chlordene, unexpectedly, gave a Cl_5 derivative (VI) by loss of the *sec*-chlorine atom rather than the allylic chlorine.

From the postulated structure, III, for α-chlordene it would appear that removal of HCl, to form a third double bond, is possible. However, to date attempts to eliminate HCl with NaOH, NaOMe, or Ag_2CO_3 have failed. The formation of a third double bond in α-chlordene would have given a skeletal comparison with "lumibullvalene", a polycyclic $C_{10}H_{10}$ hydrocarbon which has a similar tricyclo ring system (Scott and Jones, 1972) (Figure 5). However, the slow low-temperature pyrolysis of lumibullvalene gives dihydrolumibullvalene (Jones, 1967; Katz and Cheung, 1969) which has the basic α-chlordene carbon skeleton structure. Also, in keeping with the above photochemical reaction of α-chlordene is the observation that lumibullvalene is rapidly converted, even at −100°, to XX by intramolecular cycloaddition. Also of interest is the similarity of the photocycloaddition structure XVI, from γ-chlordene, to two metabolites of dieldrin (Matsumura et al., 1968) obtained by breakdown of dieldrin by soil microorganisms (Figure 5).

From the postulated structure of α-chlordene the thermal rearrangement to only γ-chlordene can be envisaged. However, this mechanism needs further clarification as do the free-radical mechanisms for the formation of α-, β-, and γ-chlordene from chlordene by the action of Cl_2 or Bz_2O_2. Further work is progressing along these lines together with a full spectral analysis, including ^{13}C NMR and chlorine nuclear quadrupole resonance of these isomers and their derivatives (Gäb et al., 1975a,b).

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Relative Significance of Dietary Sources of Nitrate and Nitrite

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Concern has repeatedly been expressed in the scientific and popular literature about the extent and effects of nitrate and nitrite in our diet. In order to provide some perspective on the amounts, a calculation has been made to evaluate the relative contribution of various known sources of nitrate and nitrite to the U.S. dietary. Combining estimates of per capita consumption of various categories of food and drink with average values for nitrate and nitrite content of these diet components (some only recently available) has made it possible to attribute the nitrate and nitrite intake of the average U.S. inhabitant to major dietary categories. It is estimated that four-fifths of the nitrate intake is from vegetables, and less than one-sixth from cured meats. Other sources (fruits, milk products, water, bread) are not significant. Two-thirds of the nitrite entering the average stomach originates in saliva and slightly less than one-third comes from cured meats. Other sources of nitrite are not significant. Since the average individual is not seen to be at special risk, attention may be directed to examination of individual situations possibly at greater risk due to special diet, age, ethnic background, sex, or area of residence.

Concern over the amount of nitrate in the U.S. diet dates at least from 1907 when Richardson (1907) asserted that most of the nitrate ingested was from vegetables. More recently, it was realized that nitrite, with its tenfold greater toxicity, presented the greater hazard. Since nitrate is easily reduced to nitrite under physiological conditions, there is a need to know the levels of both nitrate and nitrite entering the digestive tract.

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Relationships among ingested nitrate, nitrite, and infantile methemoglobinemia, as well as the interaction between nitrite and secondary amines to produce carcinogenic nitrosamines, have been adequately reviewed elsewhere (Phillips, 1968, 1971; National Research Council, 1972; Wolff and Wasserman, 1972; Fassett, 1973). Some attention has therefore been given to estimating human nitrate and/or nitrite intake. This has been done by calculation for a single meal menu including contributions from (usually

Table I. Estimated Average Daily Ingestion for U.S. Resident

Source	Nitrate mg	Nitrate %	Nitrite mg	Nitrite %
Vegetables	86.1	81.2	0.20	1.6
Fruits, juices	1.4	1.3	0.00	0.0
Milk and products	0.2	0.2	0.00	0.0
Bread	2.0	1.9	0.02	0.2
Water	0.7	0.7	0.00	0.00
Cured meats	15.6	14.7	3.92	30.7
Saliva	30.0[a]		8.62	67.5
Total	106.0	100	12.76	100

[a] Not included in total; see text.

