An Investigation of Oxygen Isotope Fractionation during Sea Spray Icing and Observation of Snow Mass Fraction of Spray Ice

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Abstract— **The contribution of snow to the growth of sea spray icing was investigated using field observation and laboratory experiments. The oxygen isotopic composition of the melted samples was analyzed to investigate the effective fractionation coefficient and the snow mass fraction of spray ice. The oxygen isotopic composition values of spray ice were higher than that of the sea-water supply. This difference suggested two reasons. The first was that isotope fractionation had occurred during the wet growth of spray ice. The other was an effect of accretion because of the very low δ^{18}O value of snow.**

The isotope fractionation during the wet growth of spray ice was investigated using artificial spray icing produced in cold room experiments. The oxygen isotopic composition of the artificial spray ice indicates that the δ^{18}O values of the spray ice was higher than the value of the water supply. The difference between the values of spray ice and of water depends on the air temperature; the fractionation decreases with the temperature decrement. In addition, height dependence of the δ^{18}O values was shown; the fractionation increased with the height of the sampling location of spray ice.

We verified the effective fractionation coefficient using samples obtained in nature. The snow mass fraction of the spray ice samples was calculated from the isotopic mass balance. The snow mass fraction of spray ice responded to ice samples from 0 % (pure spray ice) to 97 % (snow accretion).

Keywords— ***sea spray icing, snow accumulation, δ^{18}O, isotopic mass balance, Oxygen Isotope Fractionation***

I. Introduction

Formation of spray ice is a phenomenon often found in cold regions. It accumulates on ships and offshore structures, develops into a massive ice form. Heavy marine icing often disturbs safe navigation of ships sailing. Coast of Hokkaido Island on the Sea of Japan is characterized by extreme sea-water spray icing. Spray ice on light beacons severely affects their maintenance because small light beacons on breakwater are not equipped with countermeasures for spray icing (Figure 1). On the other hand, spray ice observed around Lake Inawashiro and Lake Towada, Japan, is a popular tourist attraction [1]. Heavy marine icing often disturbs safe navigation of ships sailing. In recent years, the summer conditions of the Arctic Ocean have been changing annually; the open water area expanded and the frequency of marine icing condition under cold air temperature might increase. Therefore, it is important to investigate the mechanism of ice accretion on large vessels in order to ensure the safety of commercial vessels sailing in the Northern Sea Route.

Rapid growth of ship icing occurs owing to ice accretion caused by sea-water spray. Several recent studies have investigated the feature of sea-water spray ice. [2] developed a theoretical model of salt entrapment in sea-water spray ice; a thin liquid-water film on the icing surface runs off from the surface and consequently traps liquid in the spray ice matrix. [3] studied the microstructural features of spray ice on ships and demonstrated the presence of a channelized network of brine. [4] measured the three-dimensional microstructure of sea-water spray ice using the Nuclear Magnetic Resonance (NMR) imaging technique, and confirmed the presence of such a channelized network of brine in natural sea-water spray ice samples (Figure 2).

Numerous researchers estimated sea-water spray icing. A ship-icing prediction algorithm [5] is used operationally by the National Oceanic and Atmospheric Administration office (NOAA). The factor used to estimate the severity of potential spray icing is derived from a simplified heat balance of the icing surface. [6] investigated the growth rate of sea-water spray icing using telephotographs recorded in intervals and found that the growth rate of the cross sectional area of the spray icing increases monotonically with the product of air temperature and wind speed (i.e., the heat loss by convective heat flux). [7] reviewed computer simulations of marine ice

Fig. 1 Sea spray icing on a small light beacon on the breakwater of Afun Harbor.

