CLIX. THE SULPHUR CONTENT OF FOODS

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MUCH information about the chemical composition of human foods has been collected in recent years, but there are still some wide gaps in our knowledge. Of all the systematic food tables which have been published, only those of Rosedale [1935], Sherman [1937] and possibly Schall [1938] give any original data about sulphur. Of these, Schall's figures are incomplete and sometimes uncertain, and Rosedale used a method [Morris & Rosedale, 1935] which other workers have condemned. Sherman's figures refer mostly to American foods. A search in the original literature for information about the amount of S in foodstuffs has confirmed the paucity of reliable systematic data. Schulz [1893; 1894] appears to have made the first investigation, which is now only of historic interest. Katz [1896] made a limited but very accurate study of muscular tissue. Balland [1907] and Sherman & Gettler [1912] made more extended investigations. More recently Kemmerer & Boutwell [1932] have determined S in 96 American foodstuffs. Their method seems to have been a reliable one, but unfortunately only one sample of each food was analysed. Stotz [1937] analysed 32 different plant and animal substances but he limited the value of his work by expressing the results only as percentages of the dry matter. Echevin & Crepin [1928], Frear [1930], Bertrand & Silberstein [1933, 1, 2; 1936; 1937], Schild & Jacob [1935], Painter & Franke [1936] and Balks & Wehrmann [1937] have determined S in various vegetables and plants. In recent years, since it has been shown that the addition of S or cystine to the diet of sheep improves both growth and wool production, many estimations of S in pasture grass and animal feeding stuffs have been made [Evans, 1931; Woodman & Evans, 1933; Warth & Krishnan, 1937].

Quite a number of authors have determined S in single substances. Thus S in milk has been studied by Steffen & Sullman [1931], Lachmann [1934], Kagi [1937] and Revol & Paccard [1937], and in eggs by Grossfield & Walter [1934] and Marlow & King [1936]. Friese [1929] and Sabalitschka [1931] made a detailed analysis of mushrooms, Le Matte *et al.* [1927] of animal organs used therapeutically, Morse [1929] of cranberries, Cleveland & Fellers [1932] of dates, and Wendt & Wilkinson [1931] and Steudel [1935] of lettuce and potato ash respectively. In each of these cases S was among the elements determined.

Of all these studies none has embraced the composition of foods as prepared for human consumption and few of them have yielded information of much value to those interested in human nutrition. For this reason it has been felt to be desirable to investigate and compare the relative values of a number of methods for estimating S in biological material, and then to make a systematic study of S in foods eaten in this country.

Methods, source of materials and sampling

Dried, mixed samples of nearly all the fruits, vegetables and nuts which had previously been analysed for other constituents by McCance *et al.* [1936] had been sealed up, and were available for analysis. The purchase and preparation of these samples have already been fully described by these authors. S in certain vegetables must be determined on the fresh material (*vide infra*) and new samples of these foods were purchased from different shops, mixed and analysed without being dried. A number of food materials, notably the cereals, were being analysed concurrently for other constituents by McCance & Widdowson [1939]. The collection and sampling of these were carried out in a manner similar to that described for fruit and vegetables. Special purchases of meat and fish had to be made. At least three samples of every variety to be analysed were procured and equal portions of each were mixed and dried before any estimations were made.

S was determined in most instances by a hydrogenation method, though destructive oxidation of the organic matter with a nitric-perchloric acid mixture, followed by a gravimetric estimation of the resulting sulphates as $BaSO_4$ was used in some cases, particularly for the analysis of undried material. Both methods have been described in detail by Masters [1939].

Water was determined by drying to constant weight at 50° or 100°, and N by Kjeldahl's method [McCance *et al.* 1936] using a copper selenite catalyst.

Volatile S compounds

It may safely be said that S in most foods is present as cystine, methionine and stable organic and inorganic sulphates. Bertrand & Silberstein [1929], however, analysed a number of vegetables before and after drying them at 100° and found that onions lost S when so dried. They considered that this was due to the volatilization of essential oils containing S and suggested that the members of the Cruciferae and Liliaceae should be analysed for S in the wet state. Balks & Wehrmann [1937] have also pointed out that onions and horseradish lose S on being dried at 105°. They suggested that this was due to the volatility of allyl mustard oil, C_3H_5CNS, which these plants contain.

