 2003-2005 and analyses of polyphenol concentrations were carried out for a total of 143 food samples at the laboratories of MTT AgriFood Research Finland and the University of Kuopio (16-21). The food samples, including berries, fruits, vegetables, grain products, and beverages, were purchased from 10 retail stores belonging to the 3 major food chains in 3 regions (Forssa, Helsinki, and Kuopio) in different parts of Finland. The analyses were carried out in edible parts of the food items and for each food a pooled sample was prepared (16–19).

The concentrations of total phenolic acids were determined by reversed-phase HPLC after alkaline and acid hydrolyses (16-18). Anthocyanidins were extracted as glycosides with acidic methanol and quantified for the weight of aglycone by reversed-phase HPLC (19). Other flavonoids were quantified as aglycones (20) and ellagitannins as ellagic acid (19) with reversed-phase HPLC after acid hydrolysis. Soluble flavan-3-ols (catechins and PA) were extracted with a mixture of acetone-methanol-water, purified using solid-phase extraction columns, and quantified with normal-phase HPLC according to the degree of polymerization. For quantification, an external standard consisting of PA oligomers isolated from Saskatoon berries was used along with commercially available monomeric and dimeric PA. Bound PA was hydrolyzed from the extract residue by thioacidolysis, and the flavan-3ols (terminal units) and flavan-3-ol-thioethers (extender units) thus obtained were quantified with reversed-phase HPLC (21).

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Entering polyphenol values into the food composition database. The analytical results for 44 individual polyphenol compounds and the aggregated sums for phenolic acids, anthocyanidins, other flavonoids, PA, and ellagitannins were transferred from an Excel file into the Finnish national food composition database (FCDB; 23). For 110 food items, the analyzed values were directly entered into FCDB (Supplemental Table 1). For 12 foods with analyzed concentrations available for a variety of samples or samples by year, arithmetic means were calculated. For 21 food items, values were completed with some data from other sources (8,24,25). All imputed values were agreed jointly by the analysts and the compilers. Quality indices were determined for each stored value in compliance with the USDA data quality evaluation system (26). The analyzed values (including those imputed from analyzed values) covered 67% of phenolic acid values, 29% of anthocyanidin values, 59% of proanthocyanidin values, and 19% of ellagitannin values in vegetable foods of the FCDB. The remaining food items and mixed dishes had imputed or estimated values according to recipe data and ingredients.

Concentrations of flavones (apigenin and luteolin) and flavonols (quercetin, myricetin, and kaempherol) were mainly derived from Dutch data (27) supplemented with some Finnish analytical results (20,28). Concentrations for isoflavonoids and lignans were based on the results published by Mazur et al. (29). Concentrations for carotenoids (30), fiber, sterols (31), energy, and vitamins originated from the Finnish FCDB (23).

TABLE 1 Intakes of aggregated polyphenol groups and main individual compounds in adults (FINDIET2002)¹