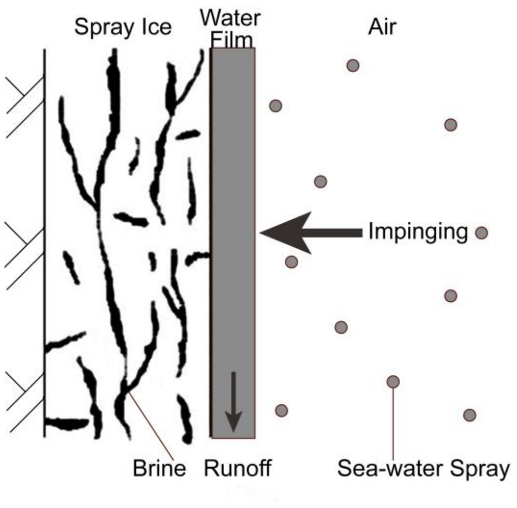

Fig. 2 A schematic vertical cross section of wet growth by sea-water spray.

accretion and discussed the U. S. Coast Guard's Cutter Midgett (115 m length and 2703 metric tons) model and a three-dimensional time-dependent vessel-icing model. [8] applied a time-dependence model, MARICE, to the prediction of marine icing. MARICE calculated the turbulent airflow, trajectories of the droplets around the complete geometry of the structure, and heat transfer from the structure. In addition, they compared the prediction of icing by MARICE and the predictions by the other 2 models (RIGICE04 and ICEMOD).

The liquid water content (LWC) of sea-water spray is an important parameter to address marine icing. Sea-water spray impinges on the superstructure, and wet growth of ice occurs from brine water flow. However, sea-water spray cloud caused by a ship is very complex because the spray generation depends on the interaction between the waves and ship. Generally, spray generation is related to the wind velocity and the wave height by open water [9]. [10] proposed the vertical distribution of LWC as a function of significant wave height, ship speed relative to the waves and height above the deck. [11] expressed LWC distribution as a function of height above the top of the wave-wash zone. [8] calculated the spray generated by the wave-structure interaction and the spray created by wind, respectively. They concluded that the wind-generated spray is unlikely to create a significant contribution to icing. [12] reported that superstructure spray flux and ice accretion on CGC Midgett. Spray cloud and droplet size measurements were made with a stroboscopic video camera set up at about 10 m above the water surface. [13] studied the water breakup phenomena of wave impact sea spray. They analyzed LWC of spray cloud for a Medium-sized Fishing Vessel (MFV). The numerical results were in reasonable agreement with field observations. [14] developed a three-dimensional model for calculating the movement of a cloud of wave-impact sea spray over an MFV.

Freezing spray is the main cause of spray icing; however, spray ice accretion often occurs during intense snowfall. It is important to estimate the contribution of snow to the growth rate of spray icing, because the release of latent heat is a rate-determining of spray ice formation, and snow that is solid water might accelerate the growth rate of icing. [15] investigated the characteristics of lake-water spray ice. The structural characteristics of the ice were analyzed using thin sections of the samples, while the snow mass fraction of the ice samples was calculated from the isotopic mass balance. They suggested that the high snow fraction in the samples indicates a significant contribution of snow. [1] investigated the contribution of snow to spray icing through field observations and laboratory experiments. They analyzed the oxygen isotopic composition of the samples of sea-water spray ice and lake-water spray ice. The δ^{18}O values of spray ice are higher than the values of the sea or lake water supply. This difference suggests two reasons. The first is that isotope fractionation has occurred during the wet growth of spray ice. The other is an effect of accretion because of the very low δ^{18}O value of snow. They verified the isotope fractionation during the wet growth of artificial spray ice produced in cold room experiments. They suggested that the difference between the values of spray ice and of water depends on the air temperature.

In this study, the contribution of snow to the growth of sea spray icing was investigated using field observation and laboratory experiments. The oxygen isotopic composition of the samples was analyzed to investigate the effective fractionation coefficient and the snow mass fraction of spray ice.

II. Method

Generally, the concentration of the heavy stable isotope ^{18}O varies with phase changes, and depends on the temperature of the phase changes [16]. The oxygen isotopic composition δ^{18}O can be calculated using the following equation:

$$\delta\,^{18}\mathrm{O} = \frac{R - R_{SMOW}}{R_{SMOW}} \times 1000 \qquad (1),$$

where R is the isotopic ratio $H_2{}^{18}O / H_2{}^{16}O$ in the sample and R_{SMOW} is the isotopic ratio in Standard Mean Ocean Water.

