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High nitrogen isotope ratio for soils of seabird rookeries

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Abstract. Soils from rookeries of penguin, of gull, and of albatross were examined for their nitrogen isotope ratio. The ratio was far higher than any so far reported for soils. Furthermore, there was an apparent dependence of the ratio on the latitude of rookery locations. The high ratio seemed to have had resulted from a relatively high ratio for incoming nitrogen to the rookeries, and from the large fractionation factor during the ammonia volatilization from the rookeries. The isotope ratio for ammonium nitrogen of the penguin rookery soils averaged 45 per mil, while that of the gull rookery soils gave the mean of 27 per mil during the breeding season of the birds. Soils of gull rookeries and of albatross rookery gave, on the average, a similar ratio of 17 per mil for Kjeldahl nitrogen, though its content of soils of gull rookeries was nearly two orders of magnitude higher than that of albatross rookery. Soils from penguin rookery showed the ratio of 32 per mil for Kjeldahl nitrogen, the enrichment of ^{15}N being two and a half times as large as that for soils of other rookeries.

Introduction

Some avian species form rookeries during their breeding season on islands that are often located for secure breeding several hundred or thousand kilometers away from the nearest continent. A large quantity of organic matter is deposited in the rookery during the breeding season and few other nutrient inputs exist there throughout the year. Along with this seasonal change in the nutrient flow, microbial activities of rookery soils exhibit a seasonally synchronized change (Mizutani, 1982; Ishizuka et al., 1985).

It has been estimated that annual inputs from bird excreta alone of nitrogen, phosphorus, and potassium to a Black-tailed Gull (*Larus crassirostris*) rookery are respectively 53 $g \cdot m^{-2}$, 45 $g \cdot m^{-2}$, and 21 $g \cdot m^{-2}$. These amounts are much higher than those to the cultivated fields in some European countries, in Korea, and in Japan (Mizutani, 1984), where the most fertilizer-intensive agriculture in the world is currently undertaken (Ohtake, 1982).

Therefore, seabird rookeries must be a unique natural environment in terms of nutrient dynamics. And, as the distribution of $^{15}N/^{14}N$ reflects sources and metabolism of nitrogen in a biogeochemical system (Wada, 1980), a study of the nitrogen isotope composition of rookery soils should help to elucidate the nature of the nutrient flow.

Materials and methods

Sampling locations

Rookeries chosen for the present study were as follows: one Adelie Penguin (*Pygoscelis adeliae*) rookery in Antarctica, six Black-tailed Gull (*Larus crassirostris*) rookeries in Japan, and one Short-tailed Albatross (*Diomedea albatrus*) rookery in Torishima (alias: Ponafidin Island), Japan. The Torishima rookery is the only rookery of the albatross known to date (Hasegawa, 1984).

The penguin rookery was in Cape Bird, Ross Island, Antarctica (77° 12′S, 166° 28′E). Soils, bottom mud of a pond, and bodies of a dead chick and a dead juvenile were obtained on January 6, 1981 (Nakaya et al., 1982), which was about a few days before the formation of crèche (Aoyanagi, 1981). The pond was located near the center of the rookery.

Black-tailed Gull rookeries were Kabushima rookery (40° 32′ 12″N, 141° 33′41″E) and Bentenshima rookery (40° 30′8″N, 141° 37′23″E), both in Hachinohe, Aomori Prefecture, Japan, Tsubakishima rookery (38° 55′59″N, 141° 43′14″E), Rikuzentakata, Iwate Prefecture, Japan, Fumishima rookery (35° 26′38″N, 132° 37′46″E), Taisha, Shimane Prefecture, Japan, and Okihebishima rookery (35° 4′48″N, 132° 19′33″E) and Matsushima rookery (35° 4′20″N, 132° 19′14″E), both in Yunotsu, Shimane Prefecture, Japan. In the following discussion, results from Okihebishima rookery and from Matsushima rookery were combined and given as those from Yunotsu area because of their mutal proximity. For the purpose of comparison, soil samples were also obtained from Aomatsushima, an island about 700 m apart from Tsubakishima, where some gulls from Tsubakishima nest in spite of a discouragement by man (Toukai Shimpo, 1976).

