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What do we know about whales and the ecosystem in the western North Pacific Ocean? Part 4: Summary of results on chemical pollution

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ABSTRACT

Three baleen whale species, common minke, sei and Bryde's whales were surveyed during the former Japanese whale research under special scientific permit programs in the western North Pacific Ocean. They were conducted from 1994 to 2019. The Institute of Cetacean Research collected and analyzed samples in order to investigate the concentration of chemical pollutants and the effect of those pollutants on the health of the whales. This paper summarizes the results found for mercury (Hg), Persistent Organic Pollutants (POPs) and radioisotope concentrations in the whales collected during the research programs. Whales are suited as biological indicators of environmental conditions because they are long-living animals with the ability to migrate long-distances. In particular, the former JARPNII research program investigated pollutant concentrations of the sampled baleen and toothed whales in relation to their sex and maturity. In addition, JARPNII investigated pollution concentrations in prey species collected from the stomach contents of whales as well as those in samples of seawater and air obtained from the research area at the time of survey. The results from direct examinations of different types of biotic and abiotic sources revealed very important information in furthering our understanding of the bioaccumulation process through the marine food chain.

INTRODUCTION

Pollutants such as Persistent Organic Pollutants (POPs) and mercury (Hg) are generally released from land and transported to coastal and pelagic waters in run-offs as well as by atmospheric dissemination and other ways. Higher trophic animals such as cetaceans generally accumulate POPs through the marine food web (Sanpera *et al.*, 1993; Borrell and Reijnders, 1999). The monitoring of pollutants concentrations in the marine environment through the examination of biological tissues of marine mammals is important, since marine mammals can serve as biological indicators of environmental conditions. Large cetaceans may be particularly useful as they are long-living animals migrating long distances.

In 1995, the International Whaling Commission (IWC) decided to 'give priority to research on the effects of environmental changes on cetaceans …' (IWC Resolution 1995/10), and its Scientific Committee (SC) decided to treat the adverse effects of pollutants in cetaceans as one of its highest priority issues.

Japan's whale research programs under special permit in the western North Pacific were conducted for approximately 25 years, from 1994 and 2019. The main target species of the programs were the common minke (*Balaenoptera acutorostrata*), Bryde's (*B. edeni*) and sei (*B. borealis*) whales. During the surveys, the Institute of Cetacean Research (ICR) collected and analyzed samples in order to investigate the concentration of chemical pollutants and the effect of those pollutants on the health of the whales.

At this point, it was considered important to summarize the knowledge on whales and their environment accumulated so far by Japan's whale research in the western North Pacific. The objective of this paper is to summarize the most relevant outputs of pollutant studies based on samples and data collected during the former whale research programs under special permit in the western North Pacific.

SUMMARY OF POLLUTANT MONITORING IN THREE BALEEN WHALE SPECIES

The ICR uses a cold-vapor atomic absorption spectrometry, ELISA, UPLC-MS/MS and UPLC-FDA for measurements of pollution and biomarker concentrations in whales (Figure 1).

Table 1 shows the measurement systems and the standard procedures used at the ICR in recent chemical pollutant studies of North Pacific baleen whales. Results on chemical pollutants in whales presented in this paper are based on those systems and procedures.

Laboratory and analytical procedures to determine pollutant concentrations in baleen whales have been described in previous documents (Yasunaga *et al.*, 2016; Yasunaga and Fujise, 2016a; b). Details of the procedures are not repeated here for the sake of brevity. The main results are presented in this section for three baleen whale species, by type of chemical pollutant.

Mercury (Hg)

It is known that Hg is mainly released on land as elemental Hg from natural sources such as volcanic activities and anthropogenic sources, e.g. thermal power plants. Once released, elemental Hg is transported to the pelagic ocean and pelagic surface water (Fitzgerald *et al.*, 1998). In the pelagic ocean, elemental Hg is converted into methyl Hg and bioconcentrated at moderate levels in animals of high trophic level, such as cetaceans, through the food chain (Knauer and Martin, 1972). Therefore, in predicting changes in Hg concentrations in baleen whales and their environment, factors such as food habitat of whales and biological information such as sex, age and pregnancy, are important.