The snow mass fraction of the spray ice is important to consider when estimating the growth rate of spray ice. In this study, we focused on the isotopic mass balance in sea-water, snow, and spray ice. The snow mass fraction was calculated from the isotopic mass balance. [17] [18] used an isotopic mass balance to estimate the contribution or fraction of snow in sea ice, where the snow ice consists of a mixture of snow and sea-water. The stable oxygen isotopic composition δ^{18}O in an ice segment (δi) can be estimated using the δ^{18}O values of sea-water (δw) and of snow (δs) in the following equation:

$$\delta\, i = (1 - f_s) \times (\delta w + f) + f_s \times \delta s \qquad (2),$$

where f_s is the snow mass fraction in the ice segment and f is an effective fractionation coefficient associated with the freezing of sea-water. For the effective fractionation coefficient f, a value of 3.0 ‰ [19] or 2.9 ‰ [20] is used. Both values are empirical values for ice grown relatively slowly from pure water.

The oxygen isotopic composition values of spray ice were higher than that of the sea-water supply [1]. This difference suggests that isotope fractionation has occurred during the wet growth of the spray ice. However it is conceivable that the fractionation coefficient of sea ice (f = 3.0) is over-estimated,

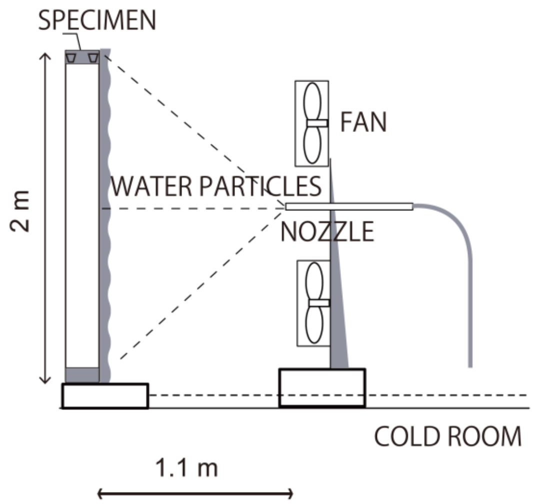

Fig. 3 Apparatus for laboratory experiment.

since the *f* value decreases with an increment in the growth rate as suggested in sea ice study of [21]. [1] obtained *f* values of spray ice as 1.41 for –20 °C air temperature, and 1.76 for –10 °C air temperature from laboratory experiment.

III. Laboratory Experiment

A. *Equipment and Method*

In this study, the data of cold room experiments in [1] were re-analyzed, and the isotope fractionation during the wet growth of spray ice was investigated in detail. The experimental apparatus was set in a cold room at Hokkaido University of Education. Figure 3 shows a schematic view of the apparatus. The air temperature in the cold room can be arbitrarily set between –10 °C and –25 °C. In this study, it was set to –20 °C and –10 °C. The height and diameter of the cylindrical test specimen were 2 m and 0.3 m, respectively. The distance of the specimen from the spray nozzle was 1.1 m. The fan-shaped spray nozzle had a particle size distribution of approximately 0.3-0.5 mm. Fresh water and 3 ‰ salt water, which is almost the same as the sea-water concentration in the Sea of Japan, were used as spray water supply. The feed temperatures of fresh water and salt water were about +1°C and about –1°C, respectively, which were approximately +1°C higher than the freezing point. The analysis was performed divided into four brocks for each height of the cylindrical specimen. Oxygen isotopic composition of the melted samples was analyzed using a standard mass spectrometer.

B. *Experimental Results of Isotope Fractionation*

Laboratory experiments of wet growth were conducted 10 times under the aforementioned conditions. The duration of spray supply was 30 min. Part of the spray water froze into spongy ice as it flowed down the specimen surface, and the surface gradually began to be covered by sheet-like ice.

Isotope fractionation has occurred during the wet growth of the spray ice. We investigated the difference between the fractionation coefficient of fresh water and salt water during the wet growth of spray ice. The δ^{18}O value of the artificial spray ice was obtained every 50 cm in height of the cylindrical specimen, and the difference between the spray ice and the supply water was calculated.

