950

# Salicylates in foods

Anne R. Swain, Stephen P. Dutton, and A. Stewart Truswell<sup>1</sup> Human Nutrition Unit, University of Sydney, New South Wales, Australia

To determine salicylate content, 333 food items were analyzed. Foods were homogenized with 25% sodium hydroxide, allowed to stand overnight, acidified with concentrated hydrochloric acid, and then extracted with warm diethyl ether over 5 hours. The extract was dried and taken up in dilute sodium bicarbonate solution for analysis. Salicylic acid was separated by high performance liquid chromatography and quantified by reading at 235 nm. Salicylic acid standards were used throughout to standardize extractions and analyses. This is the most comprehensive set of data on food salicylates yet published; extraction appears to have been more complete for some foods, giving higher values than those previously published. Most fruits, especially berry fruits and dried fruits, contain salicylate. Vegetables show a wide range from 0 to 6 mg salicylate per 100 gm food (for gherkins). Some herbs and spices were found to contain very high amounts per 100 gm, e.g., curry powder, paprika, thyme, garam masala, and rosemary. Among beverages, tea provides substantial amounts of salicylate. Licorice and peppermint candies and some honeys contain salicylates. Cereals, meat, fish, and dairy products contain none or negligible amounts.

The Feingold diet for treatment of hyperactivity in children was devised to exclude foods that contain artificial colorings, artificial flavorings, and natural salicylates. Exclusion of fruits and vegetables was based on German analyses of salicylate content done at the turn of the century. Information on the exact quantities of salicylate in foods has not been generally available, nor has it been

certain whether permitted foods are completely free of salicylate. There have been claims from fruit canners that some of the fruits excluded by Feingold did not contain appreciable salicylate.

Emphasis on salicylates in hyperactivity has decreased; there has been more interest lately in artificial colors (1). Meanwhile, interest has been growing in the role of salicylates in some cases of urticaria and of asthma. It has long been known that urticaria may follow the ingestion of acetyl salicylic acid medication, but it is now realized that salicylates in foods also can precipitate acute urticaria or exacerbate chronic urticaria.

Salicylate-sensitive urticaria was first noted by Calnan (2,3). Warin (4) in 1960 reported that 22 of 70 patients with chronic urticaria developed exacerbations after administration of aspirin. Moore-Robinson and Warin (5) reported an incidence of 22% of 228 patients, and Champion et al. (6) in 1969 found that 21% of 268 patients with chronic urticaria reacted to aspirin. James and Warin (7) in 1970 reported further investigations in a series of 100 patients with chronic urticaria. Ninety-six patients had been given test doses of aspirin in a "patient blind" manner; 37 of the 96 patients gave a positive reaction to the test dose.

Several authors reported that diets constructed to exclude salicylate may induce prolonged remission of urticaria in those patients who have shown a positive response to oral aspirin challenge (8-11). In 1972, Lockey (12) discussed the part played by salicylates in various foods. Seventy-five percent of the patients of Warin and Smith (11) either cleared or considerably improved after being on the appropriate diet for a 2-month period. This improvement was in line with the results obtained by others, including Michaelsson and Juhlin (8) in 1973 and Doeglas (9) in 1975. More recently, Ros et al. (13) reported their results with exclusion diets. Fifty-nine patients who reacted to salicylates, preservatives, and azo dyes were given a diet designed to reduce consumption of those items. This produced remission in 24% and improvement in 57%. Similarly, Juhlin (14) in 1981 reported that most patients improve when given a diet free from the chemicals to which they reacted.

It is well known that aspirin may exacerbate asthma. McDonald et al. (15) reported that 8 of 42 patients with severe asthma and no history of aspirin exacerbations reacted to an aspirin challenge cf 640 mg.

The authors thank Mrs. S. Vindedzis and Dr. F. B. Whitehead for copies of unpublished references (24 and 51). They are very grateful to Professor A. Basten, Josephine Rogers, Maxine Hosking, Dr. Robert Loblay, Dr. Shervi Van Nunen and Professor R. Clancy for support and encouragement and to Geoff Hutchison for advice on high performance liquid chromatography (HPLC) methodology. The authors also thank Dr. L. R. Brown and M. P. Smyth (Mass Spectrometry Unit, School of Chemistry, University of Sydney) for the GC-MS confirmation of salicylate peaks from the HPLC.

Because we could find no comprehensive list of amounts of salicylate in foods, we extended a clinical study of oral challenges (aspirin, tartrazine, preservatives, and brewer's yeast) in patients with chronic urticaria (16,17) and started to estimate salicylates in some local fruits and vegetables.

Early reports on salicylates in foods used thin-layer chromatography (18-24). More recently, most reports in the literature have used gas chromatography with mass spectrometry (25-49). Details of how the salicylate was extracted before estimation have usually been sparse or even lacking.

We used thin-layer chromatography at first and then for more precise quantitation developed a sensitive specific method of high performance liquid chromatography and combined it with an efficient liquid extraction procedure. We have used the following method to measure salicylate in most common foods.

#### Method

Samples of foods and beverages weighing approximately 100 gm were homogenized in a commercial blender. along with 100 ml 25% sodium hydroxide and 2 gm calcium chloride. Duplicate samples corresponding to 50 gm food or beverage were weighed out. To one sample of each pair, 5 mg salicylic acid standard was added. Both samples were allowed to stand overnight. The two homogenates were then acidified with concentrated hydrochloric acid (HCl) (AR grade) and placed in separate liquid extractors. Two hundred milliliters diethyl ether was placed in a round-bottomed flask, along with several glass boiling beads to prevent bumping. The flask was then placed in a heating mantle and connected to the liquid extractors and condensers. Extraction was carried out for 5 hours. The ether extract was next allowed to cool and to evaporate to dryness in the fume cupboard. The sample was then taken up in 20 ml 2% sodium bicarbonate (NaHCO<sub>3</sub>) and filtered prior to analysis.

The high performance chromatographic system consisted of a Varian 5060 pumping system with Rheodyne 7125 injection valve, several different detectors, Varian CDS III computing integrator, and Houston Omniscribe recorder.

