Human Nutrition and Metabolism

Several Culinary and Medicinal Herbs Are Important Sources of Dietary Antioxidants[1]

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ABSTRACT We assessed the contribution of culinary and medicinal herbs to the total intake of dietary antioxidants. Our results demonstrate that there is more than a 1000-fold difference among antioxidant concentrations of various herbs. Of the dried culinary herbs tested, oregano, sage, peppermint, garden thyme, lemon balm, clove, allspice and cinnamon as well as the Chinese medicinal herbs Cinnamomi cortex and Scutellariae radix all contained very high concentrations of antioxidants (i.e., >75 mmol/100 g). In a normal diet, intake of herbs may therefore contribute significantly to the total intake of plant antioxidants, and be an even better source of dietary antioxidants than many other food groups such as fruits, berries, cereals and vegetables. In addition, the herbal drug, Stronger Neo-Minophagen C, a glycyrrhizin preparation used as an intravenous injection for the treatment of chronic hepatitis, boosts total antioxidant intake. It is tempting to speculate that several of the effects due to these herbs are mediated by their antioxidant activities. J. Nutr. 133: 1286–1290, 2003.

KEY WORDS: • *antioxidants* • *herbs* • *reductants* • *humans* • *oxidative stress*

Herbs are most often defined as any part of a plant that is used in the diet for its aromatic properties (1–3). Recently, however, herbs have also been identified as sources of various phytochemicals, many of which possess important antioxidant activity (4–6).

Spices are dried herbs, and condiments are spices and other flavorings added to food at the table (1–3). Since prehistoric times, herbs have also been the basis for nearly all medicinal therapy until synthetic drugs were developed in the 19th century. Today, herbs are still found in 40% of prescription drugs (1–3). In addition, herbs are used for many other purposes including beverages such as tea, dyeing, repellents, fragrances, cosmetics, charms, smoking and industrial uses.

In previous studies, three different methods have been used to assess total antioxidant capacity of food. The 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid equivalent antioxidant capacity assay of Miller et al. (7), the ferric reducing ability of plasma (FRAP)[3] assay of Benzie and Strain (8) and the oxygen radical absorbance capacity assay of Glazer's laboratory (9) and others (10). We elected to use the FRAP analysis for several reasons [see discussion in (11)]. A major advantage of the FRAP method is its ability to yield absolute quantitative determination of the amounts of total antioxidants or reductants in samples such as foods, extracts, supplements or tissues. Thus, values generated by the FRAP method can be used to calculate the total intake of antioxidants and the contribution of various food groups for total dietary intake. In this study, we evaluated a variety of herbs for their total antioxidant content to elucidate whether intake of herbs is a significant contributor to antioxidant intake.

MATERIALS AND METHODS

Reagents. The reagent, 2,4,6-tri-pyridyl-s-triazine (TPTZ), was obtained from Fluka Chemie AG (Deisenhofen, Switzerland), sodium acetate trihydrate and $FeSO_4 \cdot 7\ H_2O$ from Riedel-deHaën AG (Seelze, Germany), acetic acid and hydrochloric acid from Merck (Darmstadt, Germany) and $FeCl_3 \cdot 6H_2O$ from BDH Laboratory Supplies (Dorset, England). MilliQ water (Millipore, Bedford, MA) and methanol of HPLC-grade obtained from Merck were used for all extractions; 2-propanol (HPLC-grade) was obtained from Merck.

Automated FRAP assay. The ferric reducing ability of plasma (FRAP) assay of Benzie and Strain (8) was used with minor modifications that allowed quantization of both water- and fat-soluble antioxidants (11). A Technicon RA 1000 system (Technicon Instruments, New York, NY) was used for the measurements of absorption changes that appear when the TPTZ-Fe^{3+} complex reduces to the TPTZ-Fe^{2+} form in the presence of antioxidants. An intense blue color with an absorption maximum at 593 nm develops. The measurements were performed at 600 nm. An aqueous solution of 1000 μmol/L $FeSO_4 \cdot 7\ H_2O$ was used for calibration of the instrument. The FRAP assay was fully validated in an earlier report (11). All values are means of triplicate analyses. The within-day repeatability measured as relative SD (RSD) ranged from 0.4 to 6%. The variations in the FRAP values for replicate items obtained from the same source were typically between 3 and 10 RSD%. Occasionally, some items

[1] Supported by grants from the Norwegian Research Council, the Norwegian Cancer Society, The Novo Nordic Foundation and the Throne Holst Foundation.

