

A Determination of the Salicylate Content of Ingredients Commonly Used in Korean Foods Suggested for Allergic Patients*

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ABSTRACT

Salicylate from plant foods and histamine from animal foods have been suggested to cause various food allergic symptoms. Salicylate regarded as one of the inducing agents for chronic urticaria and asthma in allergic patients was studied. There is much interest about the effects of salicylate in food allergy. It is recently suggested that the intake of salicylate from foods may have contributed to chronic urticaria and asthma. The purpose of this study is to analyze the salicylate content in various foods. 153 ingredients commonly used in Korean foods were analyzed by HPLC. Among the vegetables, chard(8.22 μ g/g), soybean sprout(5.13 μ g/g), perilla leaf(3.03 μ g/g), Chinese cabbage(2.25 μ g/g), carrot(2.16 μ g/g), and squash(1.33 μ g/g) contained higher amounts of salicylate as compared to other vegetables. Among fruits, oranges(5.13 μ g/g), grapefruits(2.90 μ g/g), and plums(1.65 μ g/g) contained the most. In animal foods, almost no salicylate was found except in chocolate milk(0.32 μ g/g). It was also found in black pepper(8.25 μ g/g), coffee(can)(5.00 μ g/g), mustard(4.16 μ g/g), curry powder(3.24 μ g/g), kochujang(2.37 μ g/g), cream powder(1.19 μ g/g), brown rice(1.04 μ g/g) and sesame oil(1.00 μ g/g). All information found in this study can be used as nutritional education materials in potentially allergic people for a prudent diet plan and can also provide proper dietary guidelines for these allergic patients. (*Korean J Nutrition* 31(5) : 949~956, 1998)

KEY WORDS : salicylate content · Korean food · food allergy · HPLC.

Introduction

Salicylate occurs naturally in many foods, predominantly in fruits and vegetables. It is also contained in aspirin in the form of acetyl salicylate, in wintergreen or mint flavoring materials added to foods,

Accepted : June 29, 1998

*This study was supported by '96-Yonsei University Research Fund

and drugs and seasoning as methyl salicylic acid or salicin¹⁾²⁾³⁾.

Salicylate has been known to exaggerate the symptoms of chronic urticaria by either acting as an antigen during an allergic reaction or increasing histamine reaction in the skin. Salicylate-sensitive urticaria was first noted by Calman⁴⁾⁵⁾. In some cases of asthmatic patients, severe attacks were reported to occur after taking aspirin containing acetyl salicylate⁶⁾⁷⁾. Since a substantial number of chronic urticaria pa-

tients were found to have high levels of salicylate in their blood and their symptoms worsened after taking aspirin, it was naturally suggested that these allergic patients should be treated in salicylate-free conditions^{8,9}. Therefore it seemed to be advantageous to treat the allergic patients with asthma and chronic urticaria using salicylic acid restricted diets¹⁰. Consequently the salicylate content in foods is of great interest.

Studies concerning salicylate have not been extensively researched in Korea, and although some research has been done in other countries, the results vary due to their different analytical methods. The salicylate contents in food should be accurately analyzed not only for the diet of allergic patients but also to prove the clinical effects of salicylate that is currently being discovered.

The salicylate content was analyzed in 153 ingredients commonly found in Korean foods. The results would provide not only a basic analysis of these common ingredients but also useful information in constructing a viable diet that would prevent or treat certain food allergies, such as chronic urticaria and asthma.

Materials and Methods

1. Food materials

Selection of the 153 different types of food was based upon analysis of the ingredients of the Korean food listed in the National Nutrition Survey¹¹ and Food Composition Table¹². Fresh materials were purchased from markets in July 1996 and refrigerated at a temperature of 0~4°C. Certain materials were kept in freezer for analysis.

1) Grains, potatoes, sugars, beans, seeds and their products are listed below: rice, white flour, barley, breakfast cereal, corn, job's tears, glutinous millet, brown rice, noodles(dried), ramyon, loaf bread, sponge cake, cracker, potato, sweet potato, starch vermicelli, chewing gum, honey, sugar, chocolate, candy, black soybean, soybean, soybean curd, soybean milk, green peas, small red bean, sesame seed, perilla seed, sunflower seed, peanut.