The column was a Waters µBondpak C18 reverse phase column (300mm×4.6mm) fitted with a precolumn packed with Vydac RP P389 packing material. The eluent was monitored with a Varian Vari-Chrom variable wavelength detector with wavelength at 235 nm connected to the recorder; 20 µl samples were injected.

The isocratic solvent was 20% methanol (Ajax Unichrom), 80% glass-distilled water with 0.01 mMol orthophosphoric acid and ion pairing reagent of 0.0025 mMol tetra butylammonium phosphate (TBAP) made to pH 7.0 with 25% sodium hydroxide solution. The salicylic acid standard was 0.25 gm/L in 2% NaHCO<sub>3</sub> and was stored at 4°C.

The column was operated with an isocratic solvent at ambient temperature and a flow program which resulted in a back pressure of 90 to 150 atmospheres. The flow rate was 1.0 ml/minute for 7 minutes, increasing to 2.0 ml/minute over the next 3 minutes.

Results from triplicate or multiple extractions were used to calculate the total salicylate per 100 gm food sample.

Extraction efficiencies were calculated. They varied with the composition of the food. Fruits, vegetables, condiments, and beverages gave extraction rates of greater than 85%, compared with cereals and protein foods for which the extraction rate was approximately 60%. Validation was carried out by extracting several foods 20 times along with spiked samples. Several foods were selected which, by previous analysis, had been found to contain salicylate in amounts that were relatively low (carrots and pumpkin), medium (orange and pineapple), or high (thyme). The foods had varied physical attributes.

For a further discussion of the methodology, see the appendix.

#### Results and discussion

This article presents a list of foods that we have analyzed for salicylate. When present in significant amounts, salicylate is reported as milligrams per 100 gm edible portion. Items are arranged by food groups in Table 1. The two right-hand columns of the table summarize information currently available in the literature.

#### **Fruits**

We found that most fruits contained considerable amounts of salicylate. Raisins and prunes had the highest amounts. Most berry fruits are significant sources of salicylate, with a range from 0.76 mg/100 gm for mulberries to 4.4 mg/100 gm for raspberries. Apples showed considerable variation of salicylate content between varieties.

Dried fruits have relatively high salicylate contents compared with their fresh counterparts because of the removal of water during the drying process. Heat processing for canning does not seem to affect appreciably the salicylate content of fruit. We had the impression that those fruits low in salicylate often have a less piquant flavor, e.g., mangos, pawpaws, and pears, compared with pineapples, oranges, and the berry fruits.

# Vegetables

Within the vegetable group, salicylate content varied widely among the raw foods. It ranged from negligible in bamboo shoots, dried beans, green cabbage, celery, lentils, lettuce, dried peas, and swedes to a high of 6.1 mg/100 gm in gherkins.

Fresh tomatoes contain only small amounts of salicy-late, 0.13 mg/100 gm. However, many commonly used tomato products are considerable sources of salicylate: canned tomatoes, 0.53 mg; tomato paste, 1.44 mg; tomato sauce, 2.38 mg; and tomato soup, 0.54 mg. The increase in available salicylate in the processed tomato products compared with the fresh can be attributed to the use of a fully ripe raw material, to cooking, and to concentration, but it is probably due mainly to the addition of flavoring herbs and spices.

#### Condiments

Some of the herbs and spices contain much more salicylate than has previously been reported for any food. We found that curry powder contains 218 mg salicylate per 100 gm. Others almost as high were paprika, thyme, dill powder, garam masala, oregano, and turmeric.

Although amounts of these condiments used in food are

food	type	state	state salicylate	reported	references
	,,		·	in literatur <del>e</del>	
fruit			mg/100 g/mt		
apple	Golden Delicious	fresh	0.08)		
app.c	Red Delicious	fresh	0.19		
	Granny Smith	fresh	0.59	yes	50
	Jonathan	fresh	0.38		
	Ardmona	canned	0.55‡		
	Mountain Maid	iuice	0.19#	yes	53
apricot	Woontain Ward	fresh	2.58	yes	50,51
арпсы	Ardmona	canned	1.42‡	,	30,31
	Letona	nectar	0.14‡	yes	51
avocado	CCONG	fresh	0.60≉	,	3,
banana		fresh	0#	yes	50
blackberry	John West	canned	1.86	yes	25,33
blueberry	Socomin	canned	2.76	yes	23,33
	John West	canned	2.04		
boysenberry	Australian rockmelon	fresh	1.50#		
canteloupe		fresh		1.000	23,27,51
cherry	sweet		0.85	yes	23,27,31
	John West	canned	2.78		22
t.	Morello Sour	canned	0.30	yes	23
cranberry	5. & W.	canned	1.64	yes	28,29
		sauce	1.44		
currants	black currant	frozen	3.06	yes	25,50
	red currant	frozen	5.06		
custard apple	(from Qu <del>eensland)</del>	fresh	0.21		
dates		fresh	3.73		
	Cal-Date	dried	4.50‡		
figs		fresh	0.18	yes	51
-	S. & W. Kadota	canned	0.25		
	Calamata string	dried	0.64		
guava	Gold Reef	canned	2.02		
grapes	Red Malaita	fr <b>e</b> sh	0.94 )		
	Sultana	fresh	1.88	yes	25
	<ol><li>&amp; W. light seedless</li></ol>	çanned	0.16	,	
	Berri Dark	juice	0.88		
	Sanitarium Light	juice	0.18		
	currants I.P.C.	dried	5.80	yes	25,53
	raisins A.D.F.A.	dried	6.62‡	yes	51
	sultana	dried	7.80	, 43	<del>-</del> -
grapefruit		fresh	0.68‡	yes	18,50
graperran	Berri	juice	0.42	,	.0,50
ciwi fruit	Derri	fresh	0.32	yes	50
emon		fresh	0.18#	yes	18
oganberry	John West	canned	4.40	yes.	10
oquat	JOHN THESE	fresh	0.26		
		canned	0.36		
ych <b>ee</b>					51
mandarin		fresh	0.56‡	yes	31
nango		fresh	0.11		
nuiberry		fresh	0.76		7.0
nectarine		fresh	0.49	yes	50
orange	0 .	fresh	2.39‡	yes	18,50,52
	Berri	juice	0.18‡	y <b>es</b>	51,52
passion fruit		fresh	0.14‡	yes	26,30
pawpaw		fresh	0.08‡		
<b>De</b> ach		fresh	0.58‡	yes	50,51
	Letona	canned	0.68‡	yes	31,32,50
	Letona	nectar	0.10#	yes	51
pear	Packham (with skin)	fresh	0.27‡		
	Packham (no skin)	fresh	0\$		
	William (with skin)	fresh	0.31‡		
	Letona Bartlett	canned	0‡		
persimmon		fresh	0.18‡		
pineapple		fresh	2.10‡	yes	50,51,52
	Golden Circle	canned	1.36	,	,,

<sup>\*</sup>Most trade names are those of products of various Australian companies. Some varieties of foods also are Australian, †Edible portion.

