SC/66b/E/08

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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

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ABSTRACT

This paper is the revised version of SC/J16/JR30, which described the results of multiple linear regression analyses of total mercury (Hg) in muscle of baleen whales based on the JARPN and JARPNII, presented to the Expert Workshop to review the results from the JARPNII Programme. To examine yearly changes of total Hg in the western North Pacific, total Hg concentrations in muscle from common minke whale (Balaenoptera acutorostrata), sei (B. borealis) and Bryde's (B. edeni) whales, were measured. Averages and standard deviations of total Hg concentrations in samples of mature males of O stock of common minke whales from sub-area 7, off Kushiro, off Sanriku, subareas 8 and 9 were 0.22±0.05, 0.22±0.05, 0.22±0.06, 0.22±0.06 and 0.24±0.10, ppm wet wt., respectively. Those of sei whales from sub-area 9 and Bryde's whales from sub-areas 8 and 9 were 0.044±0.013 and 0.044±0.013 ppm wet wt., respectively. These levels may not have any adverse effects on whale health. Multiple linear regression analysis was carried out and included adjustment for confounders, sampling years, sampling longitude, sampling latitude, sampling date, body length, blubber thickness and main prey item. Total Hg concentrations in the common minke whales from sub-area 7 were significantly associated with deficits in sampling year and main prey item, and those from sub-area 9 were significantly associated with deficits in sampling year, latitude and main prey item. Main prey items had an effect on total Hg concentrations in the common minke whales from sub-areas 7 and 9. These findings suggest that yearly changes of total Hg in common minke whales from the western North Pacific could be affected by changes of their prey items. The results of this revised version confirmed the main conclusion in SC/F16/JR30.

KEYWORDS: COMMON MINKE WHALE; SEI WHALE; BRYDE'S WHALE; NORTH PACIFIC; TOTAL MERCURY; MONITORING

INTRODUCTION

Pollutants accumulate through the food chain in the marine ecosystem. Because cetaceans are located at the top of the food chain they are frequently used for monitoring the pollutants in the marine environment (Sanpera *et al.*, 1993; Borrell and Reijnders, 1999). Further, cetaceans are mobile and long-lived animals, and these characteristics mean that pollutants can be monitored in wide areas and integrated in some way over time.

In JARPNII, surveys were designed so that quantitative behavior and fate of the pollutants in the marine environment could be monitored. The present study examines three concentrations and temporal changes of total Hg in three top predator species (baleen whales) in the western North Pacific. The analysis of this pollutant in both predators and prey species is important as the concentration of total Hg in predators reflect in part their feeding habits. To understand the pattern of accumulation of total Hg in whales it is important to consider some biological information such as sex and sexual maturity. Such information is considered in this study.

In the 2009 review workshop, Yasunaga and Fujise (2009a) presented a study on yearly changes of mercury concentrations in muscle samples of common minke (*Balaenoptera acutorostrata*), sei (*B. borealis*) and Bryde's (*B. edeni*) whales and their prey items from the western North Pacific using JARPN and JARPNII samples during 1994-2007. Yearly changes of total Hg concentrations in zooplankton and pelagic fishes were not observed in the period 1994-2007. Apart from minke whales from sub-area 9, significant yearly changes of concentrations in whales were not observed. For minke whales in sub-area 9, total Hg concentrations decreased from 1994 to 1999 but increased from 2000 to 2007. Results of a multiple linear regression analysis suggested that changes of total Hg concentrations in sub-area 9 reflect changes in food habits of minke whale rather than changes in accumulation concentrations in the environment.

This study examines concentrations and temporal changes of total Hg concentrations in mature males of three baleen whale species. Body length, blubber thickness, stomach content and sexual maturity information obtained during JARPN and JARPNII surveys is considered to assist in the interpretation of factors affecting total Hg concentrations.

This paper is a revised version of SC/F16/JR30 to cover the recommendations of the panel of the Expert Workshop held at Tokyo from 22-26 of February 2016 to review the results from the JARPNII Programme. The panel recommended that the statistical analyses should be improved and the pollutant concentrations found should be evaluated in comparison with data from previous studies conducted in comparable species and available in the literature.

MATERIALS AND METHODS

Samples

Muscle tissues from the minke whales (Okhotsk Sea-West Pacific stock) from sub-area 7, off Kushiro, off Sanriku, subareas 8 and 9, sei whales from sub-area 9 and Bryde's whales from sub-areas 8 and 9 were collected by JARPNII researchers in the surveys conducted in the period from 1994 to 2014 (Fig. 1). Biological data such as body length and blubber thickness and tissue samples were collected on board the *Nisshin Maru* (Tables 1-3). The muscle tissues were excised from the medial region part of the body, frozen and shipped to the laboratory and stored at -20°C until chemical analysis. The forestomach contents have proved sufficient for determination of the common minke whale diet in the Northeast Atlantic (Lindstrøm *et al.*, 1997). The prey composition between forestomach and fundus were very similar in this study. Therefore, this study was based on contents from forestomach.

Tables 1-3 show sample sizes n and maturity stage as well as means of body length, blubber thickness and muscular Hg by year, with standard deviations (SD). Males of minke, Bryde's and sei whales were defined as sexually mature by testis weight (larger side) of more than 290g, 560g and 1,090g, respectively (Bando *et al.*, unpublished data). Only minke whales from the Okhotsk Sea-West Pacific stock (O-stock) as identified by the microsatellite analysis (Kanda *et al.*, 2009), were used.

Laboratory analysis

The muscle samples were stripped externally to avoid contamination. For total Hg analysis, the samples were set on a ceramic boat and were subjected to cold-vapor atomic absorption spectrometry with heat-vaporization, gold amalgamation method (Nippon Instruments Co., MA-3000). Triplicate analysis was performed on each sample, to increase the accuracy of the determination. Accuracy and precision of the methods were confirmed using standard materials, DORM-3 (NRCC, muscle of dogfish).

Total Hg concentrations in prey items in SC/J09/JR23 of the 2007 JARPNII review (Yasunaga and Fujise, 2009a), were used because most of the samples for this study until 2011 were lost after the 2011 earthquake and tsunami (see IWC, 2012).