2) Vegetables, fruits and their products are egg plant, braken, chard, carrot, doraji(Chinese bellflower

er root), perilla leaf, garlic, radish, water dropwort, Chinese cabbage, leek, ginger, myungbean sprout, spinach, cabbage, onion, cucumber, soybean sprout, tomato, green onion, green hot pepper, squash, radish pickle(pickled radish in rice bran), oyster mushroom, mushroom, oak mushroom, mandarin orange, persimon, pear, apple, water melon, orange, melon, grape, chestnuts, pine nuts, walnuts, raisin, jujube (Chinese date), lemon, muskmelon, peach, avocado, Korean cherry, plum, grapefruit, kiwi, pineapple, laver, sea tangle, sea mustard.

3) Meats, eggs and its products are chicken, pork, beef, bacon, ham, spam, frankfurt sausage, egg.

4) Fish, shellfish and its products are flounder, hair tail, mackerel, Pacific saury, crab, whip-arm octopus, pacific cord, alaska pollack, anchovy, harvest fish, shrimp(tiger prawn), Spanish mackerel, common squid, yellow croaker, tuna, jelly fish, solen, fish paste, oyster, granulated ark shell, crab shell, little neck clam, hard shelled mussel, spawn of the pollack(salted roe of the pollack).

5) Milk and dairy product are milk, chocolate milk, yoghurt(urd type), yoghurt(liquid), natural cheese, mozzarella cheese, ice cream.

6) Seasoning are soy sauce, kochujang(fermented red pepper soybean paste), soybean paste, mayonnaise, salt, vinegar, ketchup, mono sodium glutamate (M.S.G. chemical seasoning), black pepper, mustard, soup powder, curry powder, strawberry jam.

7) Beverages and oils are corn oil, sesame oil, soybean oil, cider, butter, margarine, coffee(can), cola, sik hye(sweet rice beverage), ginseng tea, mugicha, green tea, black tea, coffee(powder), cream powder, beer, soju(distilled liquor), white wine, orange juice.

2. Experimental method

1) Preparation of reagents

Salicylate used to prepare the standard solution was provided by Dong-A Pharmaceutical Company. The salicylate stock solution(1,500µg/ml) was prepared by dissolving the salicylate in solvent containing acetonitrile(H₂O : acetonitril=1 : 2, v/v). Then 1, 5, 20 and 50µg/ml of the final for standard solutions were prepared by diluting the stock standard salicylate solution. As for the internal salicylate standard,

O-anisic acid was dissolved in distilled deionized water(dd H₂O) to produce 200µg/ml.

2) HPLC condition

The mobile phase consisted of water-methanol phosphoric acid(650 : 350 : 2, v/v/v) and the flow rate for the mobile phase was set at 1.35ml/min¹³. UV detection at a wave length of 305nm and a reversed-phase C₁₈ column were used. The Chart speed was set at 6cm/hr.

The voltage was adjusted to 50mV for the internal standard and 20mV and 10mV for salicylate standard solution. The Detection limit for food was about 0.1µg/ml(per 1ml or 1g of food). The retention time for salicylate and O-anisic acid were 20 and 6 min., respectively.

column	: reverse-phase with C ₁₈ column(3.9×300mm, µBondapak, Waters)
flow rate	: 1.35ml/min
mobile phase	: water : methanol : phosphoric acid(650/350/2, v/v/v)
chart speed	: 6cm/hr
detection limit	: 0.1µg/ml(per 1ml or 1g of food)
retention time	: 20min(salicylic acid) 6min(internal standard)
wave length	: 305nm
detector	: UV detector(model 1306 UV detector, Bio-Rad)
pump	: model 1330 pump(Bio-Rad, Japan Servo Company, Tokyo, Japan)

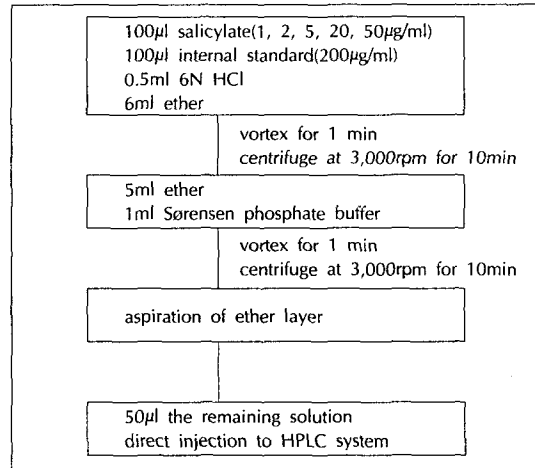
3) Food sample preparation for HPLC assay