| | Women | Men | All | | | | |
|------------------------------------|-------------------------|---|-------------------------|--|--|--|--|
| | mg/d (mg/MJ) | | | | | | |
| Phenolic acids, total ² | 571 ± 297 (92 ± 54) | 725 ± 414** (83 ± 49**) | 641 ± 363 (88 ± 52) | | | | |
| Caffeic acid | 359 ± 271 | 487 ± 368** | 417 ± 325 | | | | |
| Ferulic acid | 103 ± 47 | 139 ± 67** | 120 ± 60 | | | | |
| Gallic acid | 36 ± 59 | $29 \pm 55^{**}$ | 33 ± 57 | | | | |
| p-Coumaric acid | 15 ± 6.5 | 17 ± 8.5** | 16 ± 7.5 | | | | |
| Sinapic acid | 10 ± 5.3 | 12 ± 7.6** | 11 ± 6.6 | | | | |
| Anthocyanidins, total ³ | 53 ± 76 (8 ± 12) | 43 ± 82** (5 ± 12**) | 47 ± 79 (7 ± 12) | | | | |
| Cyanidin | 25 ± 31 | 21 ± 32** | 23 ± 32 | | | | |
| Delphinidin | 14 ± 25 | 12 ± 29* | 13 ± 27 | | | | |
| Flavonoids, total | 37 ± 44 (6.0 ± 7.5) | 27 ± 43** (3.2 ± 5.3**) | 33 ± 43 (4.7 ± 6.7) | | | | |
| Flavonols ⁴ | 5.8 ± 4.6 | 5.0 ± 5.1** | 5.4 ± 4.9 | | | | |
| Flavanones ⁵ | 31 ± 43 | 22 ± 42** | 27 ± 43 | | | | |
| PA, total | 139 ± 121 (21 ± 18) | 115 ± 118** (13 ± 14**) | 128 ± 120 (18 ± 17) | | | | |
| PA monomers ⁶ | 13 ± 13 | $10 \pm 13^{**}$ | 12 ± 13 | | | | |
| PA dimers | 14 ± 14 | 12 ± 14** | 13 ± 14 | | | | |
| PA trimers | 12 ± 14 | 10 ± 13** | 11 ± 13 | | | | |
| PA, 4–6-mers | 22 ± 26 | 18 ± 25** | 20 ± 25 | | | | |
| PA, 7–10-mers | 6 ± 8 | 5 ± 8** | 6 ± 8 | | | | |
| PA, >10 polymers | 24 ± 28 | 17 ± 24** | 21 ± 27 | | | | |
| Bound PA | 49 ± 48 | 42 ± 51** | 46 ± 50 | | | | |
| Ellagitannins | 15 ± 43 (3 ± 8) | 8 ± 28** (1 ± 3**) | 12 ± 37 (2 ± 6) | | | | |
| lsoflavonoids ⁷ | 1.0 ± 4.8 (0.2 ± 0.7) | $0.8 \pm 2.6^{**} (0.1 \pm 0.3^{**})$ | 0.9 ± 3.9 (0.1 ± 0.6) | | | | |
| Lignans ⁸ | 1.4 ± 7.5 (0.22 ± 1.12) | $0.4 \pm 2.1^{**} (0.04 \pm 0.26^{**})$ | 0.9 ± 5.7 (0.14 ± 0.85) | | | | |
| Polyphenols, total ⁹ | 817 ± 368 (130 ± 66) | 919 ± 458** (105 ± 54**) | 863 ± 415 (119 ± 62) | | | | |
| Carotenoids ¹⁰ | 6.2 ± 4.6 (1.0 ± 0.8) | 5.6 \pm 4.7 (0.6 \pm 0.6**) | 5.9 ± 4.7 (0.8 ± 0.7) | | | | |
| Sterols ¹¹ | 322 ± 398 (49 ± 58) | 424 ± 453** (48 ± 56**) | 368 ± 427 (48 ± 57) | | | | |

¹ Values are means \pm SD, n = 2007 (1095 women and 912 men). Symbols indicate different from women: *0.01 < P < 0.05, **P < 0.01.

² Includes protocatechuic, vanillic, p-hydroxybenzoic, syringic, and miscellaneous acids.

³ Includes pelargonidin, petunidin, peonidin, and malvidin. ⁴ Isorhamnetin, kaempherol, myricetin, and quercetin.

⁵ Eriodictyol, hesperetin, and naringenin.

⁶ Proanthocyanidin monomers = epicatechin, catechin (= flavan-3-ols).

⁷ Biochanin A, daidzein, genistein, glycitein, and formononetin.

⁸ Matairesinol and secoisolariciresinol.

⁹ Includes phenolic acids, anthocyanidins, flavonoids, proanthocyanidins, ellagitannins, isoflavonoids, and lignans.

 10 Includes α -carotene, β -carotene, canthaxanthin, capsantine, cryptoxanthin, γ -carotene, lutein, lycopene, and zeaxanthin.

¹¹ Includes avenasterol, brassicasterol, campesterol, sitosterol, and stigmasterol.

Conversion of dietary data to polyphenol intakes. A cross-sectional population survey (32) was carried out to assess the risk factors of cardiovascular disease. A random sample (n = 12,000) of adults aged 25– 64 y, stratified by sex, region, and 10-y age group, was drawn from the national population register for 6 regions in Finland. Subjects were invited to visit a health clinic and asked to complete a questionnaire for background information. The study design was approved by the Ethics Committee of the National Public Health Institute and an informed written consent was obtained from each participant. Subjects in a random subsample (n = 3181) from 5 regions of the population survey were asked to participate in a 48-h dietary interview (FINDIET 2002; 22). Of those invited, 64% participated and 98% of interviews were subsequently accepted (n = 2007). Food consumption data were converted into intakes of polyphenol compounds for each individual by means of in-house software together with the national FCDB (23). In addition, major food sources for polyphenols, carotenoids, and sterols were identified.