Statistical analysis

The yearly changes of total Hg concentrations in muscle of whales were assessed by multiple linear regression in the context of other factors (R Development Core Team, 2006). The following independent variables: "Year", "Date", "Latitude", "Longitude", "Body length", "Blubber thickness" and "Main prey item" were allowed, and all parameters except for main prey item were logarithmic transformed. Categorical parameters of main prey items used in the analyses were the following: sub-area 7 of minke whale (Japanese anchovy: Engraulis japonicus, Euphausiids, Japanese flying squid: Todarodes pacificus, mackerel: Scomber japonicus, Japanese sardine: Sardinops melanostictus, Pacific saury: Cololabis saira and Walleye Pollock: Theragra chalcogramma); off Kushiro and off Sanriku of common minke whale (None); sub-area 8 of minke whale (anchovy, Copepods, Euphausiids, Japanese flying squid, mackerel and Pacific saury); sub-area 9 of minke whale (anchovy, Atka mackerel: Pleurogrammus monopterygius, Copepods, Euphausiids, mackerel, armhook squid: Berryteuthis anonychus, oceanic lightfish: Vinciguerria nimbaria, Pacific pomfret: Brama japonica, Salomonids, Pacific saury); sub-area 9 of sei whale (Japanese anchovy, Copepods, Euphausiids, mackerel, armhook squid, Japanese sardine and Pacific saury); sub-areas 8 and 9 of Bryde's whale (Japanese anchovy, Euphausiids and mackerel). Anchovy is the baseline prey item in sub-areas 7-9 so the effects of anchovy being the main prey item are included in the model intercepts. Data sets were excluded from analyses in case of empty stomach and damage by harpoon. Also, multiple robust linear regression analyses were conducted to examine the effect of outliers in the same way. Furthermore, a generalized additive model (GAM) was used to examine flexion point in the yearly changes of total Hg concentrations in muscle of whales. A p value of less than 0.05 was considered to indicate statistical significance in all tests. These statistical analyses were performed using the free software R, version 3.3.0 (R Development Core Team, 2006).

RESULTS

Tables 1-3 show the total Hg concentrations in muscle of minke, sei and Bryde's whales, respectively. Averages and standard deviations of total Hg concentrations in muscle samples of mature males of O stock minke whales from subarea 7, off Kushiro, off Sanriku, and sub-areas 8 and 9 were 0.22±0.05, 0.22±0.05, 0.22±0.06, 0.22±0.06 and 0.24±0.10, ppm wet wt., respectively. Those of sei whales from sub-area 9 and Bryde's whales from sub-areas 8 and 9 were 0.044±0.013 and 0.044±0.013 ppm wet wt., respectively. To examine yearly changes of total Hg concentrations in minke, sei and Bryde's whales and their confounders, multiple linear regression analyses were conducted, and the results are given in Tables 4-10. Also, to examine those except for effect of outliers, multiple robust linear regression analyses were conducted, and the results are given in Tables 11-17. Based on the F test results in these tables, the overall regression was statistically significant except in Table 6 and Table 10. In minke whales from sub-areas 7 and 9, and sei whales from sub-area 9, total Hg concentrations were significantly associated with sampling year (Tables 4, 8 and 9). Similarly, those were significantly associated with sampling year by multiple robust linear regression analyses (Tables 11, 15 and 16). Also, simple plots and, when possible, smoothing plots using the GAM of total Hg concentrations against their sampling year are shown for common minke (Figs. 2a-2e for different sub-areas), sei (Fig. 2f) and Bryde's (Fig. 2g) whales during the 1994-2014 period except that a smoothing plot is not shown in Fig. 2g. The Bryde's whale samples were too few and too far separated in time and space to support a GAM analysis. Slight flexion points of yearly trends of muscular total Hg were observed in 2008 of minke whales from sub-area 9 and in 2012 of sei whales from sub-area 9.

DISCUSSION

In sub-area 7, total Hg concentrations in muscle of minke whales (period 1996-2012) were significantly associated with Intercept (-), Year (+) and Main prey items (Walleye Pollock) (Table 4) by multiple linear regression, and also were significantly associated with Intercept (-), Year (+), Date (+), Main prey items (Mackerel, Sardine, Saury and Walleye Pollock) (Table 11). Total Hg concentrations in muscle of minke whale observed from each main prey item in stomach and total Hg concentrations in whole body of the prey items from the same areas are shown in Table 18. Total Hg concentrations in minke whales having mackerel in the stomach were the highest and those having sardine in stomach were the lowest among minke whales from sub-area 7, whereas those for other prey items had similar concentrations. The number of minke whales having mackerel and sardine in their stomach were only one and two whales, respectively. These results indicate that total Hg concentrations of minke whales from sub-area 7 may be less affected by total Hg in the food items.

In sub-area 9, total Hg concentrations in muscle of minke whales (period 1996-2012) were significantly associated with Intercept (+), Year (-), Latitude (+), Date (-) and Main prey items (Pacific pomfret and Saury) (Table 8) by multiple linear regression, and also were significantly associated with Intercept (+), Year (-),Latitude (+), Date (-) and Main prey items (Atka Mackerel, Oceanic Lightfish, Pacific pomfret and Saury) (Table 11). Total Hg concentrations in muscle of minke whale for each main prey item in the stomach and total Hg concentrations in the prey items from sub-area 9 are shown in Table 12. Total Hg concentrations in minke whales having Pacific pomfret in stomach were the highest among minke whales from sub-area 9, and total Hg concentrations in Pacific pomfret were one or two orders of magnitude higher than those in the other prey items. Also total Hg concentrations in the zooplankton were one or two orders of magnitude lower than those in the other prey items. Furthermore, yearly changes were observed in food items of the minke whales in the same period (Konishi *et al.*, 2016). A peak of total Hg levels in the GAM plots were observed in 2008 (Fig. 2e), and Pacific pomfret were observed in the stomach contents of minke whales from sub-area 9 in the 2008 JARPNII survey (Tamura *et al.*, 2009). These results indicate that changes of total Hg concentrations in sub-area 9 reflect changes in food habits of minke whale rather than changes of background levels of total Hg in the marine environment.