Blanck *et al.* [1937] extended these researches a little further. Beet leaves and horseradish were analysed fresh, after being dried at room temperature and at 80–100°. The leaves were found to lose no S, but the horseradish did so whichever way it was dried; as much as 33% disappeared during drying at 80–100°. 10% of the S in white cabbage was found to be volatile. Potatoes were held to lose no S during the process of being dried in the air.

Similar results were obtained by Rippel & Nabel [1937]. Chives, onions, leeks, horseradish, oatseed and asparagus were analysed fresh, and after being dried at 100°. The first four substances are said to contain mustard oil, though only leeks and horseradish were found to lose S on being dried. These losses, moreover, were reduced by a preliminary drying period in the air. No losses were detected when the plants were dried at room temperature.

Table I shows the effect of drying 28 representative foods at 50° for 48 hr. It will be observed that fruit, nuts, cereals, meat and beer lost no S. Fish lost small quantities, probably because it had not been purchased really fresh. Vegetables varied very much. Onions, horseradish, and mustard and cress lost large amounts of S, due no doubt to the presence in them of volatile essential oils. Watercress, savoy cabbage and brussels sprouts also lost some S. Some plants, e.g. cabbage, contain sulphur glucosides, the best known of which is sinigrin. This substance is hydrolysed by myrosin, dilute HCl or more slowly by boiling water, to glucose, $KHSO_4$ and allyl mustard oil [Simpson & Halliday, 1928]. The last is volatile and any which had been formed would be removed on drying at 50°. If, however, sinigrin is the source of the volatile S in cabbage, it is curious that boiled and raw cabbage lost the same amount of S when they were dried. Desiccation did not remove S in significant amounts from parsnips, potatoes, peas or lentils. Vegetables, therefore, which it was thought might contain volatile, or potentially volatile S compounds, were analysed in the wet state, the others were analysed dry.

Table I. *Effect of drying on the S content of foods*

Substance	Determined after drying mg. S/100 g. or 100 ml. fresh material	Determined on fresh material mg. S/100 g. or 100 ml. fresh material
Cereals		
Pearl barley	124·5	121·5
Semolina	138·0	142·3
Drinks		
Beer	23·8	23·3
Fish		
Cod, raw	224·0	215·0
Plaice, raw	210·5	200·0
Meat		
Mutton chop, raw	237·0	233·0
Steak, raw	217·0	216·0
Fruit and nuts		
Apples	3·85	3·73
Bananas	14·4	14·9
Brazil nuts	339·0	337·0
Vegetables		
Brussels sprouts, boiled	97·5	89·2
Cabbage, savoy I, raw	204·0	174·0
Cabbage, savoy II, raw	115·0	96·7
Cabbage, savoy II, boiled	64·2	54·5
Cauliflower, boiled	76·5	79·7
Horseradish, raw	269·0	191·0
Leeks, boiled	90·0	89·0
Lentils, boiled	49·1	48·0
Lettuce, raw	13·7	15·5
Mustard and cress, raw	170·5	89·0
Onions, raw	53·7	36·7
Onions, spring, raw	70·0	47·5
Parsnips, raw	23·2	22·2
Peas, boiled	82·2	80·3
Potatoes, raw	36·2	34·2
Radishes, raw	46·7	42·7
Watercress, raw	61·6	52·8

Nitrogen: sulphur ratios

N had been determined by McCance *et al.* [1936] on all the mixed dried samples of fruits, vegetables and nuts, and an inspection of the N/S ratios has shown that they vary widely from one plant to another. This is probably to be attributed in most instances to differences in the composition of the proteins, but it may well be due to the variations in the inorganic constitution of those fruits with very little N or S in them. Table II gives the results for a few fruits and vegetables and shows the degree of variation. A study of 14 vegetables showed that the ratios were not altered appreciably by cooking. Four typical results are included in Table II. N was determined on all the mixed samples of meat and fish and the ratio was found to be relatively constant for any one class of material. The figures are given in Table IV. This constancy has a practical application for it seems unnecessary for dietary work to determine S in the muscular organs of a large number of animals and fish if N has already been determined; S may be obtained quite accurately enough for all practical purposes from the average N/S ratio. Moreover, the ratio is unaffected by cooking (Table IV), and hence the S in cooked meat or fish may be calculated from the N of the cooked material and the N/S ratio of the raw material. The figures given by McCance & Widdowson [1939] for S in cooked meat and fish were obtained in this way.

