CAROTENOIDS OF EDIBLE MOLLUSCS; A REVIEW

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ABSTRACT

This review focuses on the studies reported on carotenoids of edible molluscs in the 1980s. Following an introduction to edible molluscs, it covers the literature published in areas such as localization of carotenoids, sources of carotenoids for molluscs, functions of carotenoids in molluscs and prospective biotechnological uses of carotenoids.

INTRODUCTION

Phylum Mollusca is one of the largest phyla in the animal kingdom with more than 100,000 extant species living in marine, fresh water and terrestrial habitats (Barnes 1980). It is subdivided into seven classes: Gastropoda, Monoplacophora, Polyplacophora, Aplacophora, Bivalvia, Scaphopoda and Cephalopoda. Of these, classes Gastropoda (over 75,000 extant species) and Bivalvia (over 20,000 living species) constitute 95% of the representatives of molluscs. Cephalopoda (with 650 living species) is the other class of economic interest. Cephalopods are also remarkable for attaining the largest size of any invertebrates. Giant squid, *Architeuthis*, located in the North Atlantic Ocean has adult members with a total body length (including 6 m long tentacles) of 16 m, with a body circumference of 4 m. Comparatively, the largest octopus, *Octopus punctatus*, of the Pacific Coast possesses 5 m long arms, with a body circumference of 36 cm.

Carotenoid pigments are abundantly present in marine plants. By predominantly feeding on marine and other aquatic plant sources, molluscs accumulate carotenoids in their body tissues. A portion of carotenoids are metabolized into vitamin A derivatives in the molluscan tissues. Molluscs also play a significant role as a source of food for fishes, sea birds, marine mammals and humans. This vital function of molluscs, serving as an intermediary in incorporating carotenoids into the food chain of higher vertebrates has not been investigated extensively (Czeczuga 1980).

Chessman *et al.* (1967), Fox (1974), Simpson (1982) and Goodwin (1984) have reviewed the literature on molluscan carotenoids published until the end of 1970s. Hence, this review focuses on the studies reported since 1980 on the carotenoids of molluscs.

EDIBLE MOLLUSCS

Molluscs constitute an important food source in many regions of the world (Olley and Throwers 1977; Slabyj *et al.* 1978; Peters 1978; MacKenzie *et al.* 1978). Edible molluscs predominantly belong to classes Gastropoda, Bivalvia and Cephalopoda. The main edible types are:

(a) Abalones, Winkles and Conches
Abalones—*Haliotis* spp.
Horned turban—*Turbo cornutus*
Sea snail—*Concholepas concholepas*
Whelk—*Buccinum* spp.
Periwinkle—*Littorina littorea*
Strombid conches—*Strombus* spp.

(b) Clams, Cockles and Mussels
Clams—*Arca* spp., *Mactra sachalinensis, Spisula solidissima, Venus* spp., *Meretrix lusoria, Venerupis japonica, Saxidomus giganteus, Paphia* spp., *Protothaca thaca, Mercenaria mercenaria, Mesodesma donacium, Mya arenaria, Corbicula japonica.*
Cockles—*Anadara* spp., *Cardium edule*
Ocean quahog—*Artica islandica*
Carpet shell—*Tapes* spp.
Mussels—*Mytilus* spp., *Aulacomya alter, Perna perna*

(c) Oysters and Scallops
Oysters—*Ostrea* spp., *Crassostrea* spp.
Scallops—*Pecten* spp., *Placopecten magellanicus, Argopecten gibbus, Argopecten irradians, Chlamys opercularis*

(d) Squids, Cuttle fishes and Octopuses
Squids—*Sepiola* spp., *Loligo* spp., *Illex* spp., *Todarodes* spp.
Cuttle fishes—*Sepia officinalis*
Octopuses—*Octopus vulgaris, Eledone* spp.

The global catch of edible molluscs in the first half of the 1980s amounted to approximately 7% of the total catch of all aquatic food resources with a mean of 5.79 million metric tons per year (Table 1). The edible molluscs are primarily processed into: (1) canned products; (2) preparations in non airtight containers; and (3) fresh, frozen, salted or dried products. Between 1976 and 1985, the output of mollusc-derived preparations had almost doubled. In 1985, the global production of mollusc-derived food preparations amounted to (1) 125,296 metric tons of canned products; (2) 211,386 metric tons of preparations in non airtight containers; and (3) 982,054 metric tons of fresh, frozen, salted or dried products (FAO 1987). Major countries which process molluscs for edible purposes are, Japan, South Korea, Thailand, USA, Canada, Mexico, Chile, France, Spain and New Zealand (Table 2).