Figure 4 shows the difference of δ^{18}O value between spray ice (δi) and supply water (δw). The air temperature was –20 °C. The ($\delta i - \delta w$) values of fresh water and salt water at the height of each block were almost the same. Therefore, no difference in the isotopic fractionation was found between fresh water and salt water at –20 °C.

Figure 5 shows the difference of δ^{18}O value between spray ice and supply water at the air temperature of –10 °C. The (δi–δw) values of salt water at the height of each block were slightly larger than those of fresh water, although both were very close. It is conceivable that the difference in freezing temperature between fresh water (0 °C) and salt water (–1.9 °C) affected the growth rate of the spray ice. On the other hand, the freezing temperatures difference between the fresh water and saltwater might be ignored when the air temperature was –20 °C, which was cold enough.

Figure 6 shows the difference between the δ^{18} O values of spray ice and water supply when the temperature was –10 °C and the temperature was –20 °C at the height of each block. The (δi–δw) values at –10 °C at all heights was greater than that at –20 °C. It means that the spray ice which grew at

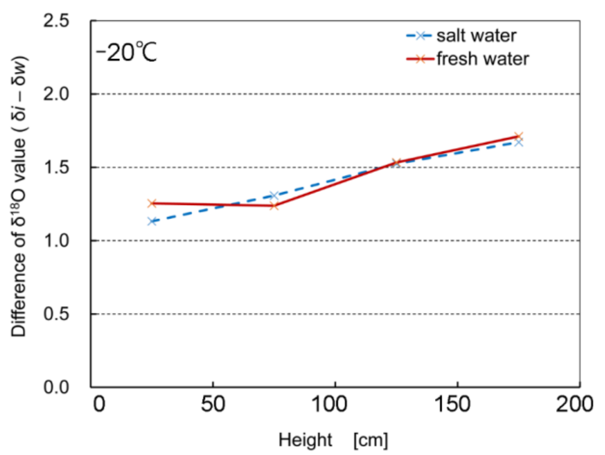

Fig. 4 Difference of δ^{18}O value between spray ice and supply water at the air temperature of –20 °C.

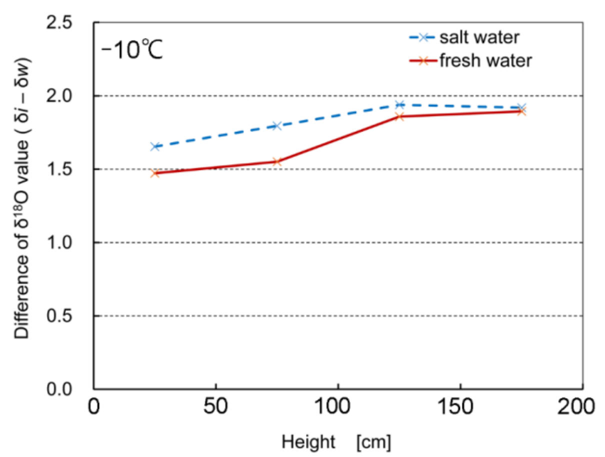

Fig. 5 Difference of δ^{18}O value between spray ice and supply water at the air temperature of –10 °C.

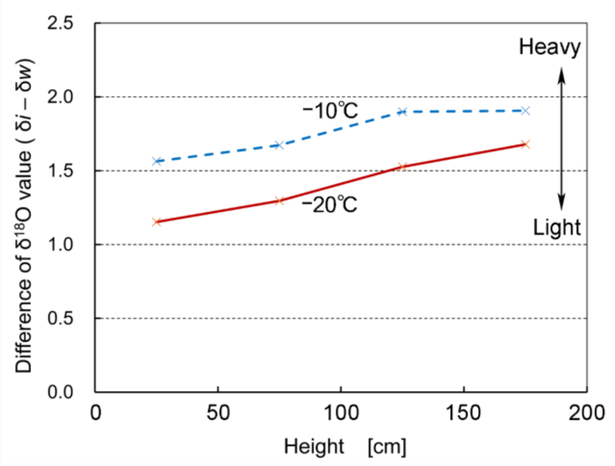

Fig. 6 Comparison of (δ*i* – δ*w*) values between –10 °C and –20 °C.

TABLE 1. Height dependence of the difference in $\delta^{18}O$ values between spray ice and supply water.