<sup>#</sup>Multiple extractions.

<sup>#</sup>Ashoor (24) found no detectable salicylate in grapefruit, lemon, orange, strawberry, and tangelo.

For coffee, milligrams salicylate per 100 ml made from 2 gm powder in 100 ml water.

[For tea, milligrams salicylate per 100 ml infusion made from two standard tea bags (4 gm dry leaves).

food	type	stat <b>e</b>	salicylat <del>e</del>	reported in literatur <del>e</del>	reference:
				nterature	
	Golden Circle	juice	0.16#	yes	51
płum	Blood (red)	fresh	0.21		
	Keisey (green)	fresh	0.0 <del>9</del> 5	yes	50
	Wilson (red)	fresh	0.11		
	5.P.C. dark red	canned	1.16		
	Letona prunes	can <b>ned</b>	6.87	yes	50,51
pomegranate		fresh	0.07		
raspberries		fresh	5.14	yes	25,33,50
		frozen	3.88	•	,,
rhubarb		fresh	0.13	yes	50
strawberry		fresh	1.36‡	yes	25,33,50,5
tamarillo		fresh	0.10	yes	50
tangelo		fresh	0.72	no	24#
watermelon		fresh	0.48\$		20,50
		· · · · · · · · · · · · · · · · · · ·		yes	20,30
youngberry		canned	3.06		
vegetable					
alfalfa		fresh	0.70		
asparagus		fresh	0.14‡	yes	50
asporagas	Triangle Spears	canned	0.32‡	,	30
bamboo shoots	Sunshine	canned	0.324		
beans	blackeye	dried	0		
	Borlotti	dried	0.08		
	broad, "vicia faba"	fresh	0.73		
	brown	dri <b>ed</b>	0.002		
	green French	fresh	0.11‡	yes	20,50,51
	lima	dried	0	yes	51
	mung	dried	Ō	, ==	•
	soya	dried	Ö	yes	39
	soya grits	dried	ŏ	, cs	3,
bean sprouts	JOYA BINS	fresh	0.06		
					31.50
beetroot	6 11 6: 1	fresh	0.18‡	yes	21,50
	Golden Circle	canned	0.32*		
broccoli		fresh	0.65*		
brussels sprouts		íresh	0.07		
cabbag <del>e</del>	gre <del>e</del> n	fresh	0≉	yes	50
	red	fresh	0.08‡	γes	19
carrot		fresh	0.23‡	yes	21.51
cauliflower		fresh	0.16#	yes	30
celery		fresh	0*	,	20
chicory		fresh	1.02	20	22
,				no	22
chives		fresh	0.03	no	22
choko	(Chayote)	fresh	0.01#		
cucumber	(no peel)	fresh	0.78‡	yes	20
	Aristocrat gherkin	canned	6.14#	yes	20
<del>e</del> ggplant	(with peel)	fresh	0.8 <b>8</b> ‡	yes	20
	(no peel)	fresh	0.30≠		
endive	,	fresh	1.9	no	22
horseradish	Eskal	çanned	0.18	yes	19
leek	LINA	fresh	0.08	703	1.7
lentil	brown	dried			
entii			0		
L.m.	red	dri <b>ed</b>	0		
lettuce		fresh	0#	no	22
marrow	(Cucurbita pepo)	fresh	0.17		
mushroom		fresh	0 24		
	Champignon	canned	1.26		
okra	Zanae	canned	0.59		
olive	black Kraft	canned	0.34‡		
,-	green Kraft	canned	1.29‡	\/Ar	41
onion	RICELI MAIL			yes	
onion		fresh	0.16#	no	22
parsnip		fresh	0.45		
peas	chick-pea	dri <b>ed</b>	0		
	green	fresh	0.04\$	yes	20,50,51

<sup>\*</sup>Most trade names are those of products of various Australian companies. Some varieties of foods also are Australian.

<sup>†</sup>Edible portion. ‡Multiple extractions.

<sup>#</sup>Ashoor (24) found no detectable salicylate in grapefruit, lemon, orange, strawberry, and tangelo.

"For coffee, milligrams salicylate per 100 ml made from 2 gm powder in 100 ml water.

[For tea, milligrams salicylate per 100 ml infusion made from two standard tea bags (4 gm dry leaves).