Though molluscs are mostly harvested from the sea, they are also actively cultivated in Japan (Ventilla 1982, 1984), USSR (Kucheryavenko 1985; Brykov *et al.* 1987; Ivanov *et al.* 1987; Lukanin *et al.* 1987; Shepel' 1987; Zhirmunskii

TABLE 1.
GLOBAL CATCH OF MOLLUSCS IN THE 1980S
(MILLION METRIC TONS)

Types	Year					
	1980	1981	1982	1983	1984	1985
Squids, cuttlefishes & octopuses	1.542	1.374	1.626	1.697	1.659	1.746
Clams, cockles & arkshells	1.232	1.319	1.276	1.327	1.510	1.641
Oysters	0.972	0.957	0.968	0.999	0.991	1.026
Mussels	0.672	0.714	0.809	0.774	0.761	0.798
Scallops & pectens	0.370	0.576	0.524	0.556	0.834	0.596
Fresh water molluscs	0.272	0.254	0.284	0.296	0.286	0.261
Abalones, winkles & conchs	0.086	0.089	0.099	0.083	0.085	0.078
Miscellaneous marine molluscs	0.101	0.116	0.113	0.127	0.134	0.147
TOTAL	5.247	5.399	5.699	5.859	6.260	6.293

Adapted from FAO (1987)

TABLE 2.
GLOBAL PRODUCTION OF CANNED MOLLUSC PRODUCTS IN 1985

Commodity	Amount (metric tons)	Producing Countries
Clam chowder	33,802	USA
Clam meat	22,464	USA, Canada, Chile, Italy, S.Korea, Spain, Venezuela
Mussel meat	20,313	Spain, Chile, Denmark, S.Korea, New Zealand
Oyster meat	11,560	S.Korea, Japan
Mussels	11,000	Netherlands
Cephalopods	9,252	Spain, Mexico, Portugal, Thailand, USA
Other clam specialities	7,933	USA
Other univalves meat	2,141	Chile, S.Korea
Scallop meat	1,300	France
Other oyster specialities	1,002	USA
Abalone meat	888	Mexico, New Zealand, South Africa
Other molluscs	3,641	USA, Argentina, Belgium, Chile, Spain

Adapted from FAO (1987)

1987) and the European Economic Community nations (Kirk 1979) for edible purposes.

LOCALIZATION OF CAROTENOIDS

The three prominent classes of molluscs are gastropods, bivalves and cephalopods. With the exception of cephalopods, molluscs are characterized by the presence of a hard external shell. Gastropods are univalve forms, while bivalves (also known as lamellibranchs and pelecypods) possess two valves. Percent of soft tissue (meat plus shell liquor) in the whole bivalve ranges from 30 to 50% (Shimizu and Monma 1968; Shimizu and Narahara 1968; Shimizu and Ohta 1968; Shimizu and Oda 1968; Shimizu and Uchida 1968). Carotenoids are predominantly located in the various regions of the soft tissues.

Goodwin's (1984) compilation of the various molluscan carotenoids reveal that β-carotene is present in many edible molluscs such as abalones (*Haliotis* spp.), oysters (*Ostrea edulis*), scallops (*Pecten* spp.) as well as cockles and clams (*Anadara* spp., *Cardium edule, Meretrix lusoria, Spisula* spp., *Mya arenaria, Tapes pullastra* and *Venus* spp.). Of the other carotenoids which have biopotency, α-carotene has been reported in a few species of fresh water, marine and terrestrial gastropods. γ-Carotene is also present in some of the fresh water and marine gastropods.