Height	–10 °C	–20 °C	Difference
150–200	1.56	1.15	0.41
100–150	1.67	1.30	0.37
50–100	1.90	1.53	0.37
0–50	1.91	1.68	0.23

–10 °C was heavier than that which grew at –20 °C. The differences at each height are shown in table 1. The average of the difference was approximately 0.35. Therefore, it is considered that a temperature difference of 10 degrees during spray icing affected to the fractionation coefficient.

In addition, the (δ*i*–δ*w*) values at low position tended to be smaller than that at the high position. The (δ*i*–δ*w*) values of the spray ice sampled at 150-200 cm were 1.91 at –10 °C and 1.68 at –20 °C, whereas the (δ*i*–δ*w*) values of those sampled at 0–50 cm were 1.56 at –10 °C and 1.15 at –20 °C. In other words, the $\delta^{18}O$ value of spray ice decreased approximately 0.3 with 100 cm going down.

C. Discussion

The fractionation coefficient of the –10 °C experiments is higher than that of the –20 °C experiments. This tendency is consistent with the sea ice study in [21], which suggested that the *f* value decreases with an increment in the growth rate. The wet growth of spray ice depends on the cooling rate of the water film, and the major factor of the cooling is convective heat flux. Therefore, it is conceivable that the difference between freezing temperature and air temperature affects the growth rate.

In Figure 5, the (δ*i*–δ*w*) values of salt water were slightly larger than those of fresh water. In the –10 °C air temperature, the difference between the freezing temperatures of fresh water and 3% salt water, which was about 2 °C, could not be ignored on the growth rate of spray ice. Therefore, the growth rate of salt water was slightly slower than that of fresh water, so that δ*i* of salt water might be slightly larger than δ*i* of fresh water.

Figure 6 shows that the (δ*i*–δ*w*) value depends on the sampling height. During the wet growth, a part of the film flow freezes and the rest runs off to the bottom (Figure 2). Then, the film flow at the lower brock consists of spray supply and advection water from the upper block. The concentration of the heavy stable isotope ^{18}O varies with phase changes from liquid phase to solid phase, so that spray ice becomes heavier than the residual liquid water. Consequently, the advection water must have been lighter than the spray supply. By repeating this process from top to bottom of the specimen, it was suggested that the $\delta^{18}O$ value of the spray ice in the lower brock became lower than that in the upper brock.

IV. OBSERVATIONS

A. Observational Sites

The observations of sea-water spray ice were conducted at the Hamamasu Harbor, the Afun Harbor, and the Toyosaki Harbor on the west coast of Hokkaido Island, Japan (Figure 7). At the Hamamasu, two dummy light beacons were set up at

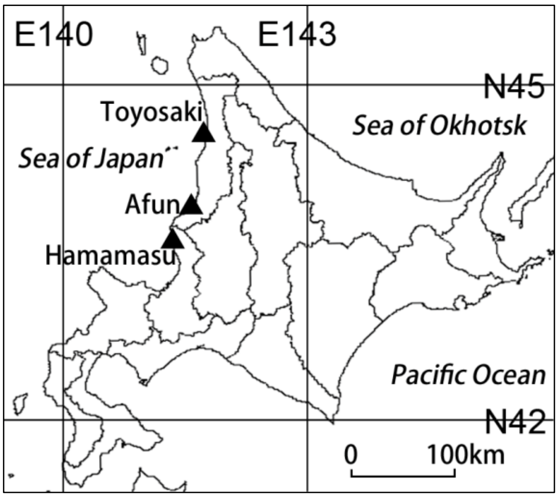

Fig. 7 Locations of observational sites. Hamamasu, Afun, and Toyosaki on the west coast of Hokkaido Island.

Fig. 8 Icebreaker Shirase (12,650 GRT).

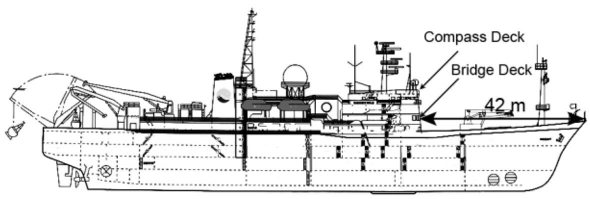

Fig. 9 R/V Mirai (8,706 GRT).