In sub-area 9, total Hg concentrations in sei whales (period 2002-2014) were significantly associated with Intercept (+), Year (-) and Body length (+) by both multiple linear regression (Table 9) and multiple robust linear regression (Table 16), whereas those in 2012 were lower than the others in the GAM plots (Fig. 2f). Total Hg concentrations in muscle of each sei whale having the same main prey items in the stomach and total Hg concentrations in the prey items from the same areas are compared in Table 20. Total Hg concentrations in minke whales having anchovy and sardine in the stomach were slightly higher than the other two whale species from sub-area 9. These results indicate that total Hg concentrations of sei whales from sub-area 9 may be less affected by total Hg in the food items.

Total Hg concentrations observed in muscle of minke whales in off Kushiro and off Sanriku, 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, yearly changes of total Hg in muscle of minke whales from sub-areas 7 and 9, and sei whales from sub-area 9 were observed reflecting change of their food items. Total Hg concentrations in surface water of the North Pacific have not changed since the 1980's (Laurier *et al.*, 2004; Sunderland *et al.*, 2010). Consequently, yearly trend of total Hg in sea surface water and the baleen whales from the North Pacific remained stable for the research period, while anthropogenic Hg emission in Asia have risen steadily over the past several decades (UNEP, 2013).

In this study, total Hg levels in baleen whales in the western North Pacific during 1994-2014 were similar. Yasunaga and Fujise (2009b) reported that total Hg concentrations in kidney and liver of common minke whales were 0.026-1.3 ppm wet wt. and 0.035-0.80 ppm wet wt., respectively, and those of Bryde's whales were 0.014-0.60 ppm wet wt. and 0.020-0.49 ppm wet wt., respectively. Thompson (1996) reported that the threshold for lethal or at least harmful effects from total Hg concentrations in kidney and liver are approximately 30mg/kg wet wt. in terrestrial wildlife mammals. Concentrations of total Hg in liver and kidney of common minke and Bryde's and sperm whales were much lower than this mercury threshold value. Furthermore, Augier *et al.* (1993) reported that striped dolphin containing high Hg (max. 2,272 ppm dry wt.) and Se (max. 1,320 ppm dry wt.) concentrations in livers did not show any symptoms of mercury or selenium poisoning because the both elements form inert material in some organs and tissues. These mercury-selenium antagonism in kidney and liver of common minke and Bryde's whales were observed (Yasunaga and Fujise, 2009b). Therefore, Hg levels in whales in this study may not have any adverse effect on whale health.

ACKNOWLEDGEMENTS

We thank the panel of the Expert Workshop to review the results from the JARPNII Programme for valuable comments on the paper. Dr. Zeh provided constructive comments and suggestions.

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Table 1. Biological data and total mercury concentrations in muscle of common minke whales (males) from western North Pacific during 1994 and 2014

subarea	year	maturity	п		body length	blubber thickness muscula		muscular Hg		
					(m)		(cm)		(ppm wet wt.)	
7	1996	mature	18	($7.36\ \pm\ 0.28$) ($3.1~\pm~0.4$) ($0.23\ \pm\ 0.05$)
	1998	mature	35	($7.50\ \pm\ 0.27$) ($2.6\ \pm\ 0.4$) ($0.20\ \pm\ 0.05$)
	1999	mature	31	($7.41\ \pm\ 0.29$) ($3.6\ \pm\ 0.8$) ($0.20\ \pm\ 0.04$)
	2000	mature	6	($7.30\ \pm\ 0.25$) ($3.6~\pm~1.0$) ($0.24\ \pm\ 0.04$)
	2001	mature	27	($7.45\ \pm\ 0.31$) ($2.4\ \pm\ 0.5$) ($0.21\ \pm\ 0.04$)
	2002	mature	33	($7.52\ \pm\ 0.27$) ($3.7\ \pm\ 0.8$) ($0.22\ \pm\ 0.03$)
	2003	mature	11	($7.42~\pm~0.40$) ($2.4~\pm~0.5$) ($0.20~\pm~0.07$)
	2004	mature	6	($7.56\ \pm\ 0.21$) ($4.1~\pm~0.6$) ($0.22~\pm~0.02$)
	2005	mature	17	($7.51\ \pm\ 0.23$) ($3.3\ \pm\ 0.9$) ($0.27\ \pm\ 0.04$)
	2006	mature	16	($7.39\ \pm\ 0.39$) ($2.4~\pm~0.7$) ($0.22~\pm~0.05$)
	2007	mature	41	($7.47\ \pm\ 0.26$) ($3.0~\pm~0.7$) ($0.23~\pm~0.04$)
	2009	mature	11	($7.71~\pm~0.26$) ($3.2~\pm~0.7$) ($0.21\ \pm\ 0.06$)
	2011	mature	23	($7.47\ \pm\ 0.34$) ($3.5\ \pm\ 0.6$) ($0.25~\pm~0.05$)
	2012	mature	19	($7.37\ \pm\ 0.34$) ($2.6~\pm~0.4$) ($0.24~\pm~0.08$)
	total		294	($7.46\ \pm\ 0.30$) ($3.1~\pm~0.8$) ($0.22~\pm~0.05$)
Kushiro	2002	mature	14	($7.22\ \pm\ 0.34$) ($4.3~\pm~1.0$) ($0.21~\pm~0.06$)
	2004	mature	23	($7.43\ \pm\ 0.24$) ($4.2~\pm~0.6$) ($0.20~\pm~0.03$)
	2005	mature	25	($7.44~\pm~0.32$) ($3.5~\pm~0.6$) ($0.25~\pm~0.05$)
	2006	mature	10	($7.52\ \pm\ 0.29$) ($4.4~\pm~0.8$) ($0.22~\pm~0.03$)
	2007	mature	14	($7.33\ \pm\ 0.59$) ($4.1~\pm~0.8$) ($0.23~\pm~0.06$)
	2008	mature	5	($7.26~\pm~0.21$) ($4.3~\pm~1.0$) ($0.25~\pm~0.01$)
	2009	mature	6	($7.54\ \pm\ 0.37$) ($4.3~\pm~0.4$) ($0.21~\pm~0.07$)
	2010	mature	3	($7.49\ \pm\ 0.21$) ($3.8~\pm~0.4$) ($0.23~\pm~0.02$)
	2011spring	mature	4	($7.40~\pm~0.41$) ($3.2~\pm~0.6$) ($0.24~\pm~0.03$)
	2011autumn	mature	10	($7.26\ \pm\ 0.37$) ($4.4~\pm~1.0$) ($0.23~\pm~0.02$)
	2012	mature	5	($7.48~\pm~0.21$) ($5.0~\pm~0.7$) ($0.21~\pm~0.06$)
	2013	mature	23	($7.33\ \pm\ 0.35$) ($5.0~\pm~0.9$) ($0.19\ \pm\ 0.04$)
	2014	mature	10	($7.33\ \pm\ 0.28$) ($5.0~\pm~0.8$) ($0.21~\pm~0.09$)
	total		152	($7.38~\pm~0.35$) ($4.3~\pm~0.9$) ($0.22~\pm~0.05$)
Sanriku	2003	mature	8	(7.10 ± 0.35) ($2.9~\pm~0.5$) ($0.18~\pm~0.04$)
	2005	mature	3	(7.27 ± 0.13) (2.5 ± 0.3) (0.29 ± 0.02)
	2006	mature	6	($7.24\ \pm\ 0.32$) ($2.8~\pm~0.2$) ($0.19~\pm~0.05$)
	2007	mature	10	(7.50 ± 0.30) (2.5 ± 0.4) (0.25 ± 0.08)
	2008	mature	3	(7.49 ± 0.32) (3.2 ± 0.3) (0.19 ± 0.03)
	2009	mature	1	($6.85 \pm$) ($2.8~\pm$) ($0.21 \pm$)
	2010	mature	5	(7.32 ± 0.24) (3.1 ± 0.6) ($0.20~\pm~0.05$)
	2012	mature	2	($7.31 \pm$) ($3.4~\pm$) ($0.18~\pm$)
	2013	mature	2	($7.52 \pm$) ($3.5~\pm$) (0.30 \pm)
	2014	mature	2	($7.38~\pm~0.39$) ($2.7~\pm~0.3$) ($0.31\ \pm\ 0.04$)
	total		42	($7.32~\pm~0.32$) ($2.8~\pm~0.5$) ($0.22~\pm~0.06$)