high-nitrate) vegetables and processed meat. Richardson (1907) estimated for two high-nitrate vegetarian meals 1.0 and 1.3 g of NO_3^-. Ashton (1970) estimated a weekly intake of 405 mg of nitrate from meat products, vegetables, and water. Phillips (1968) calculated a 313-mg nitrate content for a "typical Canadian meal consisting of meat, salad, potatoes, . . . a green vegetable, carrots, and a dessert". Fassett (1973) calculated for single portions of processed meat and spinach, with water, an intake of 300 mg of NO_3^- and 20 mg of NO_2^-. Tannenbaum (1972), discussing the significance of various sources of nitrite in the environment, notes that nitrates must also be considered because of their ready conversion to nitrite. He finds leafy vegetables to be "of only occasional significance because consumption of leafy vegetables is low relative to our total food supply". Frequency and quantity of processed meat consumption were also considered too low by Tannenbaum for this item to be a particularly significant part of the environmental nitrite load, but dependent on the individual consumption pattern. Following a suggestion from Sander, he proposed that saliva may be a significant nitrite source, contributing 6–12 mg per day. This had not been previously included in estimates of nitrate and nitrite intake.

To place into perspective the relative contributions of the various sources of nitrate and nitrite to the dietary, a different approach may be useful. Rather than calculations from hypothetical meals, the nitrate and nitrite intake of the average U.S. resident can be calculated using per capita consumption data and production statistics for the various categories of cured meats and other foods.

This has been done, with particular attention to processed and cured meats, an allegedly important nitrite source. Assessment of the contribution of vegetables to nitrite intake has not been possible heretofore because of the paucity of suitable data on nitrite content of fresh and processed vegetables as available to the U.S. consumer. New nitrite data for 34 vegetables obtained in a recent study at this laboratory (Siciliano et al., 1975) were used in the calculations summarized here.

Shown in Table I are an estimate of the daily intake of nitrate and nitrite by the average U.S. resident and the dis-

Table II. Cured Meats: Estimate of U.S. Per Capita Consumption, 1972[a]

		Thousands of pounds
U.S. Production		
Meat placed in cure		
Beef		240,000
Pork		4,090,000
Smoke, dried		
Beef		59,000
Pork		3,330,000
Sausage, dried or semidry, franks, wieners, bologna, other smoked, cooked		2,876,600
Bacon		1,520,000
	Total	12,115,600
		(5,495,510,000 kg)
U.S. Exports		
Beef, pickled or cured		8,900
Pork, hams and shoulders, cured and cooked; bacon; other pickled, salted, or otherwise cured pork; canned ham and shoulders; other canned pork; sausages, bologna, franks		22,140
	Total	31,040
		(14,080,000 kg)
U.S. Imports		
Beef, prepared or preserved, including pickled and cured		56,154
Pork, canned hams, shoulders, bacon		320,460
Pork, other pickled, cured		3,975
Mixed, sausage		6,195
Other, mixed luncheon meats		15,293
	Total	402,077
		(182,378,000 kg)

Net consumed = production minus exports plus imports = 5,663,808,000 kg

U. S. population, 1972, 207,800,000

Consumption, 27.256 kg/capita per yr; 74.7 g/capita per day

[a] Tables 502, 505, 508, Agricultural Statistics (U.S. Department of Agriculture, 1973).

Table III. Nitrate Content of Fresh and Frozen Market Vegetables[a]

	Fresh					Frozen		
Vegetable	Maynard and Barker (1972)	Wilson (1949)	Lee et al. (1972)	Jackson et al. (1967)	Siciliano et al. (1975)	Jackson et al. (1967)	Siciliano et al. (1975)	Average
Potatoes	186	63		57 (2)	120 (99)[b]	132 (2)	150	119 (106)
Tomatoes and products	89	0		71 (4)				62 (6)
Lettuce	753	1109 (2)	279	664 (5)	1210 (3)			850 (12)
Melons		433						433 (1)
Corn							45 (3)	45 (3)
Onions	62			179 (4)			80 (2)	134 (7)
Bean, snap				248 (3)		198 (1)	270 (4)	253 (8)
Pickles[c]				59 (2)				59 (2)
Carrots	142	253 (2)	337	18	72 (8)	194	97 (5)	119 (19)
Cabbage	731	693 (2)	917	314 (2)	784			635 (7)
Beans, dry								13[d]
Peas						62	20 (4)	28 (5)
Celery	2370	2236 (2)	1001	2786 (2)	2220 (3)			2340 (9)
Sweet potatoes				53				53 (1)
Cucumbers					24			24 (1)
Sweet peppers				195 (4)	62	132	50 (3)	125 (9)
Spinach	2397 (4)	1563	2073	238 (2)	2220 (7)	666 (2)	2140 (4)	1860 (21)
Beets	2658	1333	2428	1654	3010 (12)			2760 (16)
Sauerkraut[c]				253 (2)	68			191 (3)
Broccoli		2315 (2)	948			550 (2)	508 (10)	783 (15)
Cauliflower		2000	1054	53			254 (5)	547 (8)
Asparagus		50					16 (6)	21 (7)
Bean, lima				132		88	27 (4)	54 (6)
Pumpkin/squash				299	459 (3)		409 (6)	413 (10)
Eggplant					302			302 (1)