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[3] Abbreviations used: FRAP, ferric reducing ability of plasma; RSD, relative SD; TPTZ, 2,4,6-tri-pyridyl-s-triazine.

Manuscript received 24 November 2002. Initial review completed 23 December 2002. Revision accepted 16 January 2003.

TABLE 1

Total antioxidants in culinary herbs

Herb species and variety	Total antioxidants	Mean values
	mmol/100 g	
Oregano (*Origanum vulgare*)		137.5
Muster (Carl Sperling, Lüneburg, Germany)	153.8	
Wild form grown at Kise	149.3	
Wild form (Jelitto, Schwarmstedt, Germany)	142.3	
Greek Oregano (Jelitto, Schwarmstedt, Germany)	139.3	
Dost (Chrestensen, Erfurt, Germany)	139.2	
Album (Jelitto, Schwarmstedt, Germany)	138.6	
Greek Oregano (Johnny's, Winslow, Maine)	100.0	
Sage (*Salvia officinalis*)		91.2
Extrakta (Richters, Ontario, Canada)	101.9	
Extrakta (Chrestensen, Erfurt, Germany)	88.5	
Nr 934 (Johnny's, Winslow, Maine)	83.3	
Peppermint, leaves (*Mentha* × *piperita*)		78.5
Kise No 7 (Norway)	85.0	
Kise No 8 (Norway)	81.1	
Kise No 4 (Norway)	69.3	
Garden Thyme (*Thymus vulgaris*)		74.6
English Thyme (Richters, Ontario, Canada)	95.0	
German Winter (Johnny's, Winslow, Maine)	83.4	
French Thyme (Richters, Ontario, Canada)	45.4	
Lemon Balm, leaves (*Melissa officinalis*)		74.6
Unnamed variety	102.4	
8210 sample no 8 (Leüthen, Norway)	76.5	
Quedlinburger Niderligende (Quedlinburger, Germany)	44.9	
Sweet Marjoram (*Origanum majorana/ Majorana hortensis*)		55.8
Marietta (Quedlinburg, Germany)	69.2	
Erfo (Chrestensen, Erfurt, Germany)	42.3	
Hyssop, leaves (*Hyssopus officinalis*)		37.8
Schira Weiss (Quedlinburg, Germany)	49.8	
Schira Rosa (Quedlinburg, Germany)	40.1	
Piikkiö (Piikkiö, Finland)	37.7	
Schira Blau (Quedlinburg, Germany)	31.3	
Blaublühender (Chrestensen, Erfurt, Germany)	30.0	
Tansy, leaves (*Tanacetum vulgare*)		36.4
Goldsticks (Canada)	41.8	
Framnes (Norway)	41.6	
Steinvikholmen (Norway)	35.3	
Richters (Ontario, Canada)	34.8	
Alvdal (Norway)	28.7	
Anise Hyssop (*Agastache foeniculum*)		34.4
Seed lot 933 (Johnny's, Winslow, Maine)	34.4	
Tansy, flower (*Tanacetum vulgare*)		32.4
Steinvikholmen (Norway)	42.8	
Framnes (Norway)	36.0	
Richters (Ontario, Canada)	29.4	
Alvdal (Norway)	28.2	
Goldsticks (Canada)	25.6	
Purple Coneflower, leaves (*Echinacea purpurea*)		29.5
S2280 (Richters, Ontario, Canada)	29.5	
Purple Coneflower, flowers (*Echinacea purpurea*)		23.2
S2280 (Richters, Ontario, Canada)	23.2	
Roseroot/Arctic root, stem (*Rhodiola rosea* ssp. *Rosea*)		18.3
Alvdal (Norway)	19.7	
Hofstad I (Norway)	17.8	
Vesterålen (Norway)	17.4	
German Chamomile (*Chamomilla recutita*)		17.7
Bona (Johnny's, Winslow, Maine)	17.8	
Bodegold (Johnny's, Winslow, Maine)	17.5	
European Angelica, leaves/stem (*Angelica archangelica* spp. Archangelica)		14.7
Lechner (Germany)	20.2	
Aicher (Germany)	18.8	
Heyland (Germany)	15.8	
Gaissmayer (Germany)	15.7	
Kretz (Germany)	15.1	
Sørlandet (Norway)	13.2	
Hungarian	12.0	
Ringsaker (Norway)	11.3	
Voss (Norway)	10.0	
Roseroot/Arctic root, leaves (*Rhodiola rosea* ssp. *Rosea*)		5.6
Alvdal (Norway)	7.0	
Hofstad (Island)	4.1	
Vesterålen (Norway)	5.8	
European Angelica, root (*Angelica archangelica* spp. Archangelica)		5.1
Ringsaker (Norway)	7.9	
Gaissmayer (Germany)	5.9	
Kretz (Germany)	5.7	
Lechner (Germany)	5.1	
Aicher (Germany)	5.0	
Heyland (Germany)	3.9	
Hungarian	3.8	
Sørlandet (Norway)	3.6	
Coriander (*Coriandrum sativum*)		3.3
Thüringer (Chrestensen, Erfurt, Germany)	3.6	
Jantar (Chrestensen, Erfurt, Germany)	3.3	
Santo (Chrestensen, Erfurt, Germany)	3.1	