subarea	year	maturity	п		body length	b	lubber thickne	ess	muscular Hg	
					(m)		(cm)		(ppm wet wt.)	
8	1996	mature	11	($7.44\ \pm\ 0.33$) ($2.6\ \pm\ 0.5$) ($0.25~\pm~0.04$)
	1997	mature	26	($7.44\ \pm\ 0.36$) ($3.5\ \pm\ 0.6$) ($0.27~\pm~0.06$)
	1998	mature	28	($7.54\ \pm\ 0.25$) ($2.6\ \pm\ 0.4$) ($0.23~\pm~0.05$)
	2001	mature	13	($7.78\ \pm\ 0.34$) ($2.3\ \pm\ 0.4$) ($0.21\ \pm\ 0.05$)
	2002	mature	5	($7.71~\pm~0.21$) ($3.2~\pm~0.6$) ($0.20~\pm~0.11$)
	2003	mature	27	($7.50\ \pm\ 0.24$) ($2.4\ \pm\ 0.4$) ($0.18~\pm~0.06$)
	2005	mature	2	($7.54~\pm$) ($2.2~\pm$) ($0.16~\pm$)
	2006	mature	24	($7.51\ \pm\ 0.26$) ($2.9\ \pm\ 0.6$) ($0.21\ \pm\ 0.05$)
	2007	mature	10	($7.55~\pm 0.26$) ($2.3\ \pm\ 0.4$) ($0.22~\pm~0.06$)
	2008	mature	3	($7.52\ \pm\ 0.05$) ($2.7~\pm~0.4$) ($0.23\ \pm\ 0.02$)
	2009	mature	6	($7.23\ \pm\ 0.34$) ($3.2~\pm~0.6$) ($0.20~\pm~0.06$)
	total		155	($7.52~\pm~0.30$) ($2.8~\pm~0.6$) ($0.22~\pm~0.06$)
9	1994	mature	16	($7.42~\pm~0.27$) ($3.5\ \pm\ 0.7$) ($0.36\ \pm\ 0.16$)
	1995	mature	68	($7.45\ \pm\ 0.31$) ($3.1~\pm~0.5$) ($0.27~\pm~0.06$)
	1997	mature	39	($7.41\ \pm\ 0.32$) ($3.1~\pm~0.5$) ($0.25~\pm~0.11$)
	2000	mature	12	($7.51~\pm~0.23$) ($2.6~\pm~0.5$) ($0.16\ \pm\ 0.03$)
	2001	mature	19	($7.69\ \pm\ 0.29$) ($2.5~\pm 0.4$) ($0.19\ \pm\ 0.05$)
	2002	mature	21	($7.55~\pm 0.21$) ($2.9\ \pm\ 0.6$) ($0.20~\pm~0.05$)
	2003	mature	28	($7.50\ \pm\ 0.32$) ($2.9\ \pm\ 0.6$) ($0.18~\pm~0.05$)
	2004	mature	50	($7.47~\pm~0.21$) ($3.9\ \pm\ 0.7$) ($0.20~\pm~0.07$)
	2005	mature	25	($7.49\ \pm\ 0.29$) ($3.5~\pm 0.6$) ($0.17~\pm~0.05$)
	2006	mature	16	($7.56~\pm~0.30$) ($3.4~\pm~0.6$) ($0.28~\pm~0.08$)
	2007	mature	4	($7.56~\pm~0.20$) ($2.2~\pm~0.3$) ($0.18~\pm~0.09$)
	2008	mature	36	($7.45~\pm~0.26$) ($3.1~\pm~0.7$) ($0.30\ \pm\ 0.15$)
	2009	mature	5	($7.41~\pm~0.41$) ($3.0~\pm~0.8$) ($0.19\ \pm\ 0.06$)
	2010	mature	9	($7.66~\pm~0.23$) ($3.2~\pm~0.5$) ($0.22~\pm~0.03$)
	2013	mature	3	($7.59\ \pm\ 0.22$) ($4.0~\pm~1.3$) ($0.19\ \pm\ 0.06$)
	total		351	($7.49~\pm~0.28$) ($3.2~\pm 0.7$) ($0.24~\pm~0.10$)