[a] Values in parts per million of nitrate, fresh weight. Converted as needed from original data. Numbers of samples in parentheses. [b] Potato data from Heisler et al. (1973). [c] Reported here under "fresh" though actually processed. [d] From McNamara et al. (1971); see text.

tribution of this nitrate and nitrite among various categories.

It is seen from the table that four-fifths of the nitrate entering the diet originates from vegetables, with less than one-sixth from cured meats. Other sources are insignificant. Two-thirds of the nitrite entering the stomach originates in saliva, and slightly less than one-third comes from cured meats. Other sources of nitrite are not significant.

It cannot be overemphasized that the data shown above do not represent actual ingestion by any individual, but are averages calculated from the rather meager data available. Further, no inferences can be drawn regarding the variations due to special diet, age, ethnic background, sex, or to area of residence.

PROCEDURES AND DATA EMPLOYED

So that the reader may form an opinion of the validity of the estimates in Table I, the procedures by which they were reached are detailed below.

Per Capita Consumption. (a) Data were obtained on the 1972 per capita consumption of 24 vegetables in fresh, canned, and frozen forms. This degree of detail was thought necessary because some vegetables are known to average tenfold higher than others in nitrate content. Data for fresh vegetables were multiplied by appropriate factors to approximate losses in preparing for consumption, since they were originally reported on a farm weight basis. The factors were obtained from Watt and Merrill (1963). Processed (canned, frozen) vegetable data were not thus corrected since they had been reported on a processed weight basis. The values were combined to provide the estimate of total intake of each vegetable. Similar data were obtained for fruits for 1972, but all were combined because literature shows all fruits tested to be similarly low in nitrates and nitrites. Data for fruit and vegetable per capita consumption were taken directly from Tables 283, 285, 288, and 368 in Agricultural Statistics (USDA, 1973).

(b) Using Tables 502, 505, and 508 from Agricultural Statistics, the total U.S. production of meats processed with nitrite was obtained, adjusted for imports and exports, then divided by the 1972 U.S. population. Data are shown in Table II.

(c) Using the Household Food Consumption Survey 1965–1966 Report No. 11 (USDA, 1972), estimates were obtained for per capita intake of bread and milk and milk products.

Nitrate and Nitrite Values. *Vegetables.* These values are based on the recent U.S. literature. Sources used were limited to those providing data on fresh or processed vegetables as available to the consumer, rather than on experimental material. The data used for nitrate are summarized in Table III, which also shows the averages of all values with the number of samples shown.

No nitrate or nitrite values could be located for dry beans available in the market. McNamara et al. (1971) gave a value for Great Northern white seed beans of 3.4 ppm of nitrate N dry weight, which was converted to 13 ppm of nitrate air-dry for use in this calculation. The nitrite values used in the calculation are those reported from this laboratory (Siciliano et al., 1975). The nitrite value for tomatoes is from Rooma (1971). Values for potatoes are from the analytical survey of Heisler et al. (1973).

Attention is drawn to the wide variation in nitrate values of vegetables, due to varietal, agronomic, and climatic influences (Keeney, 1970; Viets and Hageman, 1971).

Fruits. Few data are available for fruits. Rooma's (1971)

Table IV. Cured Meats: Nitrate Content[a]

Item	No. of samples	Sodium nitrate, ppm	
		Mean	Range
"Fresh"			
Bacon	10	172	30–330
Beef	1	440	
Ham	2	680	360–1000
Paté	2	280	140–420
Pork	6	73	20–120
Sausage	3	247	70–380
Tongue	1	60	
Tongue–turkey roll	1	800	
	Total 26	235.3	
		(171 ppm of NO_3^-)	
Vacuum packed			
Bacon	4	77	10–120
Ham	3	417	0–1080
Chopped pork and ham	1	10	
Gammon	1	25	
	Total 9	177	
		(124 ppm of NO_3^-)	
Canned			
Beef loaf	4	186	0–260
Corned beef	31	60	0–260
Ham and beef roll	14	1190	240–1800
Ham and chicken roll	16	868	100–1500
Ham	7	440	20–1600
Ham and pork loaf	9	72	0–450
Tongue	4	85	50–140
All-pork products	12	104	10–440
Pork lunch meat	28	78	0–360
Sausage	4	185	80–300
Tongue–turkey roll	3	355	90–800
Corned mutton	2	240	
Braised kidneys	1	20	
Ham-eggroll	1	40	
	Total 136	361	
		(221 ppm of NO_3^-)	
Grand average	171	208 ppm of NO_3^-	