In Tsubakishima, a negligibly small number of Black-crowned Night Herons (*Nycticorax nycticorax*) also nest in the same area, though their breeding success is uncertain. In all the other gull rookeries, Black-tailed Gulls are the only breeding bird species. Dates of sampling were March 1, May 14, June 9, and July 21, 1980, May 13, July 24, and September 11, 1981, January 21, 1982, and June 20, 1983 for Kabushima rookery, May 14, 1981 for Bentenshima rookery, June 5, 1982 for Tsubakishima rookery, May 16, 1981 for Aomatsushima, October 4, 1980 and July 29, 1981 for Fumishima rookery, and June 8–9, 1984 for Okihebishima and Matsushima rookeries.

A Shinto shrine is located at the top of Kabushima, and tens of thousands of worshipers visit the shrine every year. The entrance to the rookery, however, is strictly forbidden during the breeding season. All other gull and albatross rookeries are on uninhabited islands, though occasional visits by fishermen and others are not impossible. Though it varies depending on the rookery and on the year, the breeding season for the gulls is about from February to August. Detailed accounts of the breeding activity and geographical features of the islands were given by Narita (1985) for Kabushima

and Bentenshima rookeries, and by Nakai (1981) for Fumishima and Okihebishima rookeries. Ishizuka (1966) gave a brief survey for Tsubakishima rookery as well as for Kabushima and Fumishima rookeries. Matsushima rookery is a newly established rookery (since about 1979), and Nakai (1986) gives the most comprehensive account yet available in literature.

From Torishima rookery (30°29′N, 140°18′E), Izu Islands, Japan, volcanic ash soils were obtained on March 25, June 22, and November 17, 1982. Black-footed Albatrosses (*Diomedea nigripes*) also nest in Torishima. Their population during the breeding season is about five hundred and exceeds that of the Short-tailed Albatross (Hasegawa, 1984). Both albatrosses nest on slopes of Tsubamezaki of the island; however, the nesting area of the Black-footeds is almost always a lower portion of the slopes while the Short-taileds nest on a higher portion. Therefore, chemical contamination of the rookery soils of the Short-tailed Albatross by the Black-footed seems minimal. The breeding season of the Short-tailed Albatross is from October to May (Hasegawa and DeGange, 1982).

Soil preparations

Soil samples from the penguin rookery were obtained by scooping the surface soil at an approximately 5 cm depth. The pond was very shallow (about 10 cm at the deepest), and the bottom mud was sampled in a similar manner to that for the soil. These samples were kept frozen until they were ready for the analyses. They were subjected without drying to the measurement of their nitrogen content and to the mass analysis. Their water content was separately obtained by drying a portion of them.

From the gull rookeries, surface soil (0–5 cm) was collected by a core sampler (core diameter: 20.5 mm). The sampling was normally done after a few consectuive days of fine weather so that the soils to be collected were fairly dry. The core sample was transferred to a plastic vial, and the vial was filled with ethanol and sealed tightly. For the study of vertical profile of various nitrogen-containing fractions, the soil from the Tsubakihsima rookery was cut to expose a vertical section, and four layers of thickness 5 cm each were sampled starting with the lowest layer (15–20 cm depth) and ending at the top layer (0–5 cm depth) in a stainless-steel cylinder (height 50 mm, I.D. 50 mm) with as much care as possible not to disturb the original structure of the soil. Immediately after collection, 8.6 g of ethanol was added to the soil in the cylinders. The purpose of the additon of ethanol was to hinder the soil microbial activity; in particular, to halt the otherwise rapid decomposition of uric acid and the resultant formation of ammonia after the sampling (Mizutani and Yamada, 1982). No ethanol was, therefore, added to the cores sampled outside the breeding season. The sealed vials and cylinders were put in a refrigerated container and moved to the laboratory. Upon arrival at the laboratory, the seal was opened, and the contents were vacuum dried by a Labconco FDC-8 freeze dryer (Labconco Corp., Kansas City, MO.,

U.S.A.). After the drying, the soil was passed through a 2 mm stainless-steel sieve to eliminate large particles such as pebbles, plant debris, and animal remains. The soil was then homogenized to pass a 0.5 mm sieve and was subjected to the analyses. When necessary, a small portion of the surface soil was kept separately in a vial without an addition of ethanol for the measurement of its water content.

For the albatross rookery, about 250 g of surface soil, taken from the uppermost 5 cm of depth, was scooped and put into a plastic bag. The soil samples appeared to be unweathered volcanic ash. Care was taken, when at all possible, to collect the soil after the absence of a precipitation for at least a few consecutive days in order to make sure that the soil was fairly dry. After the collection, the soil was spread out over a vinyl sheet, further dried in the shade for 24 h, and then returned to the plastic bag. It took about two weeks before the sample reached the laboratory. Upon its arrival, it was vacuum dried, passed through a 2 mm stainless-steel sieve to eliminate large particles, and kept in a vial until ready for the analyses.

