Accelerated bioaccumulation of total mercury in red stingray, *Hemitrygon akajei*, by ontogenetic changes of feeding habits, and selective transfer of methylmercury to the aquatic animals located at higher trophic levels

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Abstract

Mercury is a trace element circulating in the biosphere on the earth, but nevertheless some of its organic forms are highly harmful to various living organisms. There is an increasing awareness of the progress of bioaccumulation of mercury concentration in various marine ecosystems in the world, since 7,400 tons of mercury is still annually being emitted into the environment throughout the world not only from anthropogenic sources, but also natural ones including volcanos, and finally discharged to the sea. Volcanic emission has a non-negligible impact on the mercury levels of the aquatic environment. In particular, the Japanese archipelago is located on the Circum-Pacific "ring of fire", and has with about 85 active volcanoes, which have erupted repeatedly, dispersing a large amount of mercury to the atmosphere surrounding them. Approximately 1,879,000 tons of fish and shellfish have been annually caught to obtain seafoods from the coastal seas in Japan. In this meaning, approximately one hundred and twenty-five million people living in the Japanese archipelago seem to be inevitable to face the risk of bioaccumulation of mercury in their daily lives through the consumption of sea foods.

This study deals with the trophic transfer of mercury species in the bentho-pelagic food web in the Isahaya Bay, focusing on the bioaccumulation process in one of the edible ray species, red stingray (*Hemitrygon akajei*). This bay is located in the western side of the inner part of Ariake Bay, Kyushu, western Japan. An active volcano, Mt. Unzen Fugendake, is located at the center of Shimabara Peninsula, which faces the southern side of the bay. In its last eruption over 1,928 days between 1990 and 1995, about 2.95 tons of mercury were emitted to the atmosphere around it. The volcano works as one of the natural discharging sources of mercury to the bay.

In this study, I collected 22 individuals of red stingray and six species of other fishes and invertebrates from Isahaya Bay, and aimed at clarifying the mechanisms how the total mercury (THg) content of the body tissues of red stingray markedly increases as it grows up although its trophic position tends to descend in the food web system and how the mercury is transferred to the animals located at the higher trophic positions, and evaluated the potential risk of mercury intake to the human health through the dietary intake of fishes captured in the coastal seas.

In the Isahaya Bay, the THg content of the muscles of red stingray acceleratedly following the growth ($y = 160.0e^{0.03x}$, x = the disc width, y = the THg content of the muscles). The THg contents of the immature female (disc width: 25 ± 5 cm, body weight: 623 ± 421 g) are 309 ± 76 ng g^{-1} d.w. (mean ± S.D., n=7), while those of the mature ones

(disc width: 60 ± 3 cm, body weight: 8,175 ± 1,023 g) increased to 869 ± 268 ng g^{-1} d.w. (n=10). The highest value, 1,370 ng g^{-1} d.w., was recorded in the individuals with the disc width of 63 cm and the body weight of 8,615 g. However, the results of stable analysis of carbon and nitrogen of the muscles of them revealed that the trophic positions of red stingray tended to descend in the food web system as it grew up. The analysis of the stomach content of these individuals indicated the presence of the ontogenetic changes of the feeding habits following the development of mouth. In mature female, it can feed on only epifauna macro-benthic animals with soft bodies such as shrimps and small crustaceans, while mature female tends to prefer to prey on infauna macro-benthic animals with hard shells such as short-neck clam, *Ruditapes philippinarum*, and some polychaetes. These main food items burrow the sediment, and contain much higher levels of THg to the epifaunal ones, since they are apt to expose to the mercury deposited and concentrated in the sediment.

In the Isahaya Bay, a large amount of mercury has been emitted from an active volcano, Mt.Unzen Fugendake, and a part of the mercury is absorbed by phytoplankton and deposited in the sediment of the sea floor in the bay, being concentrated in the body tissues of the phytoplankton and the sediment. The mercury deposited in the sediment was taken in to the body tissues of the macro-benthic animals on the sea floor though their feeding activities and exposure to the mercury, and transferred to the animals located at the higher trophic positions through the bentho-pelagic food web of the bay. In this transferring process of mercury between the prey and predator, inorganic mercury (InHg) tends to be excreted to outside the body of each animal, while methylmercury (MeHg) is apt to remain in the body tissues and selectively transferred to the predator side. Therefore, the results of the analysis on Trophic Magnification Factor (TMF) values of the mercury species in the seven species of animals liked with the bentho-pelagic food web system revealed that the biomagnification of MeHg of the muscles proceeded approximately 2.4 (6.22/2.62) times in the animals located at one step higher trophic position in the food web system. Consequently, MeHg of the muscles of short-neck clam as the primary consumer occupied approximately 22.5 % of THg, while it increased to more than 90 % in the predatory fishes as the intermediate consumers between the secondary and tertiary ones, such as *Pennahia argentata*, *H. akajei*, and *Nuchequula nuchalis*.

In the case of the red stingray, two large individuals of mature females (56 and 63 cm of the disc width) contained the highest values of MeHg of 0.26 μg g^{-1} w.w.. They are very close to the provisional reference value of MeHg for fish and shellfish in Japan, 0.3 μg g^{-1} w.w. in Japan, although their THg contents (0.28 and 0.27 μg g^{-1} w.w., respectively) were far lower than that of THg for fish and shellfish, 0.4 μg g^{-1} w.w.. These facts indicate

that we need to pay more attention to the effects of the selective transfer of MeHg among the mercury species in the bentho-pelagic food web system in the coastal seas.

Keywords: biomagnification, *Hemitrygon akajei*, total mercury, methylmercury, ontogenetic changes of feeding habits, red stingray, *Ruditapes phillipinarum*, selective transfer, short-neck clam

アカエイ（*Hemitrygon akajei*）の成長に伴う食性変化による加速度的な総水銀の生物濃縮と高次栄養段階に位置する水棲動物へのメチル水銀の選択的移行

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