had larger variation. In such cases, the FRAP values were confirmed by reanalysis.

Collection of culinary herbs. Most of the plants were grown from seed, and transplanted from a greenhouse to a field at Kise in South Norway, (60°47′ N, 10°49′ E). Leaves, stems, flowers, seed and roots were harvested and dried in the dark at 35°C. The samples were stored at ~20°C in paper bags until analysis in fall 2001. The only exceptions were roseroot and European angelica, which also were analyzed as fresh material.

There is a wide variation in the oregano plant family. One of the oreganos in this test was collected as seed from wild growing plants at Kise in Norway, whereas the others were obtained from the seed companies indicated. Roseroot was propagated from cuttings, raised under glass and transplanted. Propagation of peppermint was also from the cuttings of green shoots. The varieties tested all belong to the red peppermint family and have been grown in Norway for many years without being named. They are now maintained in a collection at Kise. Tansy is a wild herb that grows in Norway. Plants were collected from three districts and grown on the same field at Kise as tansy propagated from seed from Richters, Goodwood, ON, Canada. Roseroot is also a native plant of Norway. One plant from each of three districts was propagated by stem cuttings and grown at Kise for 3 y before harvesting. Two varieties of chamomile and three varieties of coriander were grown and harvested at Kise. European angelica, which is native to Norway, was collected from plants in two districts.

Chinese and Japanese medicinal herbs. All Chinese medicinal herbs were generously donated by Tsumura, Tokyo, Japan. Dried extracts were prepared by the pharmaceutical company by boiling dried plants with 12 times the amount of water (wt/wt) for 1 h, filtration and concentration of supernatant under reduced pressure (40°C) followed by drying in a spray dryer (50°C). The Japanese herb-based drug, Stronger Neo-Minophagen C, was generously donated by Minophagen Pharmaceutical, Tokyo, Japan.

RESULTS

Total antioxidants in culinary herbs. We assessed total antioxidants in several herbs grown at Apelsvoll Research Centre Division Kise, Hedmark, Norway. For most of the herbs, several varieties were tested. For some herbs, antioxidant concentrations were also tested in samples harvested at different times of the year, or in different years.

Of the dried herbs tested, oregano, sage, peppermint, garden thyme and lemon balm all contained very high levels of antioxidants (**Table 1**) (between 75 and 138 mmol/100 g). Sweet marjoram, hyssop, tansy, anise hyssop and purple coneflowers were intermediate (between 23 and 56 mmol/100 g). Roseroot stem, German chamomile, European angelica and coriander all contained relatively low concentrations of antioxidants (between 1 and 18 mmol/100 g).