(1) Liquid sample

① Standard

For each 1ml of food sample in the test tube, each 100µl salicylate standard solution(1, 5, 20, 50µg) and internal standard solution(200µg/ml) were spiked, and then 0.5ml of 6N HCl and 6ml of ether were added. After separating the water and ether layers by centrifugation, 5ml of the ether layer was transferred to the tube with 1ml of Sørensen phosphate buffer solution(pH 7.0) and vortexed for 1 min and centrifuged at 3,000 rpm for 10 min. After aspirating off the upper ether layer, an aliquot of

50µl from the remaining layer was taken and injected directly to the HPLC system. The following scheme shows the procedure.



② Sample

The method was the same as for the standard, except that the sample solution was spiked instead of salicylate standard solution.

(2) Solid and powder sample

To make the sample solution, 2g of food sample was accurately weighed and dissolved in 5ml of acetonitrile containing solution(H₂O : acetonitrile= 1 : 2, v/v). Then both the salicylate standard solution and internal standard were spiked to the each of 1ml sample solution in the same way with the liquid sample mentioned above. In case of candy, however, it was homogenized after being dissolved in warm water. Also in case of gum, it was solidified by ether first, then crushed.

Results and Discussions

Amounts of salicylate were recorded either in the form of µg salicylate per 1g for solid food samples or µg salicylate per 1ml for liquid food samples. The salicylate contents were also expressed per one serving size of the foods listed in the Korean Nutrition Survey Report. A HPLC chromatogram of sample is shown in Fig. 1.

1. Salicylate contents in plant foods

Salicylate contents in plant foods were shown in

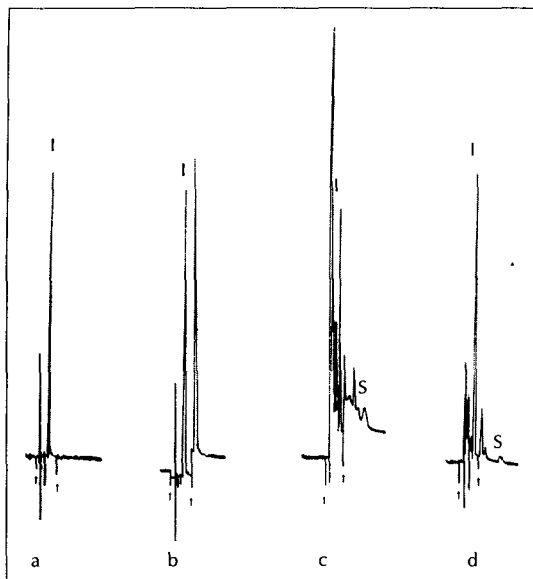


Fig. 1. HPLC chromatogram of sample.
 a : soybean oil b : milk
 c : sesame oil d : chocolate milk
 S : salicylate I : internal standard

Table 1. Chard(8.22 $\mu\text{g/g}$) showed the highest contents of salicylate. In vegetables, soybean sprout(5.13 $\mu\text{g/g}$), perilla leaf(3.03 $\mu\text{g/g}$), Chinese cabbage(2.25 $\mu\text{g/g}$), carrot(2.16 $\mu\text{g/g}$), squash(1.33 $\mu\text{g/g}$) had also considerable amounts of salicylate. Swain et al¹⁴ reported that salicylate content in vegetables was higher than other foods. Especially, red small radish(12.4 $\mu\text{g/g}$), red chilli pepper(12.0 $\mu\text{g/g}$), egg plant(8.8 $\mu\text{g/g}$), salad cucumber(7.8 $\mu\text{g/g}$), alfalfa(7.0 $\mu\text{g/g}$), spinach(5.8 $\mu\text{g/g}$) had high contents of salicylate. Fresh tomato contain only small amount of salicylate (1.3 $\mu\text{g/g}$). However many commonly used tomato product are considerable sources of salicylates : canned tomato 5.3 $\mu\text{g/g}$, tomato paste 14.4 $\mu\text{g/g}$, tomato sauce 23.8 $\mu\text{g/g}$. The increase in available salicylate in processed-tomato product 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 spices¹⁴.