Statistical methods. The mean intake of polyphenols was calculated for each person based on dietary data of 2 consecutive days. The aggregated mean \pm SD intake of each polyphenol group (phenolic acids, anthocyanidins, flavones, flavonols, flavanones, PA, ellagitannins, isoflavonoids, and lignans), the total intake of polyphenols, and the intakes of 14 individual compounds were determined. Intakes for individual compounds are presented under the relevant polyphenol group (Table 1) when intakes of men and women differed. SAS Statistical Package (version 8.2, SAS Institute, 1999) was used for the analyses. The intakes were transformed into logarithmic values for statistical analyses. Differences in intakes by gender were tested by Student's *t* test for independent samples and differences below the probability level (P < 0.05) were considered significant. Pearson correlation coefficients between the intakes of polyphenols and other dietary compounds were calculated.

Results

Polyphenol concentrations. Among the 143 food items analyzed, berries were superior in terms of their polyphenol concentrations (16–21; **Supplemental Table 1**). In the list of 20 foods with the highest total polyphenol concentrations, 16 berries were included (**Fig. 1**). Besides berries, some foods with high dry matter concentrations appeared on the list, whereas fruits and vegetables did not. Among fruits, high polyphenol concentrations (>100 mg/100 g) were found in dark plums, cherries, apples, and dark grapes. In vegetables, the highest concentrations were detected in rhubarb and red cabbage. In addition to bran (Fig. 1), rye and graham flours were good sources of polyphenols in cereals. In beverages, the highest polyphenol concentration was in coffee.

The dominant polyphenol groups as well as their concentrations varied greatly in the foods analyzed (Fig. 1). Brans and other cereal products had the highest concentrations of phenolic acids (17). Peanuts, coffee, many berries, and some vegetables also contained high levels of phenolic acids (16,18). Anthocyanidin concentrations were highest in blue and black berries, whereas red berries and all other samples had lower concentrations (19). Cocoa powder and chokeberry (*Aronia mitchurini*) had the highest concentrations of PA, but considerable concentrations were also detected in several berries, stone fruits, and peanuts (our unpublished data). Some polyphenols were detected only in specific species such as ellagitannins in cloudberries, raspberries, strawberries, and rose hips (19).

Polyphenol intake. The mean total intake of polyphenols was 863 mg/d (Table 1). The main polyphenol groups were phenolic acids (75% of total intake of polyphenols) and PA (14%), whereas anthocyanidins (6%) and other flavonoids (5%) accounted for lower proportions. Men had higher absolute intakes of total polyphenols and acids than women (Table 1), but women had



FIGURE 1 The 20 food items with the highest total concentrations of polyphenols including phenolic acids, anthocyanidins, other flavonoids, PA, and ellagitannins. Values are means of triplicate measurements of pooled samples (2–10 subsamples per pool).

higher intakes of the other polyphenol groups. Women had higher intakes per energy unit (1 MJ) than men (Table 1).

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The individual compounds with the highest intakes were caffeic acid in phenolic acids and cyanidin in anthocyanidins (Table 1). The soluble forms of PA contributed most to the total intake of PA but the intake of bound PA was also noteworthy. The mean daily intakes of flavonols, flavanones, and flavones were low compared with phenolic acids. The aggregated intake of flavonoids (i.e. anthocyanidins, PA monomers, flavonols, flavanones, flavonos, and isoflavonoids) was 103 mg/d in women and 80 mg/d in men. The intake of sterols was about one-half of the total polyphenol intake, whereas the intake of carotenoids was much below the total polyphenol intake (Table 1).

Polyphenol food sources. Coffee was the primary food item contributing to phenolic acid intake (Table 2) and also to the total intake of polyphenols. Other main contributors consisted of cereals (especially rye bread), tea, and fruits. Berries and berry products, including berry-based sweetened drinks, were the main anthocyanidin contributors. Bilberries (*Vaccinium myrtillus*) were the main food source for anthocyanidins. The most important contributors to the intake of flavonols, flavanones, and flavones were fruit (especially apple and citrus fruit) and tea. The main contributors to PA intake were apples, berries, tea, and chocolate (Table 2). Vegetables were of minor importance to polyphenol intakes. Carotenoids were obtained mainly from vegetables and sterols from margarines, bread, and fruit. Isoflavonoids came from soy products and lignans were derived from seeds, soy products, rye bread, and other cereal products (data not shown).