TABLE 2. δ^{18}O values of the spray ice, snow, and sea-water that obtained on the light beacons.

Sample	δ ^{18}O (‰)		
	Icing	Snow	Sea-water
Hamamasu			
H1	-1.61	-15.5	-0.52
Afun 1			
A1-1	2.16	-10.1	-0.39
A1-2	2.05		
Afun 2			
A2-1	1.32	-9.81	-0.02
A2-2	1.55		
A2-3	1.82		
Afun 3			
A3	1.56	-11.0	-0.22
Toyosaki			
T1	0.96	-12.1	-0.32

TABLE 3. δ^{18}O values of the spray ice, snow, and sea-water that obtained on the icebreaker Shirase and the R/V Mirai.

Sample	δ ^{18}O (‰)		
	Icing	Snow	Sea-water
Shirase			
J1-1	-13.10	-23.4	-0.16
J1-2	-5.03		
J1-3	-1.28		
J2-1	-1.06		
J2-2	-4.25		
R/V Mirai			
M1	-11.60	-11.90	-2.97

the breakwater extending from the north to the south. The height of both dummy light beacons was approximately 4 m.

The light beacons at Afun (Figure 1) and Toyosaki were both actually used, and they are located the end of a breakwater that projects from the south of the harbors to the offshore. The size of each was about 2m. All these beacons were always exposed to the strong spray jets generated by the high waves caused by the primary wind during the winter season. The spray icing grew under intense sea-water spray and cold conditions. Additionally, stormy weather often generated not only heavy spray jets, but also intense snowfall [1]. We collected samples of spray ice on the light beacons, deposited snow, and sea-water.

In Addition, samples of ship icing, deposited snow, and sea-water were collected at the icebreaker Shirase (Figure 8), and the R/V Mirai (Figure 9). The field observation was conducted on the icebreaker Shirase (12,650 GRT) in the 57th Japanese Antarctic Research Expedition. The field observation was conducted on the R/V Mirai (JAMSTEC; 8,706 GRT) from 22 August to 2 October 2016. The ship was conducting meteorological and hydrographic surveys in the northern Bering Sea and the Arctic Ocean under the Arctic Challenge for Sustainability Project.

B. Observational Results

Table 2 shows the δ^{18}O values for the spray ice, snow, and sea-water samples that obtained on the light beacons at the west coast of Hokkaido Island. The sea-water δ^{18}O values were near 0, which was considered as a standard value for seawater. The δ^{18}O values of snow varied from –10 to –16 ‰, which depended on the precipitation event. It was a sufficiently low value compared to the value of sea-water. It is remarkable that most of the δ^{18}O values of spray ice were higher than the values of the sea-water supply. It meant that not only the origin of sea-water splay, but also the isotope fractionation affected the δ^{18}O values of spray ice.

Table 3 shows the δ^{18}O values for the ship icing, snow, and sea-water samples that obtained on the icebreaker Shirase and the R/V Mirai. Two icing events were recorded in the observation of the Shirase. The sea-water sample was collected in the first event, and the snow sample was obtained on the sea ice off Syowa Station (East Antarctica). In Antarctica, it was characterized that the δ^{18}O value of the snow was very low compared to the others. We could not obtain δ^{18}O values for the snow and sea-water samples of the second event, here we assume the same values as for the first event. The δ^{18}O values of ship icing varied from –1 to –13 ‰, which depended on the icing event and the sampling location.

The icing sample of R/V Mirai was snow accretion, because it grew in weak wind condition. As a result, the δ^{18}O value of ship icing was as low as –11.60 ‰. Moreover, since the sea-water sample was obtained near the sea ice area, the value less than 0 ‰ was shown.

Air temperature, wind speed, and growth rate of the spray ice fluctuates from hour to hour in nature, therefore the effective fractionation coefficient is not settled. In this study, we assumed an effective fractionation coefficient f of 1.8, which was as same as average of the fractionation coefficient at the –10 °C experiments, for the calculation of the snow mass fraction.