The drying procedure applied to soils of the gull rookeries and the albatross rookery was necessitated to eliminate volatile components of soils and to normalize all the samples of initially various water content; however, it offered an opportunity to alter the nitrogen isotope ratio in the soils. The extent of the possible alteration was, therefore, examined with soils from Kabushima rookery.

When the soils were more or less dry like those actually collected from the rookeries, less than one hundredth of the KCl-extractable ammonium in the soils was found to come out during the drying procedure. The nitrogen isotope ratio of the outcoming ammonia was approximately 30 per mil lighter than that remained in the soils. Only the small portion of the ammonium in soils escaped, probably because of a certain degree of moisture is necessary to cause the ammonia volatilization (Bouwmeester et al., 1985), and a dry condition makes the soil ammonia tightly adsorbed on the soils and also reduces the soil bacterial production of ammonia.

Even if it is assumed that all the ammonia escaped during the vacuum drying was of soil ammonium, the resultant enrichment of ^{15}N in the remaining soil ammonium was less than 0.3 per mil. This means that the drying procedure causes in most cases an increase of less than 0.06 per mil in Kjeldahl nitrogen of the soils examined, because the ammonium usually accounts for less than 20% of the soil Kjeldahl nitrogen.

Chemicals

Water was distilled in an all-glass Yamato Auto Still Model WAG-24 (Yamato Kagaku, Chuo-ku, Tokyo, Japan) from deionized water. Potassium chloride used for the extraction of soil ammonium and potassium dihydrogenphosphate used for the chromatographic isolation of uric acid were guaranteed reagents from Wako Pure Chemical Industries, Ltd., Higashi-ku, Osaka, Japan.

Potassium chloride was baked in a muffle furnace at 500 °C for 2 h before dissolving it in water to make 2 M KCl extractant. Potassium dihydrogenphosphate was heated at 95 °C for 3 h in an Ikeda VOD-4-2 vacuum drying oven (Ikeda Rika Ltd., Chiyoda-ku, Tokyo, Japan) under a flow of an ammonia-free air.

Determination of Kjeldahl nitrogen content

Organic nitrogen in a soil sample was converted to ammonia by a Kjeldahl digestion. The digestion was done for 4 h in the presence of a small amount of 3 : 1 : 4 mixture in weight of HgO, SeO, and K_2SO_4 as a catalyst (Wada and Hattori, 1972). The ammonia thus produced was steam-distilled and collected in a 0.25 N H_2SO_4 trap. The ammonium sulphate solution was diluted to volume with water in a 100 ml volumetric flask. Kjeldahl nitrogen was determined by using an aliquot of the diluted ammonium sulphate solution. The remainder was used for nitrogen isotopic measurement of Kjeldahl nitrogen. The phenolhypochlorite method (Solorzano, 1969) was employed for the determination, and the absorbance at 640 nm was measured by a Shimadzu UV-210A double-beam spectrophotometer (Shimadzu Seisakusho Ltd., Nakagyo-ku, Kyoto, Japan). Standard solutions of ammonium chloride was used for the calibration.

Determination of uric acid content

Approximately 1 g of dried soil was weighed in a 100-ml beaker and 20 ml of 0.1 M KH_2PO_4 solution was added. The beaker was covered with a piece of aluminium foil and the soil suspension was stirred with mild heating for 10 min. The suspension was then transferred into a 50-ml centrifuge tube and centrifuged at 3000 rpm for 10 min. After centrifugation, the supernatant was pressure-filtered through a pre-heated (at 420 °C for more than 5 h) Whatman glass fiber filter (GF/C; diameter 4.7 cm; Whatman, Maidstone, U.K.) and a 0.45 μm microporous filter (Type HA; diameter 4.7 cm; Millipore, Bedford, MA, U.S.A.). The uric acid was separated from other components in the extract by a high-performance liquid chromatography (Mizutani and Wada, 1985a), and its concentration was converted to the amount originally present in the soil.