Yasunaga *et al.* (2016) examined temporal changes in Hg concentrations (including all chemical forms of Hg such as methyl Hg and inorganic Hg, i.e. total Hg) of baleen whales using muscle samples of mature males of common minke whales for the period 1994–2014, sei whales for the period 2002–2014 and Bryde's whales for the period 2002–2014 from the western North Pacific. The research areas surveyed are shown in Figure 2. The

Figure 1. UPLC-MS/MS and FDA system at the environmental chemistry laboratory of the Institute of Cetacean Research.

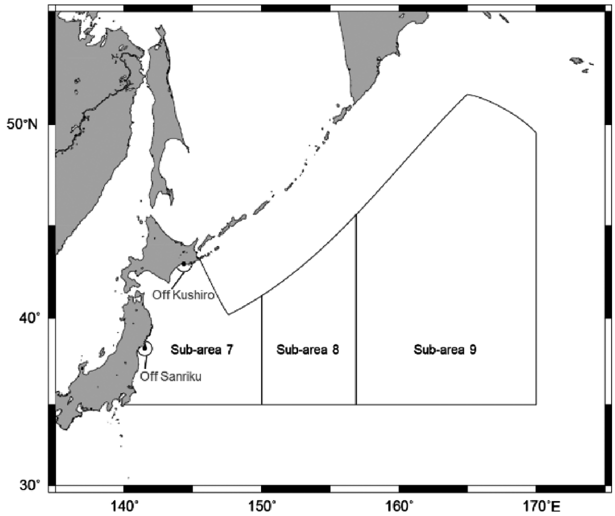

Figure 2. Sub-areas surveyed by the former Japanese whale research program under special permit. Sub-areas are based on IWC (1994), and they have been used by the IWC for the management of common minke whales.

Table 1

Measurement systems and the standard procedures used at the Institute of Cetacean Research in the recent chemical pollutant studies of North Pacific baleen whales.

Substances	Total Hg	Total PCB	Legacy POPs for determination of each isomer	Radioisotopes (the I131, Cs134 and Cs137)
Measurement systems	Direct Thermal Decomposition-Gold Amalgamation-CVAAS	Analytical method of PCBs using GC-ECD with a packed colomun	A sample decomposed with alkaline solution and extracted with hexane are measured by GC/MS-SIM	Containers were placed in 0.04-mm thick polyethylene bags and a germanium semiconductor detector
Standard procedures	Provisional regulation levels of Hg in fisheries by Ministry of Health and Welfare Japan (1972)	The public analytical method of Japan by Ministry of Welfare Japan (1973)	Analytical Manual for endocrine disrupter chemicals by Japan Environmental Agency (1998)	The analytical manual for radioactive materials in foods in emergency by Ministry of Health, Labour and Welfare (2002)

reason for using only mature males was to exclude the effects of pregnancy in case of females and growth, and to be less susceptible to age-related Hg accumulation (Braune *et al.*, 2015).

Concentrations of Hg through the trophic chain

To understand the bioaccumulation process, it is necessary to examine Hg concentrations not only in whales but also in their prey species. Figure 3 shows the relationships between Hg concentrations in common minke, sei and Bryde's whales in the western North Pacific, and those in their main prey species (two zooplanktons and six pelagic fish species) during the period 1995–2007. The Hg concentrations were in the order of:

Common minke whales (0.22±0.07 ppm wet wt.)>sei whales (0.052±0.009)≒Bryde's whales (0.046±0.008).

The Hg concentrations in krill and copepods ranged from <0.001–0.013 and 0.003–0.010 ppm dry wt., respectively. Mercury concentrations in the pelagic fishes were in the order of:

Pacific pomfret (0.232±0.027)>walleye pollock (0.045)=Pacific saury (0.039±0.016)=Japanese anchovy (adult) (0.037±0.025)>Japanese anchovy (larval fish) (0.005±0.003).