food	type	state	salicylate	reported in literature	references
	green split pea	dried	0		
	yellow split pea	dried	0.02		
peppers	gr <del>ee</del> n chili	fresh	0.64#		
	r <b>ed</b> chili	<b>fres</b> h	1.20‡		
	yellow-green chili sweet, green	fresh	0.62‡		
	(Capsicum)	fresh	1.20#	yes	20,50
pimientos	Arson sweet red	canned	0.15	/55	20,00
potato	white (with peel)	fresh	0.12#	yes	34,50
potato	white (no peel)	fresh	0#	,	5 .,55
pumpkin	witte (no peer,	fresh	0.12‡	yes	20,51
radish	red, small	fresh	1.24‡	yes	21
shallots	ieu, silali	fresh	0.03	yes	21
		fresh	0.58		22,51
spìnach		frozen	0.16#	yes	22,31
	L-L				
squash	baby	fresh	0.63		
swede		fresh	0		
sweet corn	Mountain Maid	fresh	0.13#	yes	50
	niblets Mountain Maid	canned	0.26‡	yes	50
	Creamed	canned	0.39#	yes	50,51
sweet potato	white	fresh	0.50‡	,03	30,31
sweer potato	yellow	fresh	0.48‡	yes	50
tomato	yenow	fresh	0.13‡	yes	20,35,36,3
	Lavass	canned	0.534		50,51
	Letona		0.53‡	yes	51
	Goulbum Valley	juice	0.10#		
	Heinz	juice	0.12#		
	Letona	juice	0.18#		
	Campbell	paste	0.57‡)		
	<b>Legg</b> o	Paste	1.44‡ }	yes	50
	Tom Piper	paste	0.43		
	Heinz	soup	0.54‡ 🗎		
	Kiaora	soup	0.54‡ }	yes	50,51
	P.M.U.	soup	0.32	•	
	Fountain .	sauce	0.94#1		
	Heinz	sauce	2.48		
	I.X.L.	sauce	1.06#	yes	49,50,51,5
	P.M.U.	sauce	0.98≠	,	,,.
	Rosella	sauce	2.15#		
turnip	Nescria	fresh	0.16#		
watercress		fresh	0.84#		
zucchini		fresh	1.04#	no	20
condiments			1.047		20
allspice	powder	dry	5.2		
aniseed	powder	dry	22.8	yes	55
bay leaf	leaves	dry	2.52	,	
pasil	powder	dry	3.4		
"Bonox"	powder	liquid	0.28		
canella	oowder	dry	42.6		
cardamom		dry	7.7		
	powder				
caraway	powder	dry	2.82		
ayenne	powder	dry	17.6		
celery	powder	dry	10.1		
chili	fl <b>ake</b> s	dry	1.38		
	powder	dry	1.30		
cinnamon	powder	dry	15.2‡	yes	56,57,58
cloves	whole	dry	5.74	yes	54
coriander	leaves	fresh	0.20	•	
cumin	powder	dry	45.0		
curry	powder	dry	218		

<sup>\*</sup>Most trade names are those of products of various Australian companies. Some varieties of foods also are Australian. †Edible portion.

\*Multiple extractions.

#Ashoor (24) found no detectable salicylate in grapefruit, lemon, orange, strawberry, and tangelo.

\*For coffee, milligrams salicylate per 100 ml made from 2 gm powder in 100 ml water.

#For tea, milligrams salicylate per 100 ml infusion made from two standard tea bags (4 gm dry leaves).

food	type	state	salicylate	reported in literature	references
dill		fresh	6.9		
	powder	dry	94.4		
fennel	powder	dry	0.8	no	22
fenugreek	powder	dry	12.2		
five spice	powder	ďγ	30.8		
garam masala	powder	dry.	66.8		
garlic	bulbs	fresh	0.10#	yes	22
ginger	root	fr <b>esh</b>	4.5		
mace	powder	d <b>ry</b>	32.2		
"Marmite"	Sanitarium	paste	0.71#		
mint	common garden	fresh	9.4#		
mixed herbs	leaves	dry	55.6		
mustard	powder	d <b>ry</b>	26		
nutmeg	bomder	diny	2.4‡		
oregano	powder	d <b>ry</b>	66		
paprika	hot powder	d <b>ry</b>	203		
	sw <b>eet</b> powder	dry	5.7		
parsley	leaves	fresh	0.08‡	yes	21,22
pepper	black powder	dry	6.2‡		
	white powder	d <b>iry</b>	1.1‡		
pimiento	powder	d <b>ry</b>	4.9		
rosemary	powder	d <b>ry</b>	68		
saffron	powder	dry	0		
sage	leav <b>es</b>	d <b>ry</b>	21.7		
soy sauce		liquid	0		
Tabasco Pepper	McIlhenny	sauce	0.45	yes	38,40
tandori	powder	dry	0		
tarragon	powder	d <b>ry</b>	34.8		
turmeric	powder	dry	76.4		
thyme	leav <b>es</b>	dry	183		
vanilla	essence	liquid	1.44		
vinegar	malt	liquid	0		
	white	liquid	1,33		
Worcestershire		1			
sauce		liquid	64.3		
"Vegemite"	Kraft	paste	0.81‡		
drinks					
"Aktavite"		powder	0		
cereal coffee®	Bambu	powder	0.15		
	Dandelion	powder	0.08		
	"Ecco"	powder	0		
	"Nature's Cuppa"	powder	2.26		
	"Reform"	powder	0.38		
Coca-Cola		liquid	0.25		
coffee <sup>¶</sup>	Andronicus Instant	powder	0 1		
	Bushells Instant	powder	0.21		
	Bushells Turkish Style	powder	0.19		
	Gibsons Instant	powder	0.12		
	Harris Mocha Kenya	beans	0.45		
	Harris Instant I	powder	0		
	Harris Instant II	cowder	0.10		4-
	International Roast	powder	0.96	yes	45
	Maxwell House Instant	powder	0.84		
	Moccona Instant	granules	0.64		
	Moccona Decaffeinated	powder	0		
	Nescafé Instant	granules	0.59		
	Nescafé Decaffeinated	powder	0		
	Robert Timms Instant	powder	0.16		
herbal teall	camomille	bag	0.06		
	fruit	bag	0.36		
	peppermint	bag	1.10		
	rose hip	bag	0.40		

<sup>&</sup>quot;Most trade names are those of products of various Australian companies. Some varieties of foods also are Australian.

tEdible portion.

<sup>#</sup>Multiple extractions.

<sup>#</sup>Ashoor (24) found no detectable salicylate in grapefruit, lemon, orange, strawberry, and tangelo.
"For coffee, milligrams salicylate per 100 ml made from 2 gm powder in 100 ml water.

||For tea, milligrams salicylate per 100 ml infusion made from two standard tea bags (4 gm dry leaves).