Carotenoid content in most of the edible molluscs varies between 10 and 140 μg/100g of raw edible portion (Table 3). However, *Cipangopaludina japonica*

TABLE 3.
PROTEIN, LIPID, RETINOL AND CAROTENE CONTENT OF SOME EDIBLE MOLLUSCS

Edible Molluscs	water (g)	protein (g)	lipid (g)	retinol (ug)	carotene (ug)
	--------per 100g raw edible portion-------				
Ark shell Scapharca broughtonii	78.0	15.7	0.5	30	60
Short necked clam Tapes philippinarum	86.8	8.3	1.0	12	40
Mussel Mytilus coruscus	82.9	10.3	1.4	34	0
Oyster Crassostrea gigas	81.9	9.7	1.8	7	55
Top shell Batillus cornutus	76.7	19.9	0.4	0	140
Corb shell Corbiculina leana	87.5	6.8	1.1	0	18
Vivipara Cipangopaludina japonica	78.8	13.0	1.1	15	960
Whelk Neptunea arthritica	79.0	16.4	0.5	0	19
Hen clam Buccinus spp.	84.4	11.8	0.6	12	18
Surf clam Spisula sachaliensis	75.8	17.8	1.3	6	10
Mogai clam Scapharca subcrenata	85.3	11.8	0.9	45	50
Scallop Patinopecten yessoensis	81.2	13.8	1.2	0	15
Squid Sepia spp.	81.8	15.6	1.0	3	0

Adapted from Resources Council, Japan (1982).

has been reported to contain 960 μg/100g of raw edible portion. It should be noted that the amount of carotene detected in a mollusc at a particular time of assay is dependent on factors such as (1) sexual maturity; (2) seasonal variation; (3) category of algal diet consumed by the animal; and (4) whether the animal belongs to natural stock or artificially reared stock. Generally, carotenoids are also not uniformly distributed in animal tissues (Karnaukhov *et al.* 1977).

In scallops, carotenoids are present in the ligament, mantle, ovary and spermary (Shimizu and Oda 1968; Miki *et al.* 1982). In the four species studied by Miki *et al.* (1982), carotenoid content ranged from 1.56 mg/100g in the ovaries of *Patinopecten yessoensis* to 7.15 mg/100g in *Chlamys nipponensis okazara*. Pectenolone, pectenoxanthin, pectenol and astaxanthin constitute the major carotenoids in the ovaries of scallops (Matsuno *et al.* 1981).

In a series of studies, Matsuno's group in Japan reported the carotenoid profile of various edible molluscs, such as surf clam, shortnecked clam, sea mussel, scallop, spindle shell, abalone and turban shell (Table 4). The total amount of carotenoids present in the reddish orange muscle tissue of the spindle shell *Fusinus perplexus* ranged from 4.5 to 7.6 mg/100g muscle weight (Matsuno *et al.* 1984a; Matsuno and Tsushima 1989). More than twenty carotenoids have been reported to be present, of which (3S)-phoenicoxanthin, 4,4'-dihydroxy pirarixanthin and canthaxanthin are the predominant forms. In addition, echinenone, fritschiellaxanthin, zeaxanthin, alloxanthin, (3S,3'S) astaxanthin and β-carotene were also identified. Matsuno *et al.* (1984a) inferred that the carotenoid patterns of the spindle shell, a gastropod, differs from those reported of bivalves in that the ketocarotenoids reported in the muscle tissue of the spindle shell resemble the ketocarotenoids characteristic of crustaceans. In a subsequent comparative study of three species of spindle shell, Katagiri *et al.* (1986) reported that the total carotenoid content in the (1) muscle ranged from 2.7 to 10.6 mg/100g tissue; (2) gonad and hepatopancreas ranged from 3.5 to 8.5 and 2.0–7.8 mg/100g tissue, respectively. While the gonads had β-carotene, isocryptoxanthin and echinenone as the major components, β-carotene and halocynthiaxanthin dominated the carotenoid profile in the hepatopancreas of the three species.