Among vegetables, salicylate content varied widely among the raw group. Hermann et al¹⁵ reported that salicylate in salad cucumber was 1~3 $\mu\text{g/g}$ ¹⁴ and Venema et al¹⁶ was 0.08 $\mu\text{g/g}$. Difference in salicylate contents found in foods by various authors might be caused by differences in origin, processing, storage,

analytical method or of species grown at different locations or by differences in analytical methods⁶. Swain et al found a high natural variation amounting to a 10-fold difference. Venema et al¹⁶ reported a much smaller variation. This excludes natural variation as a possible explanation for these discrepancies. Discrepancies in salicylate contents due to extraction methods may arise through differences in the content of liberation of matrix bound salicylate. We and Swain et al used a UV detector but Vernema et al used Fluorescence detector. Swain et al and Vernema et al estimated acetyl salicylate and salicylate but we estimated salicylate only.

We had found that some fruits contained considerable amounts of salicylate. Orange(5.13 $\mu\text{g/g}$), jujube(3.16 $\mu\text{g/g}$) had the highest amount in fruits. Apple showed low levels of salicylate content(0.47 $\mu\text{g/g}$). Swain et al¹⁴ reported that apples showed considerable variation in salicylate content among varieties(Golden delicious, Red delicious, Jonathan, 0.8 $\mu\text{g/g}$, 1.9 $\mu\text{g/g}$, 3.8 $\mu\text{g/g}$ respectively). Ashoor and Chu¹⁷ found no detectable salicylate in grapefruits, lemon, orange, strawberry and tangelo. Dried fruits have relatively high salicylate contents compared with their fresh counterparts because of the removal of water during the drying process.

Salicylate has not been detected in most of the cereals except in breakfast cereal(0.80 $\mu\text{g/g}$), brown rice (1.04 $\mu\text{g/g}$) and crackers(0.49 $\mu\text{g/g}$). Rice is the most important food item in Korea. Brown rice had detected salicylate but rice had not. Therefore we recommend the rice for allergic patients.

Beans and seeds are also generally safe for salicylate sensitive people.

2. Salicylate contents in animal food

In animal foods, almost no salicylate was detected (Table 2). Beef, pork, chicken, and fish did not contain salicylate. Noid et al¹⁸ had reported some amounts of salicylate in processed meat with vinegar. Dairy products did not contain appreciable amounts of salicylates. Only chocolate milk contained salicylates(0.32 $\mu\text{g/ml}$). No salicylate was found in milk, however, a small amount of salicylate was detected in chocolate milk due to food additives in its processing.

Table 1. Salicylate contents in plant food

Food group	Food item	Food group	Food item	Salicy late(µg)	S.D.	Food group	Food item	Salicy late(µg)	S.D.	Food item	Salicy late(µg)	S.D.
grains	rice		perilla seed	U.D.				N.C.		oak mushroom	U.D.	
	white flour		sunflower seed	U.D.		fruits		N.C.		mandarin orange	U.D.	
	barley		peanut	U.D.				N.C.		persimmon	U.D.	
	breakfast cereals	0.0362	egg plant	0.797	0.00564		apple	0.553	0.00564	0.470	0.0632	
	corn	N.C.	braken	N.C.			pear	U.D.		U.D.		
	job's tears	U.D.	chard	U.D.			watermelon	8.22	5.07	U.D.		
	glutinous millet	U.D.	carrot	U.D.			orange	2.16	1.94	U.D.		0.872
	brown rice	1.04	doraji	U.D.	0.0839		melon	U.D.		U.D.		
	noodles	U.D.	perilla leaf	U.D.			grape	3.03	0.834	U.D.		
	ra myon	U.D.	garlic	U.D.			chestnut	U.D.		U.D.		
loaf bread	U.D.	radish	U.D.			pine nut	U.D.		U.D.			
sponge cake	U.D.	water dropwort	U.D.			walnut	U.D.		U.D.			
cracker	0.489	Chinese cabbage	0.0185			rasin	U.D.	0.0526	U.D.			
potatoes	0.676	leek	0.0888			jujube	U.D.		3.16	0.636		
sweet potato	U.D.	ginger	U.D.			lemon	U.D.		U.D.			
starch vermicelli	U.D.	myungbean sprout	U.D.			musk melon	U.D.		U.D.			
sugars	chewing gum		spinach	U.D.		peach	U.D.		U.D.			
	honey		cabbage	U.D.		avocado	0.740	0.129	U.D.			
	sugar		onion	U.D.		Korean cherry	U.D.		U.D.			
	chocolate		cucumber	U.D.		plum	U.D.		1.65	0.390		
beans	candy		soybean sprout	U.D.		grapefruit	U.D.		2.90	0.478		
	black soybean		tomato	U.D.		kiwi	U.D.		U.D.			
	soy bean	N.C.	green onion	U.D.		pineapple	U.D.		U.D.			
	soybean curd	U.D.	green hot pepper	U.D.		laver	U.D.		U.D.			
	soybean milk	U.D.	squash	U.D.		sea tangle	U.D.		U.D.			
	green peas	N.C.	radish pickle*	N.C.		sea mustard	U.D.		U.D.			
	small red bean	U.D.	mushrooms	U.D.			U.D.		U.D.			
seeds	sesame seed	U.D.	oyster mushroom	U.D.			0.433	0.0964				
			mushroom	U.D.			0.409	0.0711				