subarea	year	matuarity	п		body length		blubber thickne	ess	musclar Hg	
	-	-			(m)		(cm)		(ppm wet wt.)	
9	2002	mature	5	(13.60 ± 0.16) ($4.7~\pm~0.5$) (0.050 ± 0.012)
	2003	mature	5	($13.80\ \pm\ 0.43$) ($4.7~\pm~0.7$) ($0.057\ \pm\ 0.007$)
	2004	mature	5	(13.61 ± 0.04) ($4.1~\pm~0.6$) ($0.050\ \pm\ 0.009$)
	2005	mature	5	($13.54\ \pm\ 0.38$) ($4.8~\pm~0.6$) ($0.049\ \pm\ 0.011$)
	2006	mature	5	(13.84 ± 0.51) ($4.9~\pm~0.6$) ($0.052\ \pm\ 0.006$)
	2007	mature	5	($13.92\ \pm\ 0.08$) ($5.8~\pm~0.7$) ($0.058\ \pm\ 0.010$)
	2011	mature	15	($13.82\ \pm\ 0.42$) ($4.8~\pm~0.8$) ($0.039\ \pm\ 0.009$)
	2012	mature	21	($13.74\ \pm\ 0.45$) ($5.0\ \pm\ 0.5$) ($0.030\ \pm\ 0.011$)
	2013	mature	26	(13.76 ± 0.50) ($5.6~\pm~0.8$) ($0.040\ \pm\ 0.009$)
	2014	mature	21	($13.80\ \pm\ 0.36$) ($4.8~\pm~0.5$) ($0.053\ \pm\ 0.009$)
total	2002-2014	mature	113	(13.76 ± 0.41) ($5.0~\pm~0.8$) (0.044 ± 0.013)

Table 2. Biological data and mercury concentrations in muscle of sei whales (males) from western North Pacific during 2002 and 2014

Table 3. Biological data and mercury concentrations in muscle of Bryde's whales (males) from western North Pacific during 2002 and 2014

subarea	year	matuarity	п		body length	1	blubber thickne	ss	musclar Hg	
					(m)		(cm)		(ppm wet wt.)	
8,9	2002	mature	5	(12.67 ± 0.43) ($4.5~\pm~0.6$) (0.051 ± 0.008)
	2004	mature	5	($12.59\ \pm\ 0.36$) ($4.4~\pm~0.7$) ($0.045\ \pm\ 0.008$)
	2006	mature	5	($12.36\ \pm\ 0.47$) ($4.5\ \pm\ 0.5$) ($0.040\ \pm\ 0.011$)
	2007	mature	5	($12.63\ \pm\ 0.35$) ($3.9\ \pm\ 0.7$) ($0.049\ \pm\ 0.008$)
	2011	mature	3	(12.31 ± 0.27) ($3.6~\pm~0.5$) ($0.022\ \pm\ 0.005$)
	2012	mature	6	($12.85\ \pm\ 0.48$) ($4.8~\pm~1.0$) ($0.046\ \pm\ 0.019$)
	2013	mature	9	($12.79\ \pm\ 0.38$) ($4.7~\pm~0.8$) ($0.047\ \pm\ 0.010$)
total	2002-2013	mature	38	(12.64 ± 0.41) ($4.4~\pm~0.8$) (0.044 ± 0.013)

Table 4. Results of multiple linear regression analysis with "total Hg levels in muscle of common minke whales from subarea 7" as the dependent variable.

a) Analysis of Variance Table

Source	Sum of Squares	DF	Mean Square	Other Statistics
Intercept B ₀	677.02	1	677.02	R^2 =0.123, Adjusted R^2 =0.0849
Regression Bo	1.91	12	0.160	F=3.212, <i>p</i> <0.05
Residual	13.395	274	0.049	SE=0.221
Total	692.301	287		
b) Variables				
Model	В	SE	Т	p value
Intercept	-196.55	52.934	-3.71	<0.05
Year	25.722	6.801	3.78	<0.05
Body length	-0.092	0.220	-0.42	0.676
Blubber thickness	0.072	0.061	1.19	0.236
Latitude	-0.078	0.529	-0.15	0.884
Longitude	-0.048	0.427	-0.11	0.911
Date	0.037	0.042	0.88	0.378
MainPrey_Euphausiids	0.073	0.045	1.62	0.106
MainPrey_JFSquid	0.067	0.095	0.70	0.485
MainPrey_Mackerel	0.374	0.223	1.68	0.094
MainPrey_Sardine	-0.285	0.159	-1.79	0.074
MainPrey_Saury	0.090	0.052	1.74	0.084
MainPrey_WalleyePollock	0.122	0.053	2.30	<0.05

Table 5. Results of multiple linear regression analysis with "total Hg levels in muscle of common minke whales from off Kushiro" as the dependent variable.

a) Analysis of Variance Tabl	е			
Source	Sum of Squares	DF	Mean Square	Other Statistics
Intercept B ₀	369.84	1	369.84	R^2 =0.115, Adjusted R^2 =0.078
Regression B ₀	1.17	6	0.195	F=3.139, <i>p</i> <0.05
Residual	9.466	145	0.065	SE=0.256
Total	380.536	152		
b) Variables				
Model	В	SE	Т	p value
Intercept	76.2	114.4	0.667	0.506
Year	-3.18	11.87	-0.27	0.789
Body length	0.845	0.430	1.97	0.051
Blubber thickness	-0.345	0.103	-3.37	<i>p</i> <0.05
Latitude	0.526	8.182	0.06	0.949
Longitude	-11.463	14.932	-0.77	0.444
Date	0.105	0.222	0.47	0.637

Table 6. Results of multiple linear regression analysis with "total Hg levels in muscle of common minke whales from off Sanriku" as the dependent variable.