It is known that common minke whales selectively feed on relatively higher trophic organisms, mainly pelagic fish such as Pacific saury and Japanese anchovy (Tamura and Fujise, 2002). This is different from sei and Bryde's whales which feed mainly on krill and copepod (Tamura and Fujise, 2002; Tamura *et al.*, 2009). Differences in feeding habitat among whale species and trophic levels of prey would be one of the reasons for higher Hg concentrations in common minke whales than in sei and Bryde's whales. The Hg concentrations in common minke whales from offshore, however, would be affected by changes in the food habitat during the research period.

Temporal trend in Hg concentrations

To examine yearly changes of Hg in common minke, sei and Bryde's whales in the western North Pacific, multiple linear regression analyses were carried out including adjustment for confounders, i.e. sampling years, sampling longitude, sampling latitude, sampling date, body length, blubber thickness and main prey item.

Results indicated that Hg concentrations observed in the muscle of common minke whales off Kushiro and Sanriku (part of sub-area 7) and sub-area 8, and Bryde's whales in sub-areas 8 and 9 were relatively stable in the western North Pacific during the research periods.

On the other hand, the association of Hg concentrations with sampling year was statistically significant in common minke whales from sub-areas 7 and 9, and sei whales from sub-area 9. A slight flexion point of yearly trends of Hg was observed in 2008 for common minke whales from sub-area 9, and in 2012 for sei whales from sub-area 9 (Figure 4). The results of multiple linear regression analyses showed that the main prey items (sub-area 7=walleye pollock, sub-aera 9=Pacific pomfret and Pacific saury) were associated with Hg concentrations in common minke whales. This association was not observed in the case of sei whales. Hg concentrations in sei whales, having sardine and saury in their stomachs, were slightly lower than those in common minke whales from sub-area 9. Therefore, Hg concentrations of sei whales from sub-area 9 may be less affected by Hg in the food items (Figure 4).

Thus, the yearly trend of Hg in sea surface water and the baleen whales from the North Pacific remain stable for the research period. This is also supported by the

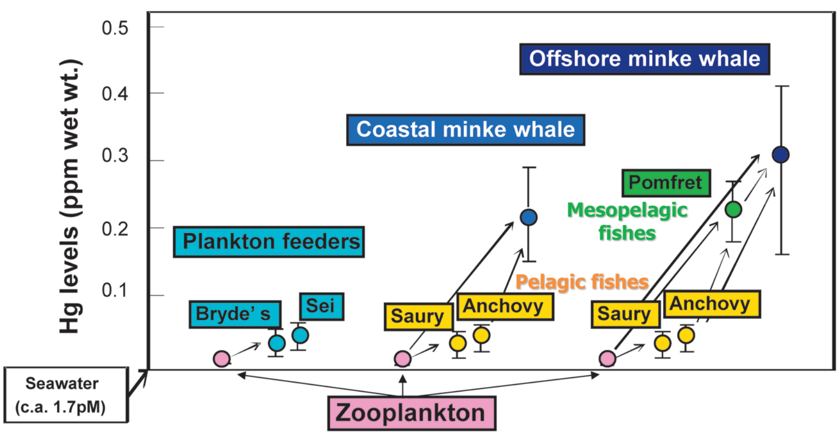

Figure 3. Relationships between Hg concentrations in muscle tissues of common minke, sei and Bryde's whales in the western North Pacific, and those in their prey species.