Table II. *The N/S ratios of fruits, nuts and vegetables*

Substance	N/S	Substance	N/S	Substance	N/S
Apples, English, eating	5·1	Artichokes, Jerusalem, boiled	11·6	Beans, haricot, raw	20·5
Greengages	40·0	Peas, fresh	18·4	Beans, haricot, boiled	23·0
Oranges	14·0	Spring cabbage, boiled	6·7	Beans, scarlet runner, raw	12·8
Strawberries	7·5	Onions, raw	3·0	Beans, scarlet runner, boiled	12·7
Brazil nuts	7·8	Onions, boiled	3·8	Swedes, raw	4·6
Walnuts	19·3	Onions, fried	3·3	Swedes, boiled	4·6

Visceral organs, the elasmobranch fish and shellfish naturally fall into a different category. The ratio for elasmobranchs (e.g. dogfish) is probably high because of their relatively high non-protein N, whereas that for shellfish is always very much lower than the average figure for fish, possibly because the former are contaminated with sulphate from the sea water.

Results

These are given in Tables III and IV. The figures are expressed as mg./100 g. or 100 ml. of fresh matter (edible portion), and except where stated to the contrary, the results refer to raw materials. The percentage of water is not given in Table III, nor is the percentage of N, since neither was determined in this investigation. For this information reference can be made to McCance & Widdowson [1939]. McCance & Shipp [1933] or McCance *et al.* [1936] may be consulted for the scientific nomenclature of all the foodstuffs.

Discussion

In fruits and most vegetables the amount of S is low and very variable. The figures given were obtained on good mixed samples, but the individual scattering is large, and there may be wide differences between the amounts of S found in the same vegetable purchased on different occasions. This is no doubt partly due to the use of different manures or fertilizers [Davidson & Le Clerc, 1936; Bertrand & Silberstein, 1937]. Nevertheless, the S found in dried apricots and peaches is very much higher than an analysis of the fresh fruit would lead one to expect, and some of the S has probably been added in the form of SO_2 as a preservative [Leach & Winton, 1920]. The high figure for carrageen moss is probably due to large quantities of S in the form of the inorganic $SO_4^{=}$ radicle. This was the only material analysed in which the sulphate was the main acidic radicle.

The amount of S in meat is very much greater than that in vegetables and very much more constant for the different species.

A comparison of the present results with those of previous workers is rather unsatisfactory, owing to the inherent variability of so many of the foods and the uncertainty as to much of the sampling. Moreover, a study of the tables published by Schall [1938], Sherman [1937] and Kemmerer & Boutwell [1932] shows that there are large variations between the figures given for nearly every food analysed by these authors.

Table III. *The S content of foods other than meat and fish. Edible portions only have been analysed*

Name	S, mg./100 g.
FRUITS, RAW	
Apples, Empire, eating	3·7
Apples, English, eating	7·6
Apples, English, cooking	2·9
Apricots	6·1
Avocado pears	19·4
Bananas	13·0
Blackberries	12·5
Cherries, eating	6·8
Cherries, cooking	7·9
Cranberries	11·1
Currants, black	33·1
Currants, red	28·6
Currants, white	23·6
Custard apples	26·7
Damsons	6·4
Figs, green	12·9
Gooseberries, green	15·9
Gooseberries, ripe	13·5
Grapes, black	7·4
Grapes, white	9·1
Grapefruit	5·1
Greengages	3·0
Lemons, whole	12·3
Lemon juice	2·0
Loganberries	18·1
Medlars	16·6
Melons, cantaloupe	11·7
Melons, yellow	6·3
Mulberries	8·8
Nectarines	10·0
Oranges	9·0
Orange juice	4·6
Passion fruit	18·7
Peaches	5·7
Pears, Empire, eating	5·6
Pears, English, eating	2·7
Pears, English, cooking	3·4
Pineapple	2·6
Plums, Victoria, dessert	3·5
Plums, cooking	4·6
Pomegranate juice	4·2
Quinces	5·2
Raspberries	17·3
Rhubarb	8·2
Strawberries	13·4
Tangerines	10·3
Tomatoes	10·7
FRUITS, DRIED	
Apricots	164·0
Currants	30·8
Dates	51·0
Figs	80·8
Peaches	240·0
Prunes	18·5
Raisins	23·0
Sultanas	44·3
TINNED FRUIT	
Tinned apricots	1·0
Tinned fruit salad	1·8
Tinned loganberries	3·0
Tinned peaches	1·0
Tinned pears	1·3
Tinned pineapple	2·7
Bottled olives (in brine)	35·6