U.D. : under detection limit N.C. : non calculable * : pickled radish in rice bran

Table 2. Salicylate contents in animal food

Food group	Food item	Food group	Salicy late(μ g)	S.D.	Food item	Food group	Salicy late(μ g)	S.D.	Food item	Salicy late(μ g)	S.D.
meats & meat products	chicken		U.D.		whip-arm octopus		U.D.		fish paste	U.D.	
	pork		U.D.		pacific cod		U.D.		oyster	U.D.	
	beef		U.D.		Alaska pollack		U.D.		crab shell	U.D.	
	bacon		U.D.		anchovy		U.D.		little neck calm	U.D.	
	ham		U.D.		harvest fish		U.D.		hard shell mussel	U.D.	
eggs & fish & shellfish	spam		U.D.		shrimp(tiger prawn)		U.D.		sprown of the Alaska pollack**	U.D.	
	frankfruit sausage		U.D.		Spanish mackerel		U.D.		granulated ark shell	U.D.	
	egg		U.D.		shrimp		U.D.	milks &	milk*	U.D.	
	flounder		U.D.		common squid		U.D.	dairy	chocolate milk*	0.318	0.0219
	hair tail		U.D.		yellow croaker		U.D.	product	yoghurt(liquid type)	U.D.	
	mackereal		U.D.		tuna		U.D.		yoghurt(liquid type)*	U.D.	
	pacific saury		U.D.		jelly fish		U.D.		natural cheese	U.D.	
	crab		U.D.		solen		U.D.		mozzarella cheese	U.D.	
									ice cream	U.D.	

U.D. : under detection limit N.C. : non calculable * : μ g/ml ** : salted roe of the pollack

Table 3. Salicylate contents in seasonings, beverages and oils

Food group	Food item	Food group	Salicy late(μ g)	S.D.	Food item	Food group	Salicy late(μ g)	S.D.	Food item	Salicy late(μ g)	S.D.
seasonings	soysauce*		U.D.		coffee(can)*		5.00	2.98	white wine*	0.309	0.0391
	kochujang ^①		2.37	0.642	cola*		U.D.		orange juice*	U.D.	
	soybean paste		U.D.		sik hye ^②		U.D.		margarine	U.D.	
	mayonnaise		U.D.		ginseng tea		U.D.		butter	U.D.	
	salt		U.D.		mugicha ^③		U.D.		corn oil*	U.D.	
	vinegar*		U.D.		green tea		N.C.		sesame oil*	0.997	0.0883
	ketchup		U.D.		black tea		N.C.		soybean oil*	U.D.	
	M.S.G		U.D.		coffee(powder)		N.C.		soup powder	U.D.	
	black pepper		8.25	2.21	cream powder		1.19	0.432	curry powder	3.24	0.339
	mustard		4.16	1.02	beer*		U.D.		strawberry jam	U.D.	
beverages	cider*		U.D.		soju ^④		U.D.				

U.D. : under detection limit N.C. : non calculable * : μ g/ml ① : Korean hot pepper paste ② : sweet rice beverage ③ : roasted barley ④ : distilled liquor

3. Salicylate contents in seasonings, beverages and oils

Seasoning materials also contained considerable amounts of salicylate such as black pepper(8.25 μ g/g), mustard(4.16 μ g/g), curry powder(3.24 μ g/g) and kochujang(2.37 μ g/g). According to Swain et al¹⁴⁾, paprika, thyme, dill powder, garam masala, oregano and tumeric were almost as high as curry powder. Although amounts of these condiments used in food are small, they can make a significant contribution to dietary salicylate.