Correlations. The intake of the predominant phenolic group, phenolic acids, correlated with the intakes of carotenoids, fiber, and vitamin C and also with ellagitannins and some flavonoids (**Supplemental Table 2**). The intakes of anthocyanidins and PA correlated with each other and with other polyphenols, except

TABLE 2 Contribution of food groups to intakes of polyphenols, carotenoids, and sterols in 2007 Finnish adults

| Food group | Phenolic acids | Anthocyanidins | Flavonoids | Proanthocyanidins | Ellagitannins | Polyphenols, total | Carotenoids | Sterols |
|----------------|----------------|----------------|------------|-------------------|---------------|--------------------|-------------|-------------------|
| | | | | % of tota | I | | | |
| Fruit | 2.2 | 3.1 | 73.9 | 33.9 | | 5.6 | 3.0 | 4.7 |
| Fruit juice | 0.8 | 1.6 | 10.1 | 1.4 | 0.9 | 1.2 | 0.6 | 1.4 |
| Berries | 0.5 | 28.0 | 1.3 | 9.7 | 64.3 | 1.6 | 0.3 | 0.4 |
| Berry dishes | 1.2 | 47.3 | 4.9 | 24.5 | 34.7 | 1.9 | 1.0 | 2.8 |
| Berry drinks | 0.2 | 12.6 | 1.5 | 13.0 | | 0.3 | 0.4 | |
| Vegetables | 2.1 | 1.6 | 2.5 | 0.4 | | 1.8 | 82.1 | |
| Potato | 1.7 | | | | | 1.6 | 1.0 | 3.0 |
| Bread, cereals | 12.3 | | | 2.0 | | 11.5 | 4.8 | 26.1 |
| Chocolate | <0.1 | 0.2 | | 6.4 | 0.1 | 0.04 | | 1.0 |
| Coffee | 67.9 | | | | | 63.3 | | |
| Теа | 9.7 | | 4.5 | 3.4 | | 9.3 | | |
| Wine | 0.4 | 2.4 | 0.1 | 2.2 | | 0.3 | | 0.3 |
| Other foods | 1 | 4.8 | 1.2 | 3.1 | 0 | 1.6 | 2.7 | 42.7 ¹ |

¹ Mainly from sterol-enriched margarines.

isoflavonoids. Vitamin C and fiber intakes correlated with intakes of all polyphenols. The highest correlation was identified between intakes of flavanones and vitamin C. The intakes of isoflavonoids and lignans as well as those of vitamin E and sterols correlated with each other.

Discussion

The most comprehensive polyphenol databases currently are those of the USDA website (24,25), containing PA in vegetables, fruits, and beverages, and flavonoids (including anthocyanidins) in almost 400 food items. However, data for quantitative intake estimation of all important polyphenol groups are limited. Our study incorporated polyphenol concentrations for a large variety of polyphenols, as well as phenolic acids and ellagitannins, into the food composition database. The coverage of polyphenol profiles in consumed food items has been considered an essential step in epidemiological studies to determine the importance of polyphenols compared with other phytochemicals (5).

For evaluation of the intake of total polyphenols, no comparable data from other studies are available. According to the study by Saura-Calixto et al. (15), where polyphenols were analyzed using unspecific spectrophotometric methods, the mean daily intake of total polyphenols ranged from 2600 to 3000 mg. These figures are much higher than those obtained here, probably due to analytical differences. Manach et al. (2) estimated that the total polyphenol intake probably reaches 1 g/d in people who eat several servings of fruits and vegetables daily. This estimation is close to the intake obtained in our study. Beverages and cereals are the most important contributors to total polyphenol intake, a finding also observed by Saura-Calixto et al. (15).

Contrary to some earlier intake estimations of polyphenols (3,4,8–12,14), intakes from cereals were included in our intake estimations and they affected total polyphenol intake and intake of phenolic acids. However, intakes of polyphenol compounds reflect individual preferences, e.g. in coffee, bread, or berry consumption, and the variation between individuals was high. A German study (8) estimated daily intake of phenolic acids at 222 mg, which was lower than our estimate. However, polyphenol concentrations in their study were derived from the literature, were based on a variety of analytical methods, and did not take into account cereal products, which are a very good source of phenolic acids (17). Consistent with our results, coffee was the best source of phenolic acids. In FINDIET 2002 (23), 86% of

women and 91% of men consumed coffee (with means of 450 mL/d and 600 mL/d, respectively).