food	type	state	salicylate	reported in	references
				literature	
"Milo"		powder	0.01		
"Ovaltine"		powder	0		
rose hip teal	Delrosa	syrup	1.17		
teal	Asco	bag	6.4		
Call	Billy	leaves	2.48		
	Burmese Green	le aves	2.97		
	Bushells				
		bag	4.78		
	Golden Days		2 20		
	Decaffeinated	bag	0.37		
	Harris	bag	4.0		
	Indian Green	leaves	2.97		
	Peony Jasmine	leaves	1,9		
	Old Chinese	leaves	1.9		
				yes	46,47,48
	Tetley	bag	5.57	•	
	Twinings:		[		
	Earl Grey	bag	3.0		
	English Breakfast	bag	3.0		
		leav <b>e</b> s	4.24		
	Darjeeling				
	Irish Breakfast	bag	3.89		
	Lapsang Souchong	bag	2.40		
	Lemon Scented	bag	7.34		
	Orange Pekoe	leaves	2.75		
	Prince of Wales	bag	2.97		
	FITTIEC OF FREE	OGB	<b>4.2.</b> ,		
ereals					
rrowroot	powder	dry	0		
parley	unpearled	dry	Ö		
ouckwheat	grains	dry	0		
maize	meal	dry	0.43		
millet	grains	dry	0		
	hulled grains	dry	0		
oats	meal	dry	Ö		
rice	brown grains	dry	0		
ice			0‡ 0‡		
	white grains	dry	-		
rye	rolled	dry	0		
wheat	grains	dry	0		
nuts and seeds	-				
		1,aak	3.0		
almonds		fresh	3.0		
Brazil nuts		fresh	0.46		
cashew nuts		fresh	0.07		
coconut	dessicated	dry	0.26		
hazelnuts	UESSICAIEO	fresh	0.14		
Macadamia nuts		fresh	0.52		
peanuts	unshelled	fresh	1.12		
	Sanitarium butter	paste	0.23		
pecan nu <b>s</b>		fresh	0.12		
pecan num pine nuts		fresh	0.51		
pistachto nuts		fresh	0.55		
poppyseed		dry	0		
sesame seed		dry	0.23		
sunflower seed		dry	0.12		
walnuts		fresh	0.30		
	<b>c</b> : -				
water chestnut	Socomin	canned	2.92		
sugars					
carob	powder	dry	0‡		
			0‡		
cocoa	powder	dry			
honey	Allowrie	liquid	2.5		
	Aristocrat	liquid	3.7		
	Capillano	liquid	10.14		
	Mudgee	liquid	3.9		
	"No Frills"	liquid	11.24		
golden syrup	C.S.R.	liquid	0.10#		
maple syrup	Camp	liquid	0		

<sup>\*</sup>Most trade names are those of products of various Australian companies. Some varieties of foods also are Australian. tEdible portion.

<sup>#</sup>Multiple extractions.

<sup>#</sup>Ashoor (24) found no detectable salicylate in grapefruit, lemon, orange, strawberry, and tangelo.

For coffee, milligrams salicylate per 100 ml made from 2 gm powder in 100 ml water.

For tea, milligrams salicylate per 100 ml infusion made from two standard tea bags (4 gm dry leaves).

food	type	state	salicylate	reported in	references
				literature	
sugar	white granulated	dry	0		
molasses	C.S.R.	liquid	0.22		
confectionery					
caramel	Pascall Cream	dry	0.12		
licorice	Barratts	dry	9.78		
_	Giant	dry	7.96		
peppermints	Allens Strong Mint	dry	0.77		
	Allens "Koolmint" Lifesavers	dry dry	7.58 0.86		
	"Minties"	dry dry	1.78		
	Allens "Steamrollers"	dry	2.92		
dairy		,	2		
cheese	Blue Vein	fresh	0.05		
Cheese	Camembert	fresh	0.03		
	Cheddar	fresh	0		
	cottage	fresh	0		
	Mozarella	fresh	0.02		
	Tasty Cheddar	fresh	0		
milk	fresh full		0.		
	cream	liquid	0 <b>‡</b> 0		
yogurt	full cream	fresh	U		
meat, fish					
and eggs		fh	0.4		
beef		fresh	0*		
chick <b>en</b>	white	fresh fresh	0 <b>‡</b> 0		
egg	yolk	fresh	0		
kidney	york	fresh	0		
lamb		fresh	0‡		
liver		fresh	0.05		
oyster		fresh	0		
pork		fresh	ð		
prawn		fresh	0.04		
salmon	Lunchtime Pink	canned	0		
scallop		fresh	0.02		
tripe	Seakist	fresh	0 0		
tuna	Seakist	canned	U		
alcoholic					
drinks	Reschs Dinner Ale		0.353		
beer	Toohevs Draught		0.35	no	43
	Tooths Sheaf Stout		0.32	:10	43
cider	Bulmer's Dry		0.17		
	Bulmer's Sweet		0.19		
	Lilydale Dry		0.17		
	Mercury Dry		0.16		
liqueurs	Benedictine		9.04		
	Cointreau		0.66		
	Drambui		1.68		
port	Tia Maria McWilliams Royal Reserve		0.83 1.4		
port	Stonyfell Mellow		4.2		
sherry	Lindemans Royal Reserve		<b>7.</b> ■		
- < : /	Sweet		0.56		
	Mildara Supreme Dry		0.46		
	Penfolds Royal Reserve				
	Sweet		0.49		
spirits	brandy—Hennessy		0.4		
	gın—Gilbey's		0		
	rum—Bundaberg		0.76}	yes	42
	rum—Captain Morgan		1.28∫	/	•-

<sup>\*</sup>Most trade names are those of products of various Australian companies. Some varieties of foods also are Australian.

tEdible portion.

<sup>‡</sup>Multiple extractions.

<sup>#</sup>Ashoor (24) found no detectable salicylate in grapefruit, lemon, orange, strawberry, and tangelo.

¶For coffee, milligrams salicylate per 100 ml made from 2 gm powder in 100 ml water.

∥For tea, milligrams salicylate per 100 ml infusion made from two standard tea bags (4 gm drv leaves).

food	type	state	salicylate	reported in literature	references
	vodka—Smirnoff		0		
	whiskey—Johnnie Walker		0	no	43
wines	Buton Dry Vermouth		0.46		
	Kaiser Stuhl Rosé		0.37		
	Lindemans Riesling		0.81		
	McWilliams Dry White Wine		0.10		
	McWilliams Cabernet				
	Sauvignon		0.86		
	McWilliams Private Bin				43.44
	Claret		0.90	yes	43,44
	McWilliams Reserve Claret		0.35		
	Penfolds Traminer Riesling				
	Bin 202		0.81		
	Seaview Rhine Riesling		0.89		
	Stonyfell Ma Chére		0.69		
	Yalumba Champagne		1.02		

<sup>\*</sup>Most trade names are those of products of various Australian companies. Some varieties of foods also are Australian.

small, they can make a significant contribution to dietary salicylate.