In beverages, almost no salicylate was detected. Only coffee(can) and cream powder(vegetable fat) had salicylate of 5.00 μ g/ml, 1.2 μ g/g, respectively. And it is probably due mainly to the addition of food additives. Swain et al¹⁴⁾ reported that some kinds of coffee(Harris Mocha kenya) and tea(Asco) had low levels of salicylate. In alcoholic beverages,

white wine had salicylate(0.31 μ g/ml). Some researchers reported that wine appears to contain about the same amount as grape juice(range 3.5 to 10 μ g/ml)¹⁵⁾.

Among oils, only sesame oil contained salicylate(1.00 μ g/g)(Table 3).

Major food salicylate sources are vegetables, fruits and seasoning. Food items listed in Table 4 were proved to contain salicylate in our study. The salicylate content of the usual "Western diet" is estimated to range from 10 to 200mg per day¹⁴⁾. However the general salicylate contents from the Korean diet and its main food sources should be systematically studied in the future.

Chronic urticaria and asthma like allergic patients should watch their intake of food items shown in Table 4. High salicylate contained foods on the basis of one serving size-chard, orange, coffee(can), jujube, perilla leaf, grapefruit, plum should particularly be a-

Table 4. Contents of salicylate in commonly used Korean food

Food group	Food item	Salicylate(μ g/g)	Recovery(%)	One serving size(g)	Salicylate(μ g)/one serving
grains	breakfast cereal	0.797	84.93	30	23.91
	brown rice	1.043	79.32	90	93.87
	crackers	0.489	68.02	30	14.67
potatoes	potato	0.676	60.00	70	47.32
vegetables	egg plant	0.553	48.55	70	38.71
	chard	8.220	18.45	70	575.40
	carrot	2.163	11.78	70	151.41
	perilla leaf	3.034	13.36	70	212.38
	Chinese cabbage	2.249	11.78	70	157.43
	cabbage	0.740	19.80	70	51.80
	soybean sprout	5.133	51.93	70	359.31
squash	squash	1.333	25.02	70	93.31
	mushrooms				
mushrooms	oyster mushroom	0.433	18.97	70	30.31
	mushroom	0.409	45.47	70	28.63
fruits	apple	0.470	43.70	100(1 each)	47.00
	orange	5.133	40.74	100(1 each)	513.30
	jujube	3.155	36.10	100	315.50
	plum	1.654	48.54	100	165.40
	grapefruit	2.896	32.06	100(1/2 each)	289.60
milk	chocolate milk*	0.318	41.69	200(1 C)	63.60
seasonings	kochujang	2.368	5.60	5(1 t.s.)	11.84
	black pepper	8.246	24.65	2	16.49
	mustard	4.155	28.67	5(1 t.s.)	20.78
	curry powder	3.236	25.97	20(4 t.s.)	64.72
beverages	coffee(can) *	4.998	33.48	100(1 can)	499.80
	cream powder	1.193	32.39	10(2 t.s.)	11.93
	white wine *	0.309	42.27	100	30.90
oils	sesame oil*	0.997	26.73	5(1 t.s.)	4.99

* : μ g/ml

voided among the allergic patients.

Conclusion

153 common ingredients of Korean cuisine were analyzed in this study : however Salicylate was detected only in 28 of the food items.

Salicylate was predominantly found in vegetables and fruits. In vegetables, chard, soybean sprout, perilla leaf, Chinese cabbage, carrot, squash, cabbage, potato and eggplant listed in the descending order have considerable amounts of salicylate on the basis of one serving size. In fruits, orange, jujube, grapefruit, plum, apple were listed. It was also found in brown rice, breakfast cereals, cracker, potato and mushroom. In animal food, almost no salicylate was found except in chocolate milk. Salicylate was also found in sesame oil, coffee(can), cream powder (vegetable fat), kochujang, black pepper, mustard and curry powder. We identified foods high in salicylate that can be used for determining diets of allergic patients.

We recommend that allergic patients with asthma and urticaria to avoid the foods listed in Table 4.

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