There was also some variation among different varieties of the same herb species (Table 1). We tested 7 different forms of oregano. All varieties contained very high levels of total antioxidants. The varieties Muster from Carl Sperling GmbH & Co (Lüneburg, Germany), the wild form grown at Kise, and the wild form obtained from Jelitto (Schwarmstedt, Germany) all contained >140 mmol/100 g. Of the oregano varieties tested, Greek oregano from Johnny's (Winslow, Maine) had the lowest concentration (i.e., 100 mmol/100 g).

The sage varieties also contained different levels of antioxidants. The varieties Extrakta from Chrestensen (Erfurt, Germany), Richters (Goodwood, Canada) and Nr 934 from Johnny's (Winslow, Maine) contained 102, 89 and 83 mmol/100 g, respectively.

Seasonal variation in herb antioxidant contents. To test seasonal variation, we collected herbs in two different seasons. For three of the varieties collected in 1998 and 2000, the total antioxidant concentration was highest in those collected in 1998 (**Table 2**). Furthermore, 7 of 8 varieties of European Angelica contained more antioxidants in one season than another (i.e., 2000 vs. 2001). Thus, there is a definite seasonal trend in antioxidant content. We also observed variation in items collected in different parts of the season because 5 of 7 herb varieties collected in the fall contained higher values than when collected earlier in the fall.

Total antioxidants in commercial spices. Of the commercial spices analyzed, clove and allspice/pimento both contained > 100 mmol total antioxidants/100 g, whereas cinnamon, rosemary, thyme and marjoram contained between 50 and 100 mmol/100 g (**Table 3**).

Total antioxidants in medicinal herbs. High concentrations of antioxidants were also detected in many of the Chinese and Japanese herbs tested (**Table 4**). Cinnamomi Cortex and Scutellariae Radix both contained >100 mmol/100 g, whereas Cimicifugae Rhizoma, Paeoniae Radix and Glycyrrhizae Radix contained between 10 and 65 mmol/100 g.

Saiko-Keishi-To, Juzaen-Taiho-To and Hocyu-Ekki-To are three Chinese herbal medicines that are composed of various combinations of these herbs. The herb medicines Saiko-Keishi-To, Juzaen-Taiho-To contained 21.4 and 14.2 mmol/100 g, respectively, whereas Hocyu-Ekki-To contained somewhat lower levels (9.7 mmol/100 g). The herbal drug, Stronger Neo-Minophagen C, which is an intravenous drug containing glycyrrhizin, contained 1.0 mmol/100 g.

TABLE 2

Seasonal variation of antioxidants in culinary herbs[1]

Herb species and variety	1998	2000	2001
		mmol/100 g	
Oregano, wild form grown at Kise (Norway)	160.0	138.5	
Oregano, wild form (Jelitto, Schwarmstedt, Germany)	134.4	150.2	
Oregano, Greek (Jelitto, Schwarmstedt, Germany)	165.0	113.5	
Garden Thyme, English Thyme (Richters, Ontario, Canada)	103.0	87.0	
European Angelica, leaves/stem, Lechner, Germany		26.2	14.2
European Angelica, leaves/stem, Aicher, Germany		24.1	13.5
European Angelica, leaves/stem, Heyland, Germany		20.4	11.1
European Angelica, leaves/stem, Gaissmayer, Germany		19.8	11.6
European Angelica, leaves/stem, Kretz, Germany		16.1	14.0
European Angelica, leaves/stem, Hungarian		14.1	9.9
European Angelica, leaves/stem, Sørlandet (Norway)		13.0	13.3
European Angelica, leaves/stem, Ringsaker (Norway)		11.6	11.0
Sage, Extrakta (Chrestensen, Erfurt, Germany)	96.2, 98.3*		
Sage, Extrakta (Richters, Ontario, Canada)	99.4, 104.4*		
Peppermint, Kise 4 (Norway)		67.9, 59.8,* 80.1†	
Peppermint, Kise 7 (Norway)		78.0, 80.8,* 96.1†	
Peppermint, Kise 8 (Norway)		82.2, 84.9,* 76.2†	