[a] From Fudge and Truman (1973).

values for strawberries were highest at 15–36 ppm of nitrate, with 13 other fruits ranging between 1.7 and 8.9 ppm of nitrate. Richardson's (1907) values ranged from 0 to 57 ppm of nitrate, while Achtzehn and Hawat (1969) reported an average of 80 ppm of nitrate for bananas, 50 for strawberries, and less than 10 ppm of nitrate for 12 other fruits. In contrast, Bodiphala and Ormrod (1971) reported over 2000 ppm of nitrate for canned apricots and peaches, which they note were anomalous when compared with relatively low values found by others. The lower values used in the present calculation are believed reasonable.

Meat. No definitive information on the amount of nitrate or nitrite in fresh meat was located. A report by Wright and Davison (1964) gives a value of 0.9 ppm of nitrate in meat of dairy cows used as a control in a sodium nitrate feeding study, and states that "the fact that dairy cattle fed barely sublethal levels of nitrate did not pass the substance into the milk, or accumulate it in their tissues, in more than trace amounts is reassuring to the human consumer".

Cured meats of course do contain variable amounts of nitrite, and when it is included in the formula, of nitrate. The use of these additives is in some controversy at present, with some pressure to reduce or if possible eliminate them from processed meats.

Two useful sources of information on the nitrate and nitrite contents of cured meats were employed. Kolari and Aunan (1972) reported 950 analyses of 18 types of meats cured in the United States for nitrite, but not for nitrate. No data of comparable scope on nitrate content of domestic cured meats were found, but a recent English report (Fudge and Truman, 1973) included nitrate data for 171 samples of mostly European meat products. Nitrite values therein, which averaged 13.5 ppm of NO_2^- for 203 samples, were not used. Both sources provided ranges of values for each category of meat product. These data, as well as the averages, are shown in Tables IV and V. The variability in nitrate and nitrite in cured meats is as wide as that of nitrates in vegetables; the content of these anions is known to decline in cured meats during storage (Kolari and Aunan, 1972). This again underlines the assertion that the calculated values shown here are of necessity only approximations.

Bread. No recent information on the nitrate or nitrite content of bread in the U.S. was found. Richardson (1907) reported two samples to contain 40 and 27 ppm of NO_3. Rooma (1971) gave an average of 22 and 27 ppm of NO_3^- for 15 samples each of wheat bread and rye bread, and 0.17 and 0.12 ppm of NO_2^-, respectively. Neither source provides information relating to modern U.S. bread, but since the concentrations of NO_3^- and NO_2^- in both reports are

Table V. Cured Meats: Nitrite Content[a]

Year	Items	No. of samples	Mean, ppm of nitrite	Remarks
1936	10	64	45.6	From nitrate–nitrite combined cure, from retail markets
1937	6	70	62.6	Do
1971	5	48	43.7	Sent to AMI lab for analysis; probably sent shortly after production
1971	12	109	26.4	Sample shortly after production, analyzed within 14 days
1972	11	224	44.4	Analyzed within 1–2 days of production
1972	7	435	63.6	Do
		Total 950	52.5	

[a] Data of Kolari and Aunan (1972). Items included and ranges found were: franks, 0-195; cold cuts, 0-180; bacon, 1.3-272; smoked ham, 0-145; Canadian bacon, picnics, center cut smoked loin, tongue, 0-178; canned hams, 3-130; corned beef, 1-216; lunch meat, 0-180; Vienna sausage, 6-16; Polish sausage, 0-60; knockwurst, 0-60; bologna, 0-185; loaves, 0-65; pork jowls, 50-75; pork belly, 50-100; beef brisket, 0-10; boiled hams, 11-87; canned spiced hams, 5-55 ppm of NO_2^-.