Source	Sum of Squares	DF	Mean Square	Other Statistics
Intercept B ₀	100.93	1	100.93	R^2 =0.2518, Adjusted R^2 =0.1235
Regression Bo	0.88	6	0.146	F=1.963, p=0.0979
Residual	2.605	35	0.074	SE=0.273
Total	104.411	42		
b) Variables				
Model	В	SE	Т	p value
Intercept	-102.5	293.1	-0.35	0.729
Year	37.21	48.90	0.76	0.452
Body length	-0.031	0.028	-1.08	0.288
Blubber thickness	-0.480	0.289	-1.66	0.106
Latitude	21.051	19.404	1.09	0.285
Longitude	-52.409	58.980	-0.89	0.380
Date	0.965	0.637	1.52	0.138

Table 7. Results of multiple linear regression analysis with "total Hg levels in muscle of common minke whales from subarea 8" as the dependent variable.

a) Analysis of Variance Table				
Source	Sum of Squares	DF	Mean Square	Other Statistics
Intercept B ₀	366.96	1	366.96	R^2 =0.235, Adjusted R^2 =0.175
Regression B ₀	3.81	11	0.346	F=3.909, <i>p</i> < 0.05
Residual	12.408	140	0.089	SE=0.298
Total	383.180	152		

b) Variables				
Model	В	SE	Т	p value
Intercept	205.5	107.1	1.918	0.057
Year	-23.42	14.79	-1.58	0.115
Body length	0.911	0.623	1.46	0.146
Blubber thickness	0.358	0.122	2.94	<i>p</i> <0.05
Latitude	0.253	1.389	0.18	0.856
Longitude	-6.367	2.648	-2.41	<i>p</i> <0.05
Date	-0.066	0.523	-0.13	0.900
MainPrey_Copepods	0.008	0.308	0.03	0.980
MainPrey_Euphausiids	-0.062	0.140	-0.45	0.656
MainPrey_JFSquid	-0.031	0.311	-0.10	0.921
MainPrey_Mackerel	0.347	0.220	1.58	0.117
MainPrey_Saury	0.186	0.069	2.68	<i>p</i> <0.05

Table 8. Results of multiple linear regression analysis with "total Hg levels in muscle of common minke whales from subarea 9" as the dependent variable.

a) Analysis of Variance Table	Sum of Squaras	DE	Moon Squara	Other Statistics
Source	Sum of Squares			
Intercept B ₀	802.96	1	802.96	R ² =0.242, Adjusted R ² =0.208
Regression Bo	12.08	15	0.805	F=7.052, <i>p</i> < 0.05
Residual	37.792	331	0.114	SE=0.338
Total	852.824	347		
b) Variables				
Model	В	SE	Т	p value
Intercept	284.2	59.4	4.782	<i>p</i> <0.05
Year	-38.44	7.85	-4.90	<i>p</i> <0.05
Body length	0.419	0.492	0.85	0.395
Blubber thickness	0.168	0.089	1.88	0.061
Latitude	2.349	0.666	3.53	<i>p</i> <0.05
Longitude	-0.122	0.947	-0.13	0.897
Date	-1.485	0.291	-5.10	<i>p</i> <0.05
MainPrey_AtkaMackerel	-0.481	0.347	-1.39	0.166
MainPrey_Copepods	-0.101	0.137	-0.74	0.460
MainPrey_Euphausiids	-0.131	0.100	-1.30	0.194
MainPrey_Mackerel	0.037	0.205	0.18	0.857
MainPrey_MAFSquid	-0.261	0.166	-1.58	0.116
MainPrey_OceanicLightfish	-0.480	0.343	-1.40	0.162
MainPrey_PacificPomfret	0.693	0.140	4.94	<i>p</i> <0.05
MainPrey_Salmonids	0.173	0.249	0.70	0.486
MainPrey_Saury	0.143	0.053	2.67	<i>p</i> <0.05

Table 9. Results of multiple linear regression analysis with "total Hg levels in muscle of sei whales from subarea 9" as the dependent variable.

a) Analysis of Variance Table

Source	Sum of Squares	DF	Mean Square	Other Statistics
Intercept B ₀	900.72	1	900.72	$R^2=0.2542$, Adjusted $R^2=0.1365$
Regression B ₀	2.81	12	0.235	F=2.159, <i>p</i> < 0.05
Residual	5.656	76	0.074	SE=0.330
Total	911.789	89		

b) Variables

b) variables				
Model	В	SE	Т	p value
Intercept	451.2	182.4	2.474	<i>p</i> <0.05
Year	-58.88	23.93	-2.46	<i>p</i> <0.05
Body length	2.777	1.298	2.14	<i>p</i> <0.05
Blubber thickness	0.046	0.263	0.18	0.861
Latitude	0.394	1.183	0.33	0.740
Longitude	-2.826	1.626	-1.74	0.086
Date	-0.452	0.542	-0.83	0.407
MainPrey_Copepods	-0.196	0.131	-1.50	0.139
MainPrey_Euphausiids	-0.272	0.164	-1.66	0.102
MainPrey_Mackerel	-0.025	0.175	-0.15	0.885
MainPrey_Min.armsquid	0.055	0.244	0.23	0.822
MainPrey_Sardine	0.220	0.270	0.81	0.419
MainPrey_Saury	-0.216	0.173	-1.25	0.217

Table 10. Results of multiple linear regression analysis with "total Hg levels in muscle of Bryde's whales from subareas 8 and 9" as the dependent variable.

a) Analysis of Variance Table				
Source	Sum of Squares	DF	Mean Square	Other Statistics
Intercept Bo	277.1	1	277.13	$R^2=0.4094$, Adjusted $R^2=0.1732$
Regression Bo	0.77	8	0.096	F=1.733, p=0.152
Residual	1.109	20	0.055	SE=0.236
Total	279.0	29		
b) Variables				
Model	В	SE	Т	p value
Intercept	353.8	237.1	1.492	0.151
Year	-51.10	32.45	-1.58	0.131
Body length	3.656	1.697	2.15	<i>p</i> <0.05
Blubber thickness	0.299	0.307	0.97	0.342
Latitude	-2.978	1.708	-1.74	0.097
Longitude	6.070	4.352	1.40	0.178
Date	1.030	0.642	1.60	0.124
MainPrey_Euphausiids	-0.017	0.123	-0.14	0.889
MainPrey Mackerel	-0.029	0.396	-0.07	0.943