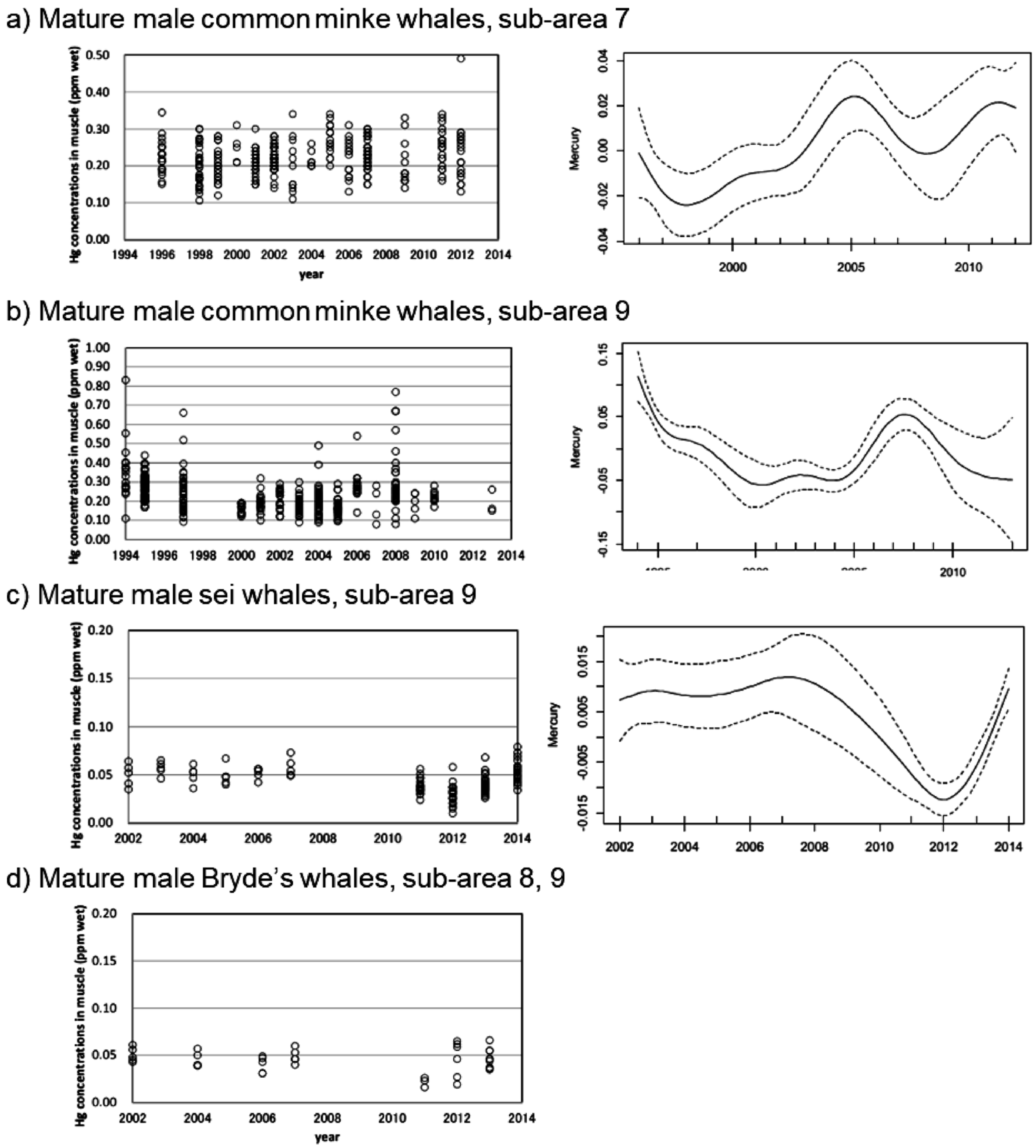

Figure 4. Simple plots (left sides) and smoothing plots using the GAM (right sides) of Hg concentrations in muscle tissues of common minke whales in sub-areas 7 (a: 1996–2012) and 9 (b: 1994–2013), sei whales in sub-area 9 (c: 2002–2014) and Bryde's whales in sub-areas 8 and 9 during research years.

results of monitoring studies on Hg concentrations from 1980s in the nearby waters (Laurier *et al.*, 2004; Sunderland *et al.*, 2009).