Name	S, mg./100 g
NUTS	
Almonds	145·0
Barcelonas	176·0
Brazils	293·0
Chestnuts	29·4
Cobs	74·5
Coconuts	44·0
Coconut milk	23·8
Peanuts	377·0
Walnuts	104·0
VEGETABLES	
Artichokes, globe, boiled	15·5
Artichokes, Jerusalem, boiled	21·6
Asparagus, boiled	46·6
Beans, baked	50·7
Beans, broad, boiled	27·0
Beans, butter, raw	109·5
Beans, butter, boiled	47·2
Beans, French, boiled	8·3
Beans, haricot, raw	166·5
Beans, haricot, boiled	46·3
Beans, runner, raw	14·1
Beans, runner, boiled	9·5
Beetroot, boiled	22·1
Broccoli tops, boiled	45·0
Brussels sprouts, boiled	77·8
Cabbage, red, raw	68·0
Cabbage, savoy, boiled	30·4
Cabbage, spring, boiled	26·7
Cabbage, winter, boiled	23·4
Carrageen moss, dried	5460·0
Carrots, old, raw	6·9
Carrots, old, boiled	5·0
Carrots, young, boiled	9·3
Cauliflower, boiled	29·4
Celeriac, boiled	12·8
Celery, raw	14·9
Celery, boiled	8·3
Chicory, raw	12·7
Cucumber, raw	11·0
Egg plant, raw	9·0
Endive, raw	25·7
Horseradish, raw	212·0
Leeks, boiled	48·9
Lentils, raw	122·5
Lentils, boiled	37·3
Lettuce, raw	11·8
Marrow, boiled	5·5
Mushrooms, raw	33·8
Mushrooms, fried	73·8
Mustard and cress, raw	170·0
Onions, raw	50·7
Onions, boiled	23·7
Onions, fried	87·8
Onions, spring, raw	50·0
Parsnips, raw	16·5
Parsnips, boiled	14·6
Peas, fresh, raw	50·0
Peas, fresh, boiled	43·5
Peas, dried, raw	129·0
Peas, dried, boiled	39·0
Peas, split, dried, raw	166·0
Peas, split, dried, boiled	45·7
Peas, tinned	43·9
Potatoes, old, raw	34·6

Table III (*cont.*)

Name	S, mg./100 g.
VEGETABLES (*cont.*)	
Potatoes, old, boiled	22·2
Potatoes, old, chips	44·7
Potatoes, old, roast	56·3
Potatoes, new, boiled	24·3
Pumpkin, raw	9·5
Radishes, raw	37·5
Salsify, boiled	25·2
Seakale, boiled	52·0
Spinach, boiled	86·5
Spring greens, boiled	28·5
Swedes, raw	39·1
Swedes, boiled	30·5
Sweet potatoes, boiled	14·9
Tomatoes, fried	9·2
Turnips, raw	22·1
Turnips, boiled	21·2
Turnip tops, boiled	39·0
Watercress, raw	127·0
CEREALS AND STARCH PRODUCTS	
All bran, Kellogg's	182·0
Arrowroot	1·6
Barley, pearl	117·0
Biscuits:	
Cream crackers	77·8
Digestive	72·0
Plain mixed	83·4
Rusks	107·0
Sweet mixed	31·8
Water	99·9
Bread:	
Currant	59·4
Hovis	77·3
Malt	114·5
White	54·5
Wholemeal	76·0
Buns, currant	73·4
Cornflakes, Kellogg's	92·5
Cornflour	1·1
Doughnuts	56·4
Dundee cake	55·0
Flour, white	108·5
Flour, wholemeal	123·5
Force	105·0
Grapenuts	145·0
Macaroni	95·0
Oatmeal	155·0
Post Toasties	83·0
Rice	78·5
Ryvita	87·0
Sago	0·45
Semolina	91·8
Tapioca	3·5
Vitaweat	93·2
DAIRY PRODUCTS	
Butter	9·1
Cheese:	
Cheddar	230·0
Dutch	186·5
Gorgonzola	177·0
Gruyère	206·0
Packet	321·0
Parmesan	251·0
St Ivel	186·0
Stilton	228·0