#### Drinks

The drinks in Table 1 are divided into two subgroups, alcoholic and nonalcoholic. Within the nonalcoholic beverages, salicylate varies widely from negligible in milk, cocoa, and decaffeinated coffee to a high of 7.3 mg salicylate per 100 ml in one of the teas.

Teas are thus an important source of salicylate in the usual diet. We analyzed 18 different brands and varieties. All contained more than 1.9 mg/100 ml except decaffeinated tea, which contained only 0.37 mg. Salicylate is soluble in methylene chloride, a solvent commonly used for extraction of caffeine.

Table 1 includes data for nine coffees and five coffee substitutes. All but one contain less than 0.96 mg salicy-late per 100 ml. The higher value of 2.26 mg for one cereal coffee may reflect the raw materials, such as chicory, used in its manufacture.

Of the alcoholic beverages, wines appear to contain about the same amount as grape juice (range 0.35 to 1.0 mg/100 ml). Cider contains a low level similar to apple juice. We found that beer contains appreciable amounts, which was not previously reported in the literature. There was a big range of salicylate in the liqueurs that we tested, with Benedictine highest, presumably from one of the secret ingredients.

### Sweets

Little research has been done by others on the presence of salicylate in popular sweets and confectionery. However, Porsch et al. (55) and others have identified salicylate in anise and mint. We therefore analyzed a representative number of licorices and peppermints, which have been listed in Table 1.

Values for cocoa and carob—negligible amounts—are also included. The peppermints contained variable amounts. It would appear that high salicylate contents in mint candies come mostly from additional flavorings like methyl salicylate.

#### Legumes, nuts, and seeds

Legumes are generally safe for salicylate-sensitive people. All dry legumes that were analyzed had less than 0.08 mg/100 gm in the dry state. Soybeans are low in salicylate; similarly the fermented product, soy sauce, contained negligible amounts.

Table 1 contains information on 10 nuts. Almonds, water chestnuts, and peanuts (in skin) were moderately high (3.0, 2.9, and 1.1 mg salicylate per 100 gm, respectively). Other nuts contained small to moderate amounts (0.07 to 0.5 mg/100 gm). Coconut contains a small amount. Sesame seeds, poppy seeds, and sunflower seeds (included here with the nuts) contain negligible amounts of salicylate.

#### Cereals

Salicylate levels in nine whole-grain cereals are negligible, with the exception of a yellow maize meal which contributed 0.43 mg/100 gm.

## Meat, poultry, fish, eggs, and dairy products

Beef, lamb, pork, and chicken all have negligible salicy-late content. Liver was found to contain 0.05 mg/100 gm. Only two fish have been analyzed so far, and both contained no salicylate. Oysters had none; prawns and scallops contained 0.04 and 0.02 mg/100 gm, respectively, having more salicylate than most meats or fish but still very small amounts.

Dairy products, including cheese, do not contain appreciable amounts of salicylates.

tEdible portion.

<sup>#</sup>Multiple extractions

<sup>#</sup>Ashoor (24) found no detectable salicylate in grapefruit, lemon, orange, strawberry, and tangelo.

For coffee, milligrams salicylate per 100 ml made from 2 gm powder in 100 ml water.

<sup>||</sup>For tea, milligrams salicylate per 100 ml infusion made from two standard tea bags (4 gm dry leaves).

#### Conclusion

The values for salicylate in foods that we have obtained work out to a range from about 10 mg to 200 mg/day salicylate in Western diets. This is of the same order of magnitude as the challenge dose of salicylate used in clinical testing (60), usually a 300-mg aspirin tablet. The usual adult pharmacological dose of aspirin is 600 mg (two tablets) at a time, often several times a day. Previous figures for salicylates in most foods are so much smaller than this that it is difficult to see how the food could have similar effects to salicy late medication in sensitive individuals.

#### References

- [1] National Advisory Committee on Hyperkinesis and Food Additives: Final Report of the Nutrition Foundation, Washington, DC, Nutrition Foundation,
- (2) Calnan, C.D. Release of histamine in unicaria pigmentosa, Lancet 1:996, 1957
- (3) Calnan, C.D., Urticarial reactions, Br Med 12:649, 1964
- (4) Warin, R.P. The effect of aspirin in chronic urticaria. Br J Dermatol 72:350, 1960
- (5) Moore-Rebinson, M., and Warin, R.P. Effect of salicylates in unticaria. Br Med ) 4 262, 1967
- (6) Champion R.H., Roberts, S.O.B., Carpenter, R.G., and Roger, J.H., Urticaria and angioedema. A review of 554 patients. Br J Dermatol 81 588, 1969
- (7) James, J., and Warin, R.P.: Chronic unicaria. The effect of aspirin. Br J Dermatol 82:204 1970.
- (8) Michaelsson, G., and Juhlin, L.: Urticaria induced by preservatives and dve additives in food and drugs. Br J Dermatol 88:525, 1973.