POPs

POPs are organic compounds that are resistant to environmental degradation that occur through chemical and biological processes. Therefore, they have the potential to be transported over long distances. The manufacturing, use and import/export of the specified compounds have been strictly restricted since the adoption of the Stockholm Convention on POPs by the United Nations Environment Programme in 2001 (Hagen and Walls, 2005). Among them, polychlorinated biphenyls (PCBs), dichlorodiphenyl-trichloroethanes (DDTs), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB) and chlordanes (CHLs) are commonly called 'legacy POPs' (French *et al.*, 2006). These legacy POPs had been produced and used in large quantities worldwide in the middle of last century. However, they remain a major environmental

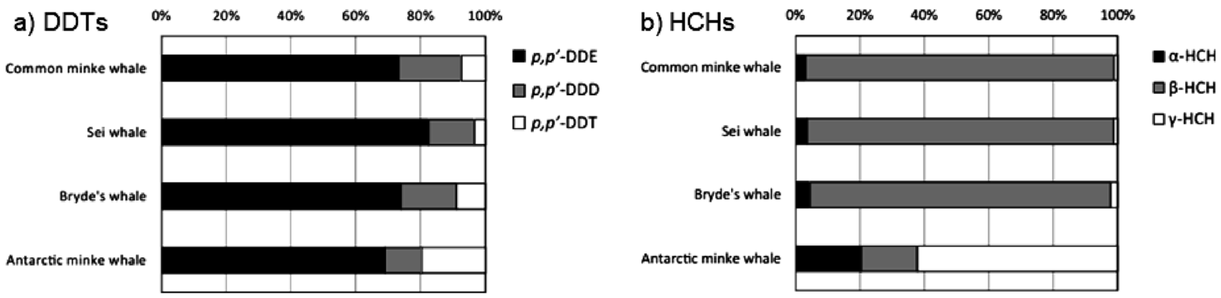

Figure 5. Compositions of a) DDTs and b) HCHs in the blubber of common minke, sei, Bryde′s whales from the western North Pacific and Antarctic minke whales.

Figure 6. Simple plots of PCB concentrations in blubber of common minke whales (mature males) in sub-areas 7 (coastal) and 9 (offshore).

challenge for human health and risk to wildlife due to their toxicity and persistency.

Accumulation features

Yasunaga and Fujise (2020) examined accumulation features of PCB congeners and DDT, HCH, HCB and CHL isomers in the blubber of common minke, sei and Bryde′s whales from the western North Pacific. DDT and HCH, the main components contained in pesticide products released into the environment, were hardly present in the samples. Concentration levels of five mature males of common minke, sei and Bryde′s whales taken by the JARPNII in 2011 were compared with those in Antarctic minke whales. Results showed that, among the legacy POPs, concentration levels of PCBs were the highest in the whales from the western North Pacific, whereas in Antarctic minke whales from the Antarctic Ocean, the levels were lower than those of HCB and DDTs.

The average percentage of *p,p*′-DDT (which is primarily released into the environment) in the three whale species from the western North Pacific was lower than that of Antarctic minke whales (Figure 5). The *p,p*′-DDT has been used to control malaria (De Jager *et al.*, 2006), and is metabolized into *p,p*′-DDE in the animal body and the environment over a long period of time (Okonkwo *et al.*, 2008). The percentage of *β*-HCH accounted for over 90% of total HCHs in common minke, sei and Bryde′s whales from the western North Pacific, whereas the percentage of *γ*-HCH was the dominant isomer of total HCHs in Antarctic minke whales (Figure 5). Technical HCH, called Lindane, had been released into the environment. Almost all of those consisted of *γ*-HCH from the 1950s and 1970s (Walker *et al.*, 1999). However, in Australia, it was used after that period (Tanabe *et al.*, 1982). These results suggest that in the western North Pacific, a great deal of time had passed from the release of DDTs and HCHs into the environment.