Mass analysis

Stable nitrogen isotope measurements were made by a Hitachi RMU-6R mass spectrometer with a dual inlet system and a double collector for ratiometry. Nitrogen isotope ratios were expressed as per mil deviations from atmospheric nitrogen as defined by the following equation:

$$\delta\,^{15}\mathrm{N}\ (\text{per mil}) = \frac{(^{15}\mathrm{N}/^{14}\mathrm{N})\ \text{sample} - (^{15}\mathrm{N}/^{14}\mathrm{N})\ \text{air}}{(^{15}\mathrm{N}/^{14}\mathrm{N})\ \text{air}} \times 1000$$

Two ammonium sulfate solutions with $\delta^{15}N$ of -3.4 and 1.3 per mil were used as running standards. The standard deviation of the nitrogen isotope measurements was 0.2 per mil.

Isotopic analysis of Kjeldahl nitrogen

The determination of the amount of Kjeldhal nitrogen and its isotopic analysis were performed as follows. An appropriate amount of a sample was weighed and its organic nitrogen was converted to ammonium by the Kjeldahl digestion as described. After determining its ammonium concentration, the ammonium sulphate solution was concentrated to approximately 1 ml in an Ikeda VOD-4-2 vacuum drying oven (Ikeda Rika Ltd.). A portion of the ammonium sulphate solution (less than 10 mg N) was transferred to a side of a dog-legged tube and 2 ml of an alkaline hypobromite solution for every 7 mg N of the ammonium sulphate solution was put in the other side of the tube. An adequate amount of 2.5 N potassium hydroxide solution to neutralize the excess sulfuric acid in the sulphate solution was added to the hypobromite solution. The tube was attached to a vacuum line for nitrogen purification. The contents of the two legs were repeatedly frozen, evacuated, and thawed to degas any atmospheric nitrogen. After the third thawing, the contents were mixed well *in vacuo* and the resulting reaction converted the ammonium nitrogen to nitrogen gas. The nitrogen gas thus produced was purified by passing it through a CuO furnace with Pt wire heated at 700 °C and through a Cu furnace heated at 400 °C. The nitrogen gas thus purified was introduced to the mass spectrometer, and the stable nitrogen isotope measurement was made.

The alkaline hypobromite solution was prepared in a hood as follows: 40 g of potassium hydroxide and 0.22 g of potassium iodide were dissolved into a 170 ml of distilled water; the solution was put in an ice bath and cooled to less than 5 °C; 25 g of bromine was added at once to the solution and the solution was stirred well; and the final solution was stored in a brown-glass bottle and kept in a refrigerator.

Preparation and isotopic analysis of ammonium fraction

The extraction procedure of soil ammonium for later isotopic analysis was the modification of that of Bremner and Keeney (1966) for the determination of soil ammonium content, and was carried out as follows (Mizutani et al., 1985a). Approximately 5 g of soil and 100 ml of 2 M KCl solution were put in a flask, and vigorously shaken at room temperature for 1 h. The content was then centrifuged at 3000 rpm for 10 min, and the resultant supernatant was pressure-filtered through a pre-heated (at 420 °C for more than 5 h) Whatman glass fiber filter (GF/C; diameter 4.7 cm) and 0.45 μm microporous filter of the same diameter (type: HA; Millipore). The filtrate was subjected to the steam-distillation, and the ammonia evolved was collected in a 0.25 N H_2SO_4 trap. The analytical procedure after the steam-distillation was the same as that given earlier for the Kjeldahl nitrogen determination.

Preparation and isotopic cnalysis of feathers and other bird organs

Fairly dry feathers of various kinds were obtained from seabird rookeries and nearby areas. They were transferred to laboratory at an ambient temperature, rinsed and cleaned with water, and then vacuum-dried. A middle portion of the dried, primary feather was cut out by a pair of stainless-steel scissors. It was weighed, further cut into small pieces, and subjected to the Kjeldahl digestion. All the procedure following the digestion was the same as that for Kjeldahl nitrogen of soil.

The dead bodies of the chick and the juvenile of the Adelie Penguin were found in the rookery in Cape Bird. The appearance of the chick body suggested that less than a few days had been passed after its death, while the juvenile must have died earlier. Various organs were obtained from these bodies. Those from the juvenile had been naturally desiccated and was kept in a vial during the transport to laboratory. The chick organs did not appear sufficiently dried, and a small amount of ethanol was sprinkled onto them after an additional few days of natural desiccation. They were, then, stored in a vial together with a small amount of ethanol until their arrival at laboratory. Both chick and juvenile organs were vacuum dried and weighed in laboratory. The procedure after the desiccation for the measurement of nitrogen content and its isotope ratio was the same as that for Kjeldahl nitrogen of soil.