1 Plants were harvested in summer except for * harvested in the fall and † harvested in the early fall.

TABLE 3

Total antioxidants in commercial herbs

Commercial spices (dried herbs)	Total antioxidants
	mmol/100 g
Clove (Black Boy, Elverum, Norway)	465.3
Allspice/pimento (Black Boy, Elverum, Norway)	101.5
Cinnamon (Santa Maria, Mölndal, Sweden)	98.4
Rosemary (Black Boy, Elverum, Norway)	66.9
Thyme (Black Boy, Elverum, Norway)	63.7
Marjoram (Santa Maria, Mölndal, Sweden)	53.9
Cinnamon (Black Boy, Elverum, Norway)	53.0
Saffron, red (Gaea, Agrinion, Greece)	47.8
Oregano (Black Boy, Elverum, Norway)	45.0
Tarragon (Black Boy, Elverum, Norway)	43.3
Common basil (Black Boy, Elverum, Norway)	30.9
Bayberry leaves (Black Boy, Elverum, Norway)	24.3
Ginger (Santa Maria, Mölndal, Sweden)	22.5
Nutmeg (Black Boy, Elverum, Norway)	20.3
Dill (Black Boy, Elverum, Norway)	15.9
Curry (Black Boy, Elverum, Norway)	13.0
Mustard (Colman's, Carrow, England)	10.4
Curcuma (Black Boy, Elverum, Norway)	10.3
Vanilla (Santa Maria, Mölndal, Sweden)	10.1
Juniper berry (Black Boy, Elverum, Norway)	9.3
Pepper, black (Black Boy, Elverum, Norway)	8.7
Chilipepper (Santa Maria, Mölndal, Sweden)	8.5
Jalapeno pepper (Black Boy, Elverum, Norway)	8.2
Vanilla (Black Boy, Elverum, Norway)	7.2
Chives (Black Boy, Elverum, Norway)	7.1
Cumin (Black Boy, Elverum, Norway)	6.8
Red pepper (Santa Maria, Mölndal, Sweden)	6.1
Piri Piri (Black Boy, Elverum, Norway)	6.0
Cayenne (Black Boy, Elverum, Norway)	5.9
Red pepper (Black Boy, Elverum, Norway)	5.6
Caraway seeds (Black Boy, Elverum, Norway)	4.5
Parsley (Black Boy, Elverum, Norway)	3.6
Coriander (Santa Maria, Mölndal, Sweden)	2.8
Vanilla seeds (Black Boy, Elverum, Norway)	2.6
Garlic (Black Boy, Elverum, Norway)	2.1
Coriander (Black Boy, Elverum, Norway)	2.1
Cardamom (Black Boy, Elverum, Norway)	0.5
Poppy seeds (Black Boy, Elverum, Norway)	0.3

DISCUSSION

We recently assessed systematically the total concentration of dietary antioxidants (i.e., electron- or hydrogen-donating antioxidants or reductants) in a variety of dietary plants including various fruits, berries, vegetables, cereals, nuts and pulses. Our results (11) demonstrated that there are large differences among total antioxidants in various dietary plants. Most berries, walnuts, sunflower seeds, ginger and pomegranates are among the high antioxidant dietary plants, whereas many of the dietary plants consumed contain few antioxidants. Fruits, berries, cereals and vegetables contribute ~0.9, 0.6, 0.3 and 0.2 mmol/d total antioxidants, respectively, in a typical Norwegian diet (11).

In the present study, we demonstrated that there is a greater than 1000-fold difference among total antioxidants in various herbs. Of the dried culinary herbs tested, oregano, sage, peppermint, garden thyme, lemon balm, clove, allspice and cinnamon all contained very high concentrations of antioxidants (i.e., >75 mmol/100 g). In a normal diet, intake of 1 g of these herbs may therefore make a relevant contribution (>1 mmol) to the total intake of plant antioxidants, and be an even better source of dietary antioxidants than many other food groups.