Yearly trend

Yasunaga and Fujise (2016a) examined yearly changes of PCBs in the western North Pacific. Multiple linear regression analysis was carried out, including adjustment for confounders, i.e. sampling years, sampling longitude, latitude, sampling date, body length, blubber thickness and main prey species. Results indicated no significant correlations between year and food items in all areas. It was suggested that PCB concentrations in minke whales from the western North Pacific were stable during 2002–2014. Results for common minke whales in coastal and offshore waters are shown in Figure 6.

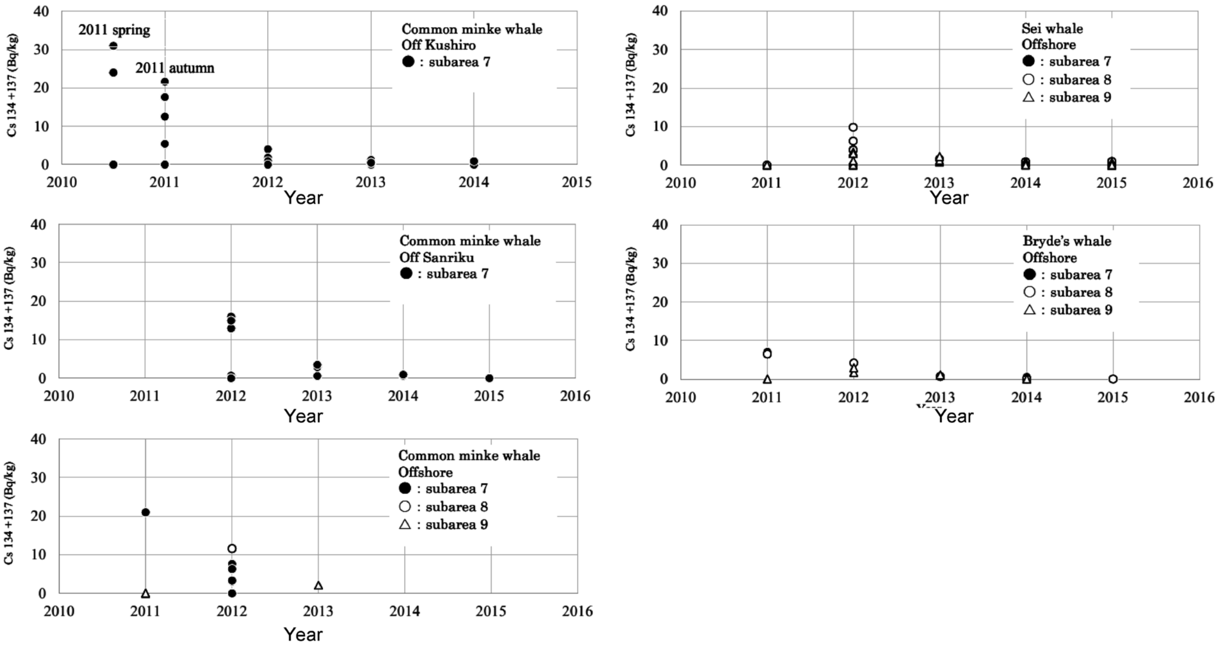

Figure 7. Concentrations of Cs 134+137 in muscle of common minke, sei and Bryde′s whales taken from the western North Pacific from 2011 to 2015.

Radio Isotope

On 11 March 2011, the Great East Japan Earthquake and the tsunami that followed destroyed the nuclear power plant in Fukushima. This accident led to the release of large amounts of radioactive materials into the environment.

To monitor the impact of this incident on large whales in the western North Pacific, Yasunaga and Fujise (2016b) investigated the I131, Cs134 and Cs137 concentrations in muscle samples of 53 common minke, 16 Bryde′s, 32 sei and 3 sperm whales sampled in the period 2011–2015. Results are shown in Figure 7. Iodine131 was not detected in Bryde′s, sei and sperm whales, with the exception of two common minke whales taken from Kushiro in 2012. The ranges of Cs134 + Cs137 concentrations in common minke, sei, Bryde′s and sperm whales were ND (not detected)-31, ND-9.8, ND-7.1 and ND-0.59 Bq/kg wet wt., respectively. The radioisotope concentrations in all four whale species examined have been decreasing since 2011.