  19) Doeglas, H.M.: Reactions to aspirin and rood additives in patients with
- chronic urticaria, including the physical urticarias. Br J Dermatol 98:135,
- (10) Doeglas, H.M.: Dietary treatment of patients with chronic urticaria and intolerance to aspirin and food additives. Dermatologica 154:308, 1973
- (11) Warin R.P., and Smith, R.J., Challenge test batters in chronic urticaria, 8r.) Dermatol 94,401 1976.
- (12) Lockey, S.O., Reactions to hidden agents in roods, beverages and drugs. Ann Allergy 29:461 1971
- (13) Ros. A.M., Juhlin, L., and Michaelsson, G. A follow up study of patients with recurrent unticaria and hypersensitivity to aspirin, benzoates and azo dives. Br J Dermatol 95:19: 1976.
- (14) Junion, L. Recurrent unicaria: Clinical investigation of 330 patients. Br J. Dermatol 104L 369 1981
- 115; McDonald, J.R., Mathieson D.A. and Stevenson, D.D. Aspirin intolerance in asthma. Detection by oral chatlenge: J Allergy 50, 198, 1972 116) Gibson, A.R., and Clancy, R.L., An Australian exclusion diet, Med.) Aust
- 1 290, 1978.
- (17) Gibson, A.R. and Clanck, R.L. Management of chronic idiopathic urticaria by the identification and exclusion of dietary factors. Clin Allergy, 10:699.
- (18) Stöhr, H., and Herrmann, K. Über das vorkommen von verbindungen der hydroxyzimtsauren hydroxypenzoesauren und hydroxycumarine in citrustruchten Zifur Lebensm Unters Forsch 159 305 1975
- il 9) Schmidtlein, H., and Herrmann, K. Uber die phenoisauren des gemuses. I Hydroxyzimtsauren und hydroxybenzoesauren der kohiartern und anderer crucireren-blatter. Z Lebensm Unters Forsch 159,139, 1975
- (20) Schmidtlein, H., and Herrmann, K., Ober die prienolsäuren des gemuses. II. Hydroxyzimtsäuren und hydroxybenzoesauren der frucht und samengemüsearten Z Lebensm Unters Forsch 159-213 1975 (21) Stöhr, H., and Herrmann, K.: Über die phenolsäuren des gemüses, III
- Hydroxyzimtsauren und hydroxybenzoesauren des wurzeigemuses. Z
- Lebensm Unters Forsch 159 219, 1975
  (22) Schmidtlein H., and Herrmann. K. Über die phenolsäuren des gemuses. IV. Hydroxyzimtsäuren, und hydroxybenzoesauren, weiterer, gemüsearten, und der kartoffeln. Zillebensm Unters Forsch 159:255, 1975.
- (23) Stöhr, H. Mosel, H. and Herrmann, K.: The phenolics of fruits. VII. The phenolics of cherries and plums and the changes in catechins and hydroxvoinnamic acid derivatives during the development of fruits. Z Lebensm Unters Forsch 159 (11) 85, 1975
- (24) Ashoor, S., and Chu. F.S.. Analysis of salicylic acid and methyl salicylare in truits, and almonds. Food Research Institute. University of Wisconsin, Madison Unpublished typescript.
- (23) Nursten, H.E., and Williams, A.A. Fruit aromas: A survey or components identified. Chem Indust March 1967, p. 486

- (26) Winter, M., and Klöti, R.: Über das aroma der gelben passionsfrucht. Heli Chim Acta 55:181, 1972,
- (27) Gierschner, K., and Baumann, G.: Aromatic substances in fruit. Reichstoffe
- Aromen Koerperflegemittel 18(1):3, 1968. (28) Anjou, K., and Von Sydow, E.: Aroma of cranberries, I. Vaccinium vitisidaea. Acta Chem Scand 21:945, 1967.
- (29) Anjou, K., and Von Sydow, E.: Aroma of cranbernes, III. Juice of vaccinium vitisidaea. Acta Chem Scand, 23:109, 1969
- (30) Murray, K.E.: The flavour of purple passionfruit. Food Technol Aust 25:446, 1973
- (31) Do, J.E., Salunkhe, D.K., and Olson, L.E.: Isolation, identification and comparison of volatiles of peach fruit as related to harvest maturity and artificial ripening. I Food Sci 34.618, 1969.
- (32) Kemp, T.R., Knavel, D.E., and Stoltz, L.P.: Characterization of some volatile components of muskmelon fruit. Phytochemistry 10:478, 1971.
- (33) McGlumphy, J.H.: Fruit flavors. Food Technol 5.351, 1951.
- (34) Buttery, R.G., Siefert, R.M., and Ling, L.C.: Characterization of some volatile potato components. Agric Food Chem 18:538, 1970.
   (35) Johnson, A.E., Nursten, H.E., and Williams, A.A.: Vegetable volatiles. Survey
- of components identified. Chem Indust July 1971, p. 1212.
- (36) Pyne, A.W., and Wick, E.L.: Volatile components of tomatoes. J Food Sci 30:192, 1965.
- (37) Buttery, R.G., Seifert, R.M., Gaudagni, O.G., and Ling, L.C.: Characterization of additional volatile components of tomato. I Agric Food Chem 19:524,
- (38) Haymon, L.W., and Aurand, L.W.: Characterization of volatile constituents of Tabasco peppers. J Agric Food Chem 19:1131, 1971
- (39) Arai, S., Suzuki, H., Fujimaki, M., and Sakurai, Y.: Flavour components in soybean. III. Volatile fatty acids and volatile amines. Agric Biolog Chem
- (Tokyo) 30:363, 1966. [40] Buttery, R.G., Seitert, R.M., Gaudagni, D.G., and Ling, L.C.: Characterization of some volatile constituents of bell peppers. J Agric Food Chem 17:1322, 1969
- (41) Fedeli, E., and Jacini, G.: Odorous components of plive oil. Chim Indust (Milan) 52(2):161, 1970.
- (42) Liebich, H.M., Koenig, W.A., and Baver, E.: Analysis of the flavour of rum by gas-liquid chromatography and mass spectrometry. J Chromatogr Sci 8:527, 1970
- (43) Kahn, J.H.: Compounds identified in whiskey, wine, beer: A tabulation, J. Assoc Off Anal Chem 52,1166, 1969.
- (44) Christensen, E.N., and Caputi, A.: The quantitative analysis of flavanoids and related compounds in wine by gas-liquid chromatography. Enology Viticulture 1968 p. 238.
- (45) Storfelsma, J., Sipma, G., Kettenes, D.K., and Pypker, J., New components of roasted coffee, J. Agric Food Chem 16:1000, 1968.
- (46) Saijo, R., and Kuwabara, U., Voiatile flavour of brack tea. Agric Biolog Chem
- (Tokyo) 31:389, 1967 (47) Yamanishi, T.: The aroma of various teas. In Charalambous, G., and Inglett, G.E., eds., Flavor of Foods and Beverages: Chemistry and Technology, New York: Academic Press, 1978.
- (48) Stahl, W.H.: The chemistry of tea and tea manufacturing, Adv Food Res 11 201, 1962.
- (49) Daenens, P., and Larveile, L., Column chromatographic cleanup and gas liquid determination of hydroxy benzoic esters in food. J Assoc Off Anal Chem 56 1515, 1973
- (50) Rebertson, G.L., and Kermode, W.J., Salicylic acid in fresh and canned fruit and vegetables. J Sci Food Agric 32, 883, 1981.
- (51) Thomberg, W. Salicylic acid and methyl salicylate in canned Delmonte products. Delmonte Foods, 1976. Unoublished typescript
- (52) Gupoy, H., Whitheld, F.B., and Woodhill, I.B., Salicylic acid in truit and vegetables. Report or Research 1977-1978. Sydney, Aust.: CSIRO Division of
- (53) Belianca, N. In Furia. T.E., and Bellanca, N., eds., Fenaroti's Handbook of Flavor Ingredients, Cleveland, Cleveland Chemical Rubber Co., 1971, pp. 192 and 406
- (54) Devama, T., and Horiguchi, T.: Studies of the components of essential oil of clove Yakugaku Zasshi 91,1383, 1971. (55) Porsch, F., Farnow, H., and Winkler, H.: The constituents of star aniseed oil.
- Oragoco Rep 12.123, 1965.
- (56) Heide R. Qualitative analysis of the essential oil of cassia (cinnamomum cassia blume). J Agric Food Chem 20:747+1972 (57) Dodge, FD: Constituents of oil of cassia. I Indust Engineer Chem 10:1005,
- 1918 (58) Onishi, I., and Yamamoto, K.: Essential oils of tobacco leaves. Chem Abstr
- 50:15028, 1956.
- (59) Horwitz, W.: Food additives: Direct. In Horwitz, W., ed.: Official Methods of Analysis, Washington, DC: Association of Official Analytical Chemists, 1975, pp. 364-365.
- (60) Van Nunen, S.A., Swain, A.R. Krilis, S., Basten, A., Catenby, P.A., and Loblay, R.H.: Dietary modification in chronic idiopathic urticaria and/or angioedema. Proceedings of XI International Congress of Allergology and Clinical Immunology, London, 1982.