Name	S, mg./100 g.
DAIRY PRODUCTS (*cont.*)	
Cream	33·0
Egg yolk	164·5
Egg white	182·5
Ice cream	30·6
Milk, fresh, whole	29·2
Milk, cond., unsweetened	75·0
Milk, cond., sweetened	82·5
Milk, cond., skimmed, sweetened	94·3
SWEETMEATS, JAMS, ETC.	
Chocolate, milk	67·0
Chocolate, plain	32·0
Cherries, glacé	21·0
Honey	0·8
Honeycomb	0·8
Jam (edible seeds)	6·5
Jam (stone fruits)	3·2
Jelly, packet	36·6
Marmalade	2·1
Mincemeat	28·4
Sugar, Demerara	14·0
Syrup, golden	53·8
Treacle, black	68·5
BEVERAGES	
Bournvita	243·0
Bovril	362·0
Cocoa	160·0
Coffee	110·0
Malted milk, Horlick's	167·0
Marmite	382·0
Ovaltine	183·0
Oxo	321·0
Tea	177·0
Virol	82·9
Beers:	
Pale ale, draught	23·2
Pale ale, bottled	23·8
Mild ale, draught	20·4
Mild ale, bottled	25·2
Strong ale	34·1
Stout	23·1
CONDIMENTS	
Curry powder	86·0
Ground ginger	145·0
Mustard	1280·0
Pepper	99·2
Salt: block	401·0
Table salt A	34·7
Table salt B	23·3
Vinegar	18·6
FATS	
Dripping	9·2
Lard	24·8
Margarine	12·1
Suet	20·0
SAUSAGES AND PASTES	
Beef sausage, fried	163·0
Black sausage	173·0
Breakfast sausage	78·5
Pork sausage, fried	95·0
Fish paste	185·0
Meat paste	131·0

Table IV. *The S content of flesh foods; edible portions only analysed*

Name	Water, g./100 g.	S, mg./100 g.	N/S
MEATS			
Beef, corned	55·6	222	17·0
Beef steak	72·6	203	16·4
Beef steak, fried	59·3	271	15·4
Beef steak, stewed	62·6	287	16·2
Beef, topside	70·4	212	14·8
Beef, topside, stewed	58·0	341	16·1
Chicken, roast	47·2	232	14·9
Duck, roast	42·6	395	13·8
Goose, roast	57·5	326	13·8
Ham, boiled	53·7	233	13·8
Hare, roast	—	347	—
Lamb cutlet	65·3	166	15·1
Mutton chop	65·6	197	15·5
Mutton, leg	52·4	164	15·4
Pheasant, baked	51·5	302	13·7
Pork, leg	66·3	195	16·1
Rabbit	75·0	169	17·8
Turkey, roast	—	234	—
Veal	75·0	191	15·3
Average			15·2
ORGANS			
Brain, sheep's	80·1	108	14·9
Heart, sheep's	75·4	176	14·1
Kidney, sheep's	72·1	141	17·2
Kidney, ox	76·0	154	17·5
Liver, calves'	66·8	264	11·1
Liver, ox	69·0	263	11·8
Sweetbread	71·1	98	20·0
Tripe	81·6	103	20·6
FISH			
Bloaters	69·3	234	12·2
Brill	80·2	172	15·8
Catfish	81·2	149	15·9
Cod	82·6	171	14·1
Cod, baked	71·3	256	13·7
Cod's roe	75·3	212	14·1
FISH (*cont.*)			
Dabs	76·3	227	12·3
Dogfish*	72·4	132	17·4
Eel	70·1	130	14·2
Fillet, smoked, boiled	75·9	249	13·1
Haddock, fresh	80·2	226	12·0
Haddock, smoked	73·4	221	14·7
Halibut, steamed	73·2	227	14·9
Hake	80·3	164	16·1
Herring	68·3	212	14·1
Herring, fried	55·3	270	13·5
Herring's roe*	76·9	175	15·9
Kippers	67·4	225	14·6
Mackerel	70·1	162	16·3
Plaice	81·0	203	12·1
Plaice, fried	68·0	246	12·6
Salmon	72·0	192	16·9
Salmon, tinned	67·3	241	14·6
Sardines, tinned in oil	53·6	246	12·3
Smelts	76·8	168	14·2
Sprats, smoked	62·3	222	15·2
Sole, Dover	78·4	233	12·5
Sole, lemon	79·1	195	13·6
Trout, rainbow	77·7	169	17·2
Turbot	80·0	188	14·1
Whiting	79·9	257	10·9
Whitebait	78·9	208	11·6
Witch	79·1	181	12·6
Average			13·8
SHELLFISH†			
Cockles	65·7	286	5·6
Mussels	81·3	326	5·3
Mussels, boiled	79·0	262	7·9
Prawns	66·3	335	9·9
Scallops	79·6	342	6·5
Shrimps	59·4	339	11·2
Whelks	75·9	401	6·6
Winkles	75·1	265	6·5