Table VI. Source of Nitrate and Nitrite Ingestion of Average U.S. Resident, 1972[a]

Item	Per capita consumption		Composition, ppm		Daily ingestion, μg	
	lb/yr	g/day	Nitrate	Nitrite	Nitrate	Nitrite
Potatoes, white	96.0	119	119	0.4	14,200	47.6
Tomatoes and products	25.8	31.9	62	1.3	1,980	41.5
Lettuce	17.9	22.2	850	0.4	18,900	8.9
Melons	17.4	21.6	433		9,350	
Corn	13.8	17.1	45	2.0	770	34.2
Onions	9.60	11.9	134	0.7	1,590	8.3
Beans, snap	8.23	10.2	253	0.9	2,580	9.2
Pickles	7.70	9.5	59		560	
Carrots	7.02	8.70	119	0.8	1,040	7.0
Cabbage	6.96	8.63	635	0.5	5,480	4.3
Beans, dry	6.2	7.7	13		100	
Peas	5.70	7.07	28	0.7	198	4.9
Celery	5.52	6.84	2340	0.5	16,000	3.4
Potatoes, sweet	4.02	4.98	53		264	
Cucumbers	2.64	3.27	24	0.5	78	1.6
Peppers, sweet	2.16	2.68	125	0.7	335	1.9
Spinach	1.82	2.26	1860	2.7	4,200	6.1
Beets	1.60	1.98	2760	6.0	5,460	11.9
Sauerkraut	1.40	1.74	191	0.4	332	0.7
Broccoli	1.31	1.62	783	1.0	1,270	1.6
Cauliflower	1.15	1.43	547	1.1	782	1.6
Asparagus	1.10	1.36	21	0.9	28	1.2
Beans, lima	0.99	1.23	54	1.1	66	1.3
Pumpkin/squash	0.74	0.92	413	0.7	380	0.6
Eggplant	0.40	0.49	302	0.5	148	0.2
Total vegetables					86,091	198
All fruits	104.5	129.6	10	0	1,300	0
Juices	43.3	53.7	2	0	107	0
Breads		90	22	0.17	1,980	15
Milk and products		500	0.5	0	250	0
Water		1000	0.71	0	710	0
Cured meats		74.7	208	52.5	15,540	3,920
Saliva		1250	24	6.9	30,000[b]	8,620
				Total μg	105,978	12,753
				Total mg	106.0	12.76

[a] Sources; see text. [b] Not included in total; see text.

relatively small, Rooma's values for wheat bread were used.

Milk. Normal fresh cows' milk is reported to contain less than 1 ppm of nitrate and nitrite (Davis and Macdonald, 1953; Hanni, 1954). A value of 0.5 was assigned for nitrate, since Sander (1967) reported no nitrite in milk.

Water. The analysis of the 100 largest public water supplies in 1962 by the U.S. Public Health Service was used (National Research Council, 1972).

Saliva. Tannenbaum et al. (1974) have reviewed literature on the nitrate and nitrite content of saliva, and verified previous reports that prove ductal saliva contains nitrate but no nitrite. Nitrite is produced from nitrate by oral microorganisms, is relatively constant for an individual, and does not seem to be strongly influenced by composition of a meal (Goaz and Biswell, 1961; Tannenbaum et al., 1974). The average nitrate and nitrite contents for 20 individuals, reported by Tannenbaum et al., were used in the calculations. For the average 24-hr volume of secretion, the midpoint of the range given by Hawk et al. (1954) was used. Brendgen (1932) analyzed saliva from 150 adults and 50 children in Cologne for nitrite content. He was unable to correlate the nitrite content with the time of day, state of health of the oral cavity or teeth, smoking, or frequency of tooth cleaning. Children averaged 31 mg % NO_2^- (range 2.6–76) and adults averaged 42 mg % NO_2^- (range 0–140).

The question of the origin of saliva nitrate remains unanswered. Tannenbaum et al. (1974) showed saliva nitrite to be unaffected by diet but gave no data for the effect of diet on nitrate content of saliva. The calculation in Table VI showing the sources of ingested nitrate assumes salivary nitrate to originate from the food; hence, it is not included in the total of ingested nitrate. Nitrite in saliva, originating as it does in the mouth, is included in intake.

It is not the intent of this paper to discuss the public health implications of this estimate. It is hoped that information will be useful to those who must evaluate the current situation and make recommendations in this area. The impossibility of eliminating all nitrite ingestion by removing it from cured meats is obvious. The data indicate that the average individual is not exposed to an extraordinary risk from the common food and water supply; attention may therefore be focused on finding situations which do present such a risk.

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