Based on the carotenoid analyses in the spindle shell *Fusinus perplexus* and edible mussel *Mytilus edulis*, Matsuno (1985), Maoka and Matsuno (1989) as well as Partali *et al.* (1989) proposed the derivation of different carotenoids by oxidative (and reductive) metabolic pathways. These include the following:

(1) β-carotene → echinenone → canthaxanthin → (3S)-phoenicoxanthin → (3S,3'S)-astaxanthin.
(2) lutein A → fritschiellaxanthin
(3) fucoxanthin → fucoxanthinol → halocynthiaxanthin → mytiloxanthin
fucoxanthinol → amarouciaxanthin A

TABLE 4.
CAROTENOIDS FROM EDIBLE MOLLUSCS REPORTED IN THE 1980S

Molluscan species	Carotenoid	Reference
Abalone *Haliotis discus*	3R,3'R-zeaxanthin	Maoka et al. (1986)
Edible mussel *Mytilus edulis*	alloxanthin	Partali et al. (1989)
	mytiloxanthin	
Sea mussel *Mytilus cornuscus*	pectenol	Matsuno et al. (1984c)
	pectenol B	Maoka & Matsuno (1988)
Scallop *Patinopecten yessoensis*	3R,3'R-astaxanthin	Matsuno et al. (1984b)
Short necked clam *Paphia euglypta*	amarouciaxanthin A	Matsuno et al. (1985)
Spindle shell *Fusinus* spp.	alloxanthin	Matsuno et al. (1984a);
	3S,3'S-astaxanthin	Katagiri et al. (1986);
	canthaxanthin	Matsuno & Tsushima (1989)
	β-carotene	
	diatoxanthin	
	echinenone	
	fritschiellaxanthin	
	fucoxanthin	
	isocryptoxanthin	
	lutein A	
	mytiloxanthin	
	3S-phoenicoxanthin	
	3R,3'R-zeaxanthin	
Surf clam *Mactra chinensis*	mactraxanthin	Matsuno & Sakaguchi (1983)
Turban shell *Batillus cornutus*	3R,3'R-zeaxanthin	Maoka et al. (1986)

The structures of newly reported carotenoids have appeared in Matsuno (1985) and Partali *et al.* (1989).

Astaxanthin is one of the major carotenoids present in many marine phyla. A comparative survey of the occurrence of the stereoisomers of astaxanthin among the phyla revealed that in the muscles of molluscan species *Semisulcospira libertina, Fusinus perplexus* and *Patinopecten yessoensis,* more than 99% of the astaxanthin is present as 3S,3'S-astaxanthin (Matsuno *et al.* 1984b).

In comparison to the studies on the carotenoids of gastropods and bivalves, the carotenoids of cephalopods have not been investigated in detail so far. Recently, Maoka *et al.* (1989) reported the presence of carotenoids in the gonads of *Watasenia scintillans* and the livers of *Sepia modokai, S.officinalis, Octopus vulgaris, O.ocellatus* and *O.minor*. Free and esterified forms of astaxanthin were identified as the major components (70–80%) in these six species. The carotenoid profile of cephalopods resembled those of the crustaceans, and since the latter constitute the prey of cephalopods, Maoka *et al.* (1989) postulated that the resemblance is due to the food chain relationship shared by the cephalopods with crustaceans.

Carotenoids, in combination with protein, could also occur as carotenoproteins in the molluscs. But, with the exception of a few reports (Arvanitaki and Chalazonitis 1949a,b; Goodwin and Taha 1950; Paparo and Murphy 1978), hardly any studies have been made on the carotenoproteins of molluscs.

SOURCES OF CAROTENOIDS FOR MOLLUSCS

Traditionally, carotenoids originate as a component of a mollusc's algal food (Miki *et al.* 1982; Goodwin 1984). Gastropods have a multitude of food preferences; herbivorous, carnivorous, detritophagus or any opportunistic feeding habit including carrion-eating. Among the herbivores, the marine gastropods feed on micro and macro-algae. Tender parts of aquatic and terrestrial vascular plants, decaying vegetation and fungi constitute the diet of fresh water and terrestrial gastropods. Most of the bivalves feed on fine phytoplankton as minute as 1 μm. Cephalopods are adapted for a carnivorous diet (Barnes 1980).

Japanese abalone, *Haliotis discus hannai,* fed with alga *Ulva pertusa,* accumulated β-carotene and lutein in their shells (Tajima *et al.* 1980a). β-carotene, fucoxanthin and neofucoxanthin were found commonly in the juvenile abalone shells and in diatoms fed to abalones. Muscles of mature abalones predominantly contained β-carotene. Other types of carotenoids were also detected in the testis and ovary of mature abalones (Tajima *et al.* 1980b).