* Not included in average. † Analysed as purchased.

The present figures for fruit are of the same order as those given by the above workers, although Schall's results tend to be higher. It is not possible to compare the amounts of S found in vegetables with the results of the other authors, because all their analyses have been made on raw materials. The figures for raw pulses, however, are lower than those of Sherman. An average value only is given for the S in meat and fish by Sherman, but Katz and Schall have analysed the muscular tissues of a limited number of species, and the figures given in the present paper are of the same order and often agree closely with the results obtained by these workers.

The present figure for milk (29·2 mg. S/100 g.) agrees quite well with figures reported by other workers. Steffen & Sullman [1931] gave 30·52 mg., Revol & Paccard [1937] 27·44 mg., Schall [1938] 39·6 mg., Sherman [1937] 34 mg. and Kemmerer & Boutwell [1932] 28 mg.

Schall appears to have been the only one to have analysed different varieties of cheese, and the present authors' results agree with his findings, excepting where his figures are uncertain.

The yolk and white of eggs have been analysed separately, and the values obtained were 164 and 182 mg. S/100 g. respectively. These are in agreement with the findings of Kemmerer & Boutwell [1932], but are lower than those of Grossfield & Walter [1934]. The latter calculated the S in yolk and white from the most reliable figures available for the protein constituents of these substances. In this way they found yolk contained 201 mg. S/100 g. and white 216 mg. S/100 g. which agree well with the figures of 222 and 212 which they obtained on analysis. Schall, however, gives 176 and 276 mg. S/100 g. for yolk and white respectively, and Sherman gives an average figure of 204 mg. 100 g. for S in the whole egg.

Table V. *The amount of S found in cooked dishes, as determined by calculation and analysis*

	S calculated mg./100 g.	S found mg./100 g.
Apple charlotte	12·4	14·0
Fish pie	105·0	108·5
Macaroni cheese	68·0	64·0
Queen cakes	81·0	72·0

As a general test of the reliability and applicability of the figures given in Tables III and IV, four cooked dishes were prepared as they would be normally for human consumption. The ingredients were not "mixed samples". The recipes were known and from these and the change of weight on cooking the S in the cooked dish was calculated, using the data in Tables III and IV. The food was also analysed and it will be seen from Table V that the amount of S found on analysis agrees satisfactorily with the calculated figure.

Summary

1. Drying introduces errors into the determination of S in certain vegetables owing to the presence in them of volatile or potentially volatile S compounds.

2. Figures are given for the total S found in about 300 different foodstuffs.

3. The N/S ratio has been shown to be relatively constant for all muscular organs. The average figure for meat is 15·3 and for fish, 13·8. It is lower than this in all shellfish, and varies from one visceral organ to another. The ratio varies widely in different plants so that generalization about them is not justified.

4. The results have been compared very briefly with those of previous authors.

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