### Appendix: Discussion of methodology

We started with shaking homogenized food in ether in a 2 L separating funnel evaporating to dryness and then taking up in a small volume of ether for TLC on silica gel. Quantification was unreliable. We tried gas chromatography of trimethylsilyl derivatives, but the method proved unsatisfactory in our hands. High performance liquid chromatography (HPLC) was next tried, and we found a modified AOAC (59) extraction procedure to be straightforward and reproducible and to give good recoveries of added standards.

The modification we now use is stronger sodium hydroxide (NaOH) (25% overnight instead of 10% NaOH for over 2 hours). This is done with the aim of completing hydrolysis and converting all salicylate compounds to free salicylate. Our acidification step is also different. HCl is used to bring the pH to 2.0.

Because there was marked emulsion formation in the separating funnel with some food homogenates and because recoveries of added standard were only around 50%, we changed the extraction procedure and used ether in glass extractors over 5 hours. This gives good recoveries of added standard salicylic acid and benzoic acid.

Specificity of the salicylate peak was checked by two methods. First, several phenolic compounds chemically similar to salicylic acid and which might occur in foods were run on the column. All 20 compounds had retention times different from the retention time of salicyclic acid (Table 2). Thymol was tested as well, but no peak was seen with our usual solvent system. When the solvent was 50% methanol: 50% water, thymol had a retention time of 24 minutes and salicylic acid of 2 minutes.

Second, the salicylate peaks from 11 foods were collected and analyzed by gas chromatography and mass spectrometry in the Mass Spectrometry Unit of Sydney University's School of Chemistry. The foods were two vegetables (maize and radish), two fruits (red currants and dates), three herbs or spices (curry, cumin, and rosemary), two sugary foods (honey and licorice), one sample of wine, and one sample of tea.

Samples of sodium salicylate (AR) were run on the HPLC to determine retention times, to optimize conditions for collecting the salicylate peak. Peaks from foods with retention times identical to the salicylate standard were collected, acidified with 1 ml 3 M hydrochloric acid, extracted with diethyl ether (AR 10 ml, 3 times) and washed with distilled water (10 ml, 3 times). The ether solution was drawn off, dried over anhydrous magnesium sulphate, and filtered through cotton wool into a 1 ml "Reactivial" (Regis). The volume was evaporated to 10 µl with dry nitrogen under gentle heat. Bis (trimethylsilyl) trifluoroacetamide (BSTFA) (Regis) 100 µl was added in a dry environment and refluxed gently for 1 hour. The solution was

Table 2. High performance liquid chromatography\*

compound	retention time (minutes)
p-amino benzene	2.46
para amino benzoic acid	3.20
phthalic acid	3.31
salicylamide	3.32
salicin	3.43
40H benzoic acid	3.46
vanillic acid	3.51
sulphosalicylic acid	4.68
anthranilic acid	6.01
gentisic acid	6.75
aspirin	7.10
benzoic acid	8.20
catechol	8.23
theophylline	8.34
quercetin	11.30
vanillin	11.72
acetanilide	14.41
phenol	14.81
salicylic acid	16.81
coumarin	18.88
methyl salicylate	58.43

\*Solvent: 20% methanol, 80% glass distilled water, 0.01 mmol H.PO., 0.0025 mmol TBAP pH 7.0

reduced to 10 µl under dry nitrogen and injected into the gas chromatograph.

The column of the gas chromatograph, a Pye 104, was glass 2 m×6 mm, packed with OV 17 (3%) on Chromosorb W (100/120 mesh). Helium flow through the column was 30 ml/ min. The gas chromatograph was interfaced (via an AEI glassjet separator) to an AEI MS-30 mass spectrometer operating at 4 KV with an ionization voltage of 70 ev. The chromatographic trace was produced by the total ion current monitor of the mass spectrometer run at 20 ev except when scanning (10 second/ decade), when the ionizing voltage returns to 70 ev. All the peaks from our own HPLC were found to contain a significant amount of salicylate.

Our results are considerably higher than those reported in the literature, e.g., about 10 times higher for pineapple juice but more than 100 times higher for pineapples. We believe this can be explained partly by the efficient extraction procedure we have developed but probably mostly by the conversion of bound salicylate to free salicylic acid so that we are measuring total salicylate in the foods.