Table 11. Results of multiple robust linear regression analysis with "total Hg levels in muscle of common minke whales from subarea 7" as the dependent variable.

a) Residual SE and regression of	coefficient
Robust residual SE	0.20
R^2	0.135
Adjusted R ²	0.0969

b) Variables

Model	В	SE	Т	p value
Intercept	-198.59771	53.16048	-3.736	<i>p</i> <0.05
Year	26.054	6.788	3.84	<i>p</i> <0.05
Body length	-0.138	0.237	-0.58	0.559
Blubber thickness	0.059	0.060	0.98	0.328
Latitude	-0.156	0.541	-0.29	0.773
Longitude	-0.064	0.568	-0.11	0.911
Date	0.038	0.017	2.21	<i>p</i> <0.05
MainPrey_Euphausiids	0.076	0.045	1.70	0.089
MainPrey_JFSquid	0.062	0.054	1.15	0.250
MainPrey_Mackerel	0.372	0.026	14.53	<i>p</i> <0.05
MainPrey Sardine	-0.294	0.036	-8.25	<i>p</i> <0.05
MainPrey_Saury	0.097	0.047	2.07	<i>p</i> <0.05
MainPrey_WalleyePollock	0.138	0.049	2.83	<i>p</i> <0.05

Table 12. Results of multiple robust linear regression analysis with "total Hg levels in muscle of common minke whales from off Kushiro" as the dependent variable.

a) Residual SE and regression coefficient		
Robust residual SE	0.2834	
R^2	0.3472	
Adjusted R^2	0.3176	

b) Variables

Model	В	SE	Т	p value
Intercept	63.6	98.2	0.648	0.518
Year	-2.14	10.61	-0.20	0.840
Body length	0.904	0.843	1.07	0.285
Blubber thickness	-0.322	0.077	-4.15	<i>p</i> <0.05
Latitude	5.384	6.941	0.78	0.439
Longitude	-14.215	13.618	-1.04	0.298
Date	0.119	0.149	0.80	0.424

Table 13. Results of multiple robust linear regression a	analysis with "total Hg	; levels in muscle of	common minke
whales from off Sanriku" as the dependent variable.			

a) Residual SE and regression c	oefficient
Robust residual SE	0.2543
R^2	0.2374
Adjusted R ²	0.1067

b) Variables				
Model	В	SE	Т	p value
Intercept	-50.2	286.8	-0.175	0.862
Year	31.50	44.65	0.71	0.485
Body length	-0.029	0.025	-1.15	0.257
Blubber thickness	-0.400	0.274	-1.46	0.153
Latitude	23.792	17.674	1.35	0.187
Longitude	-56.288	43.990	-1.28	0.209
Date	1.079	0.569	1.90	0.066

Table 14. Results of multiple robust linear regression analysis with "total Hg levels in muscle of common minke whales from subarea 8" as the dependent variable.

a) Residual SE and regression of	coefficient
Robust residual SE	0.2439
R^2	0.3021
Adjusted R ²	0.2473

c) Variables

Model	В	SE	Т	P value
Intercept	163.35009	95.210	1.716	0.088
Year	-17.288	13.636	-1.27	0.207
Body length	1.219	0.534	2.29	<i>p</i> <0.05
Blubber thickness	0.288	0.108	2.67	<i>p</i> <0.05
latitude	-0.281	1.628	-0.17	0.863
longitude	-7.181	2.778	-2.59	<i>p</i> <0.05
Date	0.495	0.699	0.71	0.481
MainPrey_Copepods	-0.020	0.080	-0.25	0.805
MainPrey_Euphausiids	-0.104	0.144	-0.72	0.471
MainPrey_JFSquid	-0.137	0.101	-1.36	0.175
MainPrey_Mackerel	0.309	0.170	1.82	0.072
MainPrey_Saury	0.180	0.063	2.84	<i>p</i> <0.05

Table 15. Results of multiple robust linear regression analysis with "total Hg levels in muscle of common minke whales from subarea 9" as the dependent variable.

a) Residual SE and regression coefficient		
Robust residual SE	0.2834	
R^2	0.3472	
Adjusted R^2	0.3176	

b) Variables

Model	В	SE	Т	p value
Intercept	330.3	55.7	5.932	<i>p</i> <0.05
Year	-43.74	7.31	-5.99	<i>p</i> <0.05
Body length	0.150	0.441	0.34	0.735
Blubber thickness	0.164	0.092	1.78	0.076
Latitude	1.809	0.672	2.69	<i>p</i> <0.05
Longitude	-0.784	0.899	-0.87	0.384
Date	-1.424	0.301	-4.73	<i>p</i> <0.05
MainPrey_AtkaMackerel	-0.368	0.076	-4.86	<i>p</i> <0.05
MainPrey_Copepods	0.044	0.224	0.20	0.843
MainPrey_Euphausiids	-0.177	0.159	-1.12	0.266
MainPrey_Mackerel	0.050	0.139	0.36	0.722
MainPrey_MAFSquid	-0.386	0.209	-1.84	0.066
MainPrey_OceanicLightfish	-0.439	0.060	-7.30	<i>p</i> <0.05
MainPrey_PacificPomfret	0.930	0.335	2.78	<i>p</i> <0.05
MainPrey_Salmonids	0.213	0.132	1.61	0.108
MainPrey_Saury	0.188	0.050	3.73	<i>p</i> <0.05

Table 16. Results of multiple linear regression analysis with "total Hg levels in muscle of sei whales from subarea 9" as the dependent variable.