It is also possible that carotenoids could occur in the molluscan body tissues, if they harbor symbiotic microbes which produce carotene pigments. Van den Branden *et al.* (1980) reported that the accessory nidamental glands of the cuttle fish *Sepia officinalis* show an intense orange-red coloration in mature females. This color was found to be due to carotenoid pigments occurring in the symbiotic bacteria which are located in the tubular lumina of the glands. Of the six different pigmented strains of bacteria isolated, β-carotene was the main pigment in five. The bacteria were not identified in this study.

FUNCTIONS OF CAROTENE IN MOLLUSCS

Some of the molluscs possess extreme anoxic tolerance. Karnaukhov (1971) was of the opinion that carotenoids present in the cytosomes assist in maintaining anoxic tolerance by storing oxygen. Presence of conjugated double bonds in the form of unstable peroxides or aldehydes is suggested as a possible way of providing an intracellular reserve of oxygen. Zs-Nagy (1974) provided an alternative opinion which excludes the possibility of the role of carotenoid.

From a study of nine species of bivalves and two species of gastropods inhabiting the littoral region of the Black Sea, Karnaukhov *et al.* (1977) found that (1) the carotenoid content in the body tissues of molluscs showed an increase, depending on the polluted region of the sea in which the species live and (2) the molluscs with high carotenoid content in their body tissues showed a high resistance to environmental pollution. From these results, the Soviet researchers deduced that the carotenoids take part in oxygen metabolism of molluscs. However, other laboratories have been unable to reproduce their studies (Goodwin 1986).

Paparo and Murphy (1978) have investigated the two neuronal chromoproteins, one of which is a yellow carotenoprotein, which initiate changes in ciliary activity on the gill of clam *Mytilus*. They inferred that, since the photic, electrical and chemical stimulation of these chromoproteins correlate with fluctuations in calcium and iron deposits in gill bulk tissue, this is evidence that carotenoproteins are involved in the ionic transport of clams.

The physiology of ink secretion in Cephalopods or the biochemistry of the ink itself have fascinated zoologists for a long time. Fox and Crane (1942), while investigating the pigments present in two-spotted octopus *Paroctopus bimaculatus* and the common squid *Loligo opalescens*, reported that the liver-pancreas of octopus contained a variety of carotenoids amounting to 3.5 mg lutein equivalents/100g moist tissue. The ink sac, adjacent to the liver-pancreas, had 0.55–0.70 mg lutein equivalents/100g. Also of interest was the observations that (1) starvation resulted in the gradual disappearance of carotenoids from liver and ink; (2) autolysis of the liver or incubation of its ground tissues resulted in only a slow loss of the pigments. Since the release of ink by Cephalopods is interpreted as an escape mechanism to disorient the oncoming predator (Barnes 1980), carotenoids seem to play a protective role to the Cephalopods. It is unfortunate that the results of Fox & Crane have not been substantiated further in recent years. In a recent survey of three species of *Octopus* and six species of squids (including *Sepia officinalis* and *Loligo edulis*), Maoka *et al.* (1989) couldn't identify any carotenoids in the ink secretions.

RESEARCH NEEDS FOR BIOTECHNOLOGICAL USES OF CAROTENOIDS

In this decade, marine gastropods have elicited much interest among biochemists and pharmacologists in the search of naturally occurring chemicals which may act as anti-tumor agents (Matsuno 1985). In this respect, carotenoids from molluscs offer good prospects for study on antitumor activity.

Carotenoids are also used in the food industry as natural food additives to provide yellow, red and orange pigmentation to foods (Newsome 1987). A range of popularly consumed food products such as ice cream, egg nog, macaroni products, soups, jams, jellies, juices, butter, cheese, margarine and salad dressing derive their delightful yellowish orange colors due to addition of β-carotene, β-apo-8'-carotenal and canthaxanthin. Hence, carotenoids from nonedible portions of molluscs which are presently wasted could be extracted for use as food and feed additives.

Extensive studies are needed for maximum utilization of carotenoids present in the molluscs. These include, studies on (1) stability of molluscan carotenoids under various processing conditions; (2) efficient practical methods for extraction and purification; (3) prevention of loss of biopotency using protease inhibitors, antioxidants and antimicrobial agents; and (4) bioassays on antitumor activity.

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