CONCLUDING REMARKS

It is important to monitor the concentrations of different chemical pollutants in whale tissues and their environment, and the possible effect of such pollutants on whales. The ICR has contributed to this monitoring by analyzing samples and data collected over a long period in the western North Pacific, not only from whales but also from their prey species and their environment. Biological data available from the whales are important for the interpretation of the concentrations of chemical pollutants found. The results from the direct analyses of samples from biotic and abiotic sources revealed very important information in furthering our understanding of the bioaccumulation process through the marine food chain.

The Minamata Convention on Mercury is a global treaty which aims to protect human health and the environment from the adverse effects of mercury. The monitoring of the marine environment using bioindicators from marine mammals is encouraged by the Convention as one of the recommended actions in the Hg reduction plans (UNEP, 2019). Thus, the findings from this study are expected to contribute directly to such monitoring programs.

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REFERENCES

Borrell, A. and Reijnders, P.J. 1999. Summary of temporal trends in pollutant levels observed in marine mammals. *J. Cetacean Res. Manage.* (special issue) 1: 149–155.

Braune, B., Chételat, J., Amyot, M., Brown, T., Clayden, M., Evans, M., Fisk, A., Gaden, A., Girard, C., Hare, A., Kink, J., Lehnherr, I., Letcher, R., Loseto, L., Macdonald, R., Mann, E., McMeans, B., Muir, D., O′Driscoll, N., Poulain, A., Reimer, K. and Stern, G. 2015. Mercury in the marine environment of the Canadian Arctic: review of recent findings. *Science of the*

Total Environment 509: 67–90.

DeJager, C., Farias, P., Barraza-Villarreal, A., Avila, M.H., Ayotte, P., Dewailly, E., Dombrowski, C., Rousseau, F., Sanchez, V.D. and Bailey, J.L. 2006. Reduced seminal parameters associated with environmental DDT exposure and *p,p*′-DDE concentrations in men in Chiapas, Mexico: A cross-sectional study. *Journal of Andrology* 27 (1): 16–27.

Fitzgerald, W.F., Engstrom, D.R., Mason, R.P. and Nater, E.A. 1998. The case for atmospheric mercury contamination in remote areas. *Environmental Science & Technology* 32 (1): 1–7.

French, T.D., Campbell, L.M., Jackson, D.A., Casselman, J.M., Scheider, W.A. and Hayton, A. 2006. Long-term changes in legacy trace organic contaminants and mercury in Lake Ontario salmon in relation to source controls, trophodynamics, and climatic variability. *Limnol. Oceanogr.* 51 (6): 2794–2807.

Hagen, P.E. and Walls, M.P. 2005. The Stockholm Convention on persistent organic pollutants. *Natural Resources & Environment* 19 (4): 49–52.

International Whaling Commission. 1994. Report of the Working group on North Pacific minke whale management trials. *Rep. int. Whal. Commn* 44: 120–125.

Knauer, G.A. and Martin, J.H. 1972. Mercury in a marine pelagic food chain 1. *Limnol. Oceanogr.* 17 (6): 868–876.

Laurier, F.J.G., Mason, R.P., Gill, G.A. and Whalin, L. 2004. Mercury distributions in the North Pacific Ocean—20 years of observations. *Marine Chemistry* 90 (1-4): 3–19.

Ministry of the Environment Government of Japan. 1998. Analytical Manual for endocrine disrupter chemicals. Tokyo, Japan (in Japanese). http://www.env.go.jp/chemi/end/sympo/manual199810/water.html.