a) Residual SE and regression of	coefficient
Robust residual SE	0.2526
R^2	0.3344
A diusted R^2	0.2293

h	Variables
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.,				
Model	В	SE	Т	p value
Intercept	405.9	137.4	2.954	<i>p</i> <0.05
Year	-54.46	18.35	-2.97	<i>p</i> <0.05
Body length	3.579	1.029	3.48	<i>p</i> <0.05
Blubber thickness	0.024	0.262	0.09	0.927
Latitude	0.910	1.188	0.77	0.446
Longitude	-1.233	1.431	-0.86	0.392
Date	-0.602	0.481	-1.25	0.215
MainPrey_Copepods	-0.182	0.096	-1.90	0.061
MainPrey_Euphausiids	-0.259	0.159	-1.63	0.107
MainPrey_Mackerel	-0.064	0.097	-0.66	0.510
MainPrey_Min.armsquid	-0.012	0.105	-0.12	0.906
MainPrey_Sardine	0.193	0.104	1.85	0.069
MainPrey_Saury	-0.136	0.096	-1.41	0.164

Table 17. Results of multiple robust linear regression analysis with "total Hg levels in muscle of Bryde's whales from subareas 8 and 9" as the dependent variable.

a) Residual SE and regression of	coefficient
Robust residual SE	0.2025
R^2	0.3592
Adjusted R ²	0.1029

b) Variables				
Model	В	SE	Т	p value
Intercept	-101.1	162.8	-0.621	0.541
Year	9.93	22.02	0.45	0.657
Body length	2.431	1.453	1.67	0.110
Blubber thickness	-0.320	0.317	-1.01	0.325
Latitude	1.693	1.534	1.10	0.283
Longitude	1.933	2.810	0.69	0.499
Date	0.503	0.376	1.34	0.196
MainPrey_Euphausiids	-0.106	0.071	-1.51	0.148
MainPrey_Mackerel	-0.240	0.274	-0.88	0.391

Table 18. Comparis	son between	n muscular Hg	levels in common r	ninke whale:	s per main prey	items observed	from subarea 7 and	d the whole
body Hg levels in th	he prey item	s from the we	stern North Pacific					
		Anchovy	Euphausiids	JFSquid	Mackerel	Sardine	Saury	Walley ePollock
Muscle of whales	Ave.±SD (0.21 ± 0.051) (0.22 ± 0.049) (0.22 ± 0.03)(0.33 ±)(0.17 ±) (0.23 ± 0.043)	(0.24 ± 0.056)
	и	238	33	8	1	2	26	28
whole of prey spp.	Ave.±SD (0.037 ± 0.025) (0.005 ± 0.003) ($0.058 \pm$) (0.020 ± 0.002	2)(0.018 \pm) (0.038 ± 0.015)	(0.045 ±)
	и	20*	19*	57**	5*	e6**	41*	2*

*: Yasunaga and Fujise (2009); **: Ministry of Health, Labour and Welfare (2005) wh

Table 19. Comparison between muscular Hg levels in common minke whales per main prey items observed from subarea 9 and the whole body Hg levels in the prey items from the western Saury Salmonids **Pacific Pomfret** Oceanic Lightfish **MAFSquid** Mackerel Euphausiids Cop ep ods Atka M ackerel Anchovy North Pacific

Muscle of whales	Ave.±SD (0.22 ± 0.10)(0.13 ±) (0.17 ± 0.07) (0.18 ± 0.09	(0.19 ± 0.04)	0.19 ± 0.12)(0.11	+H) (0.44 \pm 0.26) (0.32 ± 0.08
	и	83	1	9	15	3	5		1	9	5
whole of prey spp.	Ave.±SD (0.037 ± 0.025) ($0.086 \pm$)(0.005 ± 0.003) (0.005 ± 0.003	$(0.020 \pm 0.002)($	H)(++)(0.23 ± 0.)3) (0.027 \pm
1	и	20*	61**	5*	19*	5*				3*	41**
*: Yasunaga and F	¹ ujise (2009);	**: Ministry (of Health, Labo	ur and Welfare (2	2005)						
Table 20. Compar.	ison between	n muscular Hg	; levels in sei wl	hales per main pre	y items observe	d from subarea 9 an	id the whole	: body H	g levels	8	

from the western North Pacific items in the prev

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		Anchovy	Cop ep ods	Euphausiids	M ackerel	M in.arm squid	Sardine	Saury
Muscle of whales	Ave.±SD (0.049 ± 0.013) (0.043 ± 0.014) (0.045 ± 0.016) (0.045 ± 0.016)	(0.040 ± 0.005)	(0.052 ±) (0.041 ± 0.015)
	и	11	48	6	6	б	2	7
whole of prey spp.	Ave.±SD (0.037 ± 0.025) (0.005 ± 0.003) (0.005 ± 0.003) (0.020 ± 0.002)	· ()	(0.018 ±) (0.038 ± 0.015)
	и	20*	5*	19*	5*		66**	41*
					1 0 0			

*: Yasunaga and Fujise (2009); **: Ministry of Health, Labour and Welfare (2005)

) (0.038 ± 0.015)

41*

) (0.24 ± 0.09

257



Figure 1. Sub-areas surveyed by the JARPNII research. Sub-areas based on IWC (1994), excluding the EEZ of Russia



Figure 2a. Simple plots and smoothing plots using the generalized additive model of total Hg concentrations in muscle of common minke whales (mature males, O-stock) in sub-area 7 against research years during the period 1996-2012



Figure 2b. Simple plots and smoothing plots using the generalized additive model of total Hg concentrations in muscle of common minke whales (mature males, O-stock) in off Kushiro against research years during the period 2002-2014



Figure 2c. Simple plots and smoothing plots using the generalized additive model of total Hg concentrations in muscle of common minke whales (mature males, O-stock) in off Sanriku against research years during the period 2002-2014



Figure 2d. Simple plots and smoothing plots using the generalized additive model of total Hg concentrations in muscle of common minke whales (mature males, O-stock) in sub-area 8 against research years during the period 1996-2009



Figure 2e. Simple plots and smoothing plots using the generalized additive model of total Hg concentrations in muscle of common minke whales (mature males, O-stock) in sub-area 9 against research years during the period 1994-2013



Figure 2f. Simple plots and smoothing plots using the generalized additive model of total Hg concentrations in muscle of sei whales (mature males) in sub-area 9 against research years during the period 2002-2014



Figure 2g. Simple plots of total Hg concentrations in muscle of Bryde's whales (mature males) in sub-areas 8 and 9 against research years during the period 2002-2013