Figure 10 indicates the snow mass fractionation of the spray ice samples in Table 2 and Table3. The snow mass fraction of spray ice responded to ice samples from pure spray ice to snow accretion. In this study, most samples obtained from light beacons (H, A and T) showed no snow accretion effect. The values for A1-1 and A1-2 were slightly negative, which might be due to the assumption of the f value. We considered these samples to be pure spray ice.

The contribution of snow accretion was observed in the ship icing samples. In the event of J1, the contribution rate varied from 10 to 60 %, which depended on the sampling location on the ship. In the R/V Mirai event (M1), the snow mass fractionation was 97%, the largest value observed. This result was consistent with the observation that it was a snow accretion event. As a result of calculating snow mass fraction of the spray ice samples, it was suggested that the effective fractionation coefficient of 1.8 was appropriate.

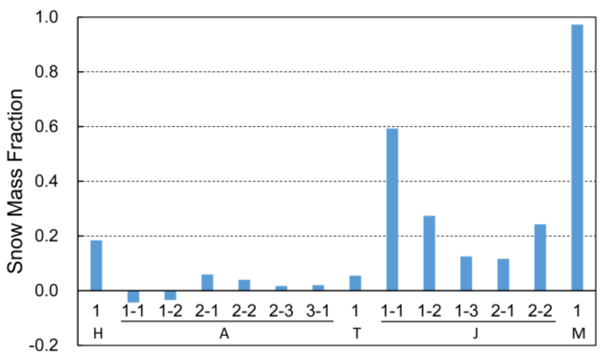

Fig. 10 Snow mass fraction of spray ice. H: Hamamasu, A: Afun, T: Toyosaki, J: Shirase, M: R/V Mirai.

V. CONCLUSIONS

In this study, the contribution of snow to the growth of sea spray icing was investigated using laboratory experiments and field observations. In the laboratory experiments, the $\delta^{18}O$ value of the artificial spray ice was obtained every 50 cm in height of the cylindrical specimen, and the difference between the spray ice and the supply water was calculated. The difference between the fractionation coefficient of fresh water and salt water during the wet growth of spray ice was investigated, and the difference was negligible when the temperature was –20 °C. On the other hand, it is conceivable that the difference in freezing temperature between fresh water (0 °C) and salt water (–1.9 °C) affected the growth rate of the spray ice, and affected to the fractionation coefficient. In addition, the (δi-δw) values at low position tended to be smaller than that at the high position. By repeating this process from top to bottom of the specimen, it was suggested that the $\delta^{18}O$ value of the spray ice in the lower brock became lower than that in the upper brock.

We verified the effective fractionation coefficient using samples obtained in nature. For calculating the snow mass fraction, effective fractionation coefficient f was assumed to be 1.8. The observations of sea-water spray ice were conducted on the light beacons at the west coast of Hokkaido Island, and the ship icing at the icebreaker Shirase and the R/V Mirai. The snow mass fraction of spray ice responded to ice samples from pure spray ice (0 %) to snow accretion (97 %). In this study, most samples obtained from light beacons showed no snow accretion effect. On the other hand, the contribution of snow accretion was observed in the ship icing samples.

As a result of calculating snow mass fraction of the spray ice samples, it was suggested that the effective fractionation coefficient of 1.8 was appropriate.

ACKNOWLEDGMENT

We wish to express our gratitude to Prof. H. Yamaguchi of University of Tokyo, and Prof. J. Inoue of National Institute of Polar Research for their support to this research. We would like to thank to Mr. K. Yamane of Hokkaido University of Education, and Mr. S. Toda of University of Tokyo for their support in laboratory experiments and field observations. We would like to thank to Ms. M. Kitagawa of Hokkaido University for their support of isotope ratio measurements. We are grateful to 1st Regional Coast Guard Headquarters for their support in field observation at Hokkaido. The field observation on the icebreaker Shirase was performed in collaboration with 57th Japanese Antarctic Research Expedition. We wish to acknowledge useful support from crews of the icebreaker Shirase and R/V Mirai. We wish to acknowledge support from Arctic Challenge for Sustainability Research Project. This research was supported by JSPS KAKENHI Grant Number 18K02929.

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