Anthocyanidin intakes were higher in the Finnish diet than reported elsewhere (4,12). This may partly be due to the berry consumption in Finland, with the mean consumption of berries in adults being 52 g/d (23). Differences may also exist in the databases, e.g. in the coverage of anthocyanidin values. The aggregated intake of anthocyanidins and other flavonoids gave a more than 100% higher estimate (190 mg/d) in the US (4) than in Finland (80 mg/d), which may be due to different consumption of such flavonoid contributors as beans or soy.

Contrary to scant studies on phenolic acids, anthocyanidins, and PA, numerous studies (3–6,9–11) have estimated daily intakes for flavonols, flavones, and flavanones. In our study, the intake estimation for flavonols, flavones, and flavanones was higher than in a previous Finnish study (24 mg/d; 10), which likely is due to increased vegetable and fruit consumption. The intake of these compounds was higher in women than in men, supporting an earlier finding (4).

The intake of PA was higher in our study (128 mg/d) than in previous estimations for American adults (58 mg/d; 13). The methodology of soluble PA assessments was very similar and PA forms larger than trimers were the predominant soluble procyanidins in both studies. However, our study also covered bound PA, which represented \sim 38% of the total PA intake. The major sources of PA in the American diet were apples (32%), chocolate (18%), and grapes (18%) (13), whereas the sources of PA in Finland were berries and berry products (45%) followed by fresh fruits (34%, especially apples) and chocolate (7%). Chocolate is considered an important source of antioxidants and PA; a chocolate bar may contain levels up to 400 mg (33). The daily intake of ellagitannins (12 mg/d) was higher in Finland than in Germany (5 mg/d) (8). Berries were the main contributors of ellagitannins in both studies.

Although analytical results for some processed foods (e.g. berry drinks and jams) were available, retention factors for losses of polyphenols due to domestic and industrial processing in other dishes were not examined. Many processes, such as storing, cooking, and peeling, can cause variable losses in the concentrations of polyphenols (2). This may cause some overestimation of intakes. Therefore, we present cooked contributors of polyphenols separately from fresh food items (Table 2). On the other hand, although polyphenol concentrations decrease during processing, the bioavailability of these compounds

may improve (1). These aspects are important in, for instance, berry dishes as major sources of anthocyanidins, PA, and ellagitannins (Table 2). However, our intake figure for total polyphenols is the most reliable estimate currently available.

Polyphenols originate from plants and their intakes are expected to correlate with each other. The intakes of polyphenol groups were also associated with other phytochemicals, fiber, and vitamins in the Finnish diet, indicating the same food sources. Vitamin C correlated especially highly with flavanones, because all of these compounds exist in large amounts in citrus fruits. However, the intakes of isoflavonoids and sterols were not associated with the intakes of other phytochemicals due to different food sources.

As sources of polyphenols, berries, fruits, and cereals are preferred among the recommended foods groups. Berries contained very high concentrations of polyphenols. Some lignans and resorcinols from cereals (17) were not included in our intake calculation; their inclusion would further improve the position of cereals. In Finland, rye bread contributes greatly to the intake of phenolic acids. Commonly consumed vegetables were only minor contributors to the total polyphenol intake. However, vegetables have other recommended nutrient profiles. Polyphenol absorption has been confirmed by biomarkers, e.g. urinary excretion (2,6,34), but more knowledge of the metabolism of the various polyphenols is needed. The absorption of polyphenols may depend on the compound, food material, and processing (1,2,7,15). Polyphenols bound to fiber fractions of food items, for instance, may be less bioavailable (1,15,34).

To summarize, this study combines complete polyphenol concentrations with individual food consumption data for accurate intake estimations. The total intake of polyphenols was substantial, although interindividual variation was high. The main contributors to total polyphenol intake were coffee and cereal products, and phenolic acids were the main group of compounds. In the future, the polyphenol database compiled for this study may serve as a valuable tool for the purpose of clarifying the relationships between polyphenol intake, biomarkers, and chronic diseases.

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