Ministry of Health and Welfare Japan. 1972. Provisional regulation levels of Hg in fisheries. Tokyo, Japan (in Japanese). https://www.mhlw.go.jp/web/t_doc?dataId=00ta5730&dataType=1&pageNo=1.

Ministry of Welfare Japan. 1973. The public analytical method of Japan. Tokyo, Japan (in Japanese). https://www.mhlw.go.jp/web/t_doc?dataId=00ta5729&dataType=1&pageNo=1.

Ministry of Health, Labour and Welfare. 2002. The analytical manual for radioactive materials in foods in the emergency. http://www.mhlw.go.jp/stf/houdou/2r9852000001558e-img/2r98520000015cfn.pdf.

Okonkwo, J.O., Mutshatshi, T.N., Botha, B. and Agyei, N. 2008. DDT, DDE and DDD in human milk from South Africa. *Bulletin of Environmental Contamination and Toxicology* 81: 348–354.

Sanpera, C., Capelli, R., Minganti, V. and Jover, L. 1993. Total and organic mercury in North Atlantic fin whales: distribution pattern and biological related changes. *Marine Pollution Bulletin* 26 (3): 135–139.

Sunderland, E.M., Krabbenhoft, D.P., Moreau, J.W., Strode, S.A. and Landing, W.M. 2009. Mercury sources, distribution, and bioavailability in the North Pacific Ocean: Insights from data and models. *Global Biogeochemical Cycles* 23 (2).

Tamura, T. and Fujise, Y. 2002. Geographical and seasonal changes of the prey species of minke whale in the Northwestern Pacific. *ICES Journal of Marine Science* 59 (3): 516–528.

Tamura, T., Konishi, K., Isoda, T., Okamoto, R. and Bando, T. 2009. Prey consumption and feeding habits of common minke, sei and Bryde′s whales in the western North Pacific. Paper SC/J09/JR16 presented to the JARPNII Review Workshop, January 2009 (unpublished). 36 pp. [Available from the IWC Secretariat].

Tanabe, S., Tatsukawa, R., Kawano, M. and Hidaka, H. 1982. Global distribution and atmospheric transport of chlorinated hydrocarbons: HCH (BHC) isomers and DDT compounds in the western Pacific, eastern Indian and Antarctic Oceans. *Journal of the Oceanographical Society of Japan* 38: 137–148.

UNEP. 2019. Minamata Convention on mercury - Text and Annexes. https://mercuryconvention.org/en/resources/minamata-convention-mercury-text-and-annexes.

Walker, K., Vallero, D.A. and Lewis, R.G. 1999. Factors influencing the distribution of lindane and other hexachlorocyclohexanes in the environment. *Environmental Science & Technology* 33 (24): 4373–4378.

Yasunaga, G., Hakamada, T. and Fujise, Y. 2016. Update of the Analyses on Temporal trend of Total Hg levels in three baleen whale species based on JARPN and JARPNII data for the period 1994–2014. Paper SC/66b/E/08 presented to the IWC Scientific Committee, June 2016 (unpublished). 18 pp. [Available from the IWC Secretariat].

Yasunaga, G. and Fujise, Y. 2016a. Temporal trend of PCB levels in common minke whales from the western North Pacific for the period 2002–2014. Paper SC/F16/JR31 presented to the JARPNII Special Permit Expert Panel Review Workshop, February 2016 (unpublished). 9 pp. [Available from the IWC Secretariat].

Yasunaga, G. and Fujise, Y. 2016b. Status of I131, Cs134 and Cs137 in baleen and sperm whales from the western North Pacific during 2011-2015. Paper SC/F16/JR35 presented to the JARPNII Special Permit Expert Panel Review Workshop, February 2016 (unpublished). 6 pp. [Available from the IWC Secretariat].

Yasunaga, G. and Fujise, Y. 2020. Concentration of persistent organic pollutants (POPs) in three species of baleen whales in the western North Pacific. *Cetacean Population Studies* 2: 39–53.