In previous studies, alternative methods to assess total antioxidant capacities have been used for some of the herbs included in the present study. In general, these studies (6,12–14) report similar rankings among the individual herbs, although their relative values are not easily compared to the absolute values in this report.

The Chinese herbs Cinnamomi Cortex and Scutellariae Radix both contained very high levels of antioxidants (i.e., >75 mmol/100 g). The Chinese herb medicines Saiko-Keishi-To, Juzaen-Taiho-To and Hocyu-Ekki-To, which are used for various kinds of inflammatory and infectious diseases, are all taken in daily doses of 7.5 g, corresponding to 1.6, 1.1 and 0.7 mmol antioxidants/d, respectively.

The antioxidant activity of the Japanese herb medicine Sho-saiko-to, which is composed of the herbs tested in this study, was calculated to be ~1.3 mmol/7.5 g (i.e., the daily dose). This drug, which is commonly used to treat chronic hepatitis in Japan, may also inhibit the development of hep-

TABLE 4

Total antioxidants in Chinese and Japanese medicinal herbs

Medicinal herbs	Botanical name	Total antioxidants
		mmol/100 g
Cinnamomi Cortex	*Cinnamomum cassia,* Blume	120.2
Scutellariae Radix	*Scutellaria baicalensis,* Georgi	111.5
Cimicifugae Rhizoma	*Cimicifuga simplex,* Wormskjold	64.3
Paeoniae Radix	*Paeonia lactiflor,* Pallas	55.1
Aurantii Nobilis Pericarpium	*Citrus unshiu,* Markovich	17.5
Glycyrrhizae Radix	*Glycyrrhiza uralensis,* Fisher/*Glycyrrhiza Glabra,* Linne	11.6
Zingiberis Rhizoma	*Zingiber officinale,* Roscoe	7.5
Atractylodis Lanceae Rhizoma	*Atractylodes Lancea,* De Candolle/*Atractylodes Chinenshis,* Koidzumi	7.4
Cnidii Rhizoma	*Cnidium officinale,* Makino	6.7
Zizyphi Fructus	*Zizyphus jujube* Miller var. *inermis,* Rehder	5.9
Bupleuri Radix	*Bupleurum falcatum,* Linne	5.7
Astragali Radix	*Astragalus mambranaceus,* Bunge/ *Astragalus mongholicus,* Bunge	4.9
Rhemanniae Radix	*Rhemanniae glutinosa,* Liboschitz	3.9
Angelicae Radix	*Angelicae acutiloba,* Kitagawa	3.0
Holelen	*Paria cocos,* Wolf	2.8
Ginseng Radix	*Panax ginseng,* C. A. Meyer	1.5
Pinelliae Tuber	*Pinelliae*	0.3
Medicinal herb product	Manufacturer	
Saiko-Keishi-To	Tsumura Company	21.4
Juzen-Taiho-To	Tsumura Company	14.2
Hocyu-Ekki-To	Tsumura Company	9.7
Stronger Neo-Minophagen	Minophagen Pharmaceutical Company	1.0

atocellular carcinoma (15), and reduce lipid peroxidation and hepatic fibrosis in experimental animals (16,17).

The herbal medicine, Stronger Neo-Minophagen C, a glycyrrhizin preparation, has also been used extensively with considerable success in Japan for the treatment of chronic hepatitis in intravenous doses up to 100 mL/d (18). Our analyses reveal that this injection volume equals ~1 mmol antioxidants. Thus, such injections will boost the total antioxidant intake. It is tempting to speculate that several of the effects observed with these herbal medicines are mediated by their antioxidant activity.

More data are required concerning bioavailability and bioactivity of herb antioxidants. However, these data represent a first crucial step that will be followed up in future studies. To perform such studies, we recently developed an in vivo transgenic mouse model for noninvasive detection of antioxidant activity (19). This model is based on the luciferase reporter regulated by various oxidative stress–related response elements. We intend to use this model to elucidate the role of dietary antioxidants in counteracting oxidative stress.

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