Results and discussion

Local variation of content and isotope ratio of soil ammonium and Kjeldahl nitrogens

The local variation of soil ammonium content was examined with the soil collected on May 13, 1981 in Kabushima rookery. A quadrat of the size of about 1 m by 1 m was selected in the rookery, and a grid pattern, each line separating approximately 20 cm from one another, was drawn in the quadrat. The analyses of soil cores from 23 arbitrarily chosen crossing points of the grid gave 1.0 ± 1.4 mg N·(g dry soil)$^{-1}$ as the soil ammonium content. Since most ammonium must have had come from the aerobic degradation of uric acid (Mizutani and Wada, 1985a), the large fluctuation of the soil ammonium content appeared to reflect a short-term heterogeneity of the mode of the deposition and the degradation of uric acid, and of the mode of the loss of ammonium.

Though not as much as the ammonium content, the nitrogen isotope ratio for soil ammonium also fluctuated greatly. Four samples from a site in Kabushima collected on June 20, 1983 gave the average of 29.7 per mil with the standard deviation of 4.6 per mil. The fluctuation was greater for four samples obtained from the same site on July 24, 1981; the mean of the ratio was 34.7 per mil and the standard deviation was 9.1 per mil.

In a clear contrast with the large variation in soil ammonium content, Kjeldahl nitrogen content and its isotope ratio were relatively constant. In order to know the extent of fluctuation of Kjeldahl nitrogen content and its isotope ratio of rookery soils, a 3 m by 3 m area whose sides running from north to south and from east to west was arbitrarily chosen in Fumishima rookery. Soil cores were sampled from the four corners of the area. After the collection, a 1 m by 1 m square whose centre was located at the same position as that of the larger one was set up, and the cores were obtained from the four corners of the small square. Then, like a Chinese box, another square of 50 cm by 50 cm in size was established inside the small square, and the cores were taken from its four corners and the middles of its every side. And finally, a core was sampled from the common center of the three squares.

Figure 1 shows the Kjeldahl nitrogen content of these soil cores and their nitrogen isotope ratio. For the outermost four corners of the 3 m by 3 m square, the content averaged 7.4 mg N·(g dry soil)$^{-1}$ with the standard deviation of 4.6, which gave 61% as the coefficient of variation, whereas the four corners of the 1 m by 1 m square yielded 9.4 ± 3.0 mg N·(g dry soil)$^{-1}$ (coefficient of variation: 32%) and eight points of the 50 cm by 50 cm square gave 9.3 ± 2.1 mg N·(g dry soil)$^{-1}$ (coefficient of variation: 23%). The overall average for Kjeldahl nitrogen content was 8.8 ± 2.9 mg N·(g dry soil)$^{-1}$. As Kjeldahl nitrogen includes ammonium nitrogen that accounts, on the average, for about 15% of Kjeldahl nitrogen in rookery soils during breeding season, the content of organic nitrogen must fluctuate less than that of Kjeldahl nitrogen.

The isotope ratio of Kjeldahl nitrogen varied still less. The ratio for the outermost four corners was 18.5 ± 1.2 per mil, while the four corners of the 1 m by 1 m square yielded 16.9 ± 0.7 per mil and eight points of the 50 cm by 50 cm square 16.6 ± 0.7 per mil. The overall average of the ratio was 17.1 ± 1.1 per mil. With a standard deviation of about 1 per mil, an isotope ratio of Kjeldahl nitrogen from one soil core could, therefore, be regarded representative for a site whose area is at least several square meters.

Content and isotope ratio of soil Kjeldahl and ammonium nitrogens of various seabird rookeries

Table 1 summarizes Kjeldahl nitrogen content of soils and their nitrogen isotope ratio. The bottom mud of the pond in the penguin rookery exhibited nearly the same content and ratio as those of the soil, and the pond is included in Table 1 as one of the five studied sites of Cape Bird rookery. Table 2 shows ammonium nitrogen content of soils and their isotope ratio. Because the amount and the nitrogen isotope ratio of ammonium present in the soils appear dependent on the breeding activity of seabirds, Table 2 distinguishes whether they were obtained during the breeding season or outside the breeding season. Mean and standard deviation in the two tables were obtained by treating equally all the data given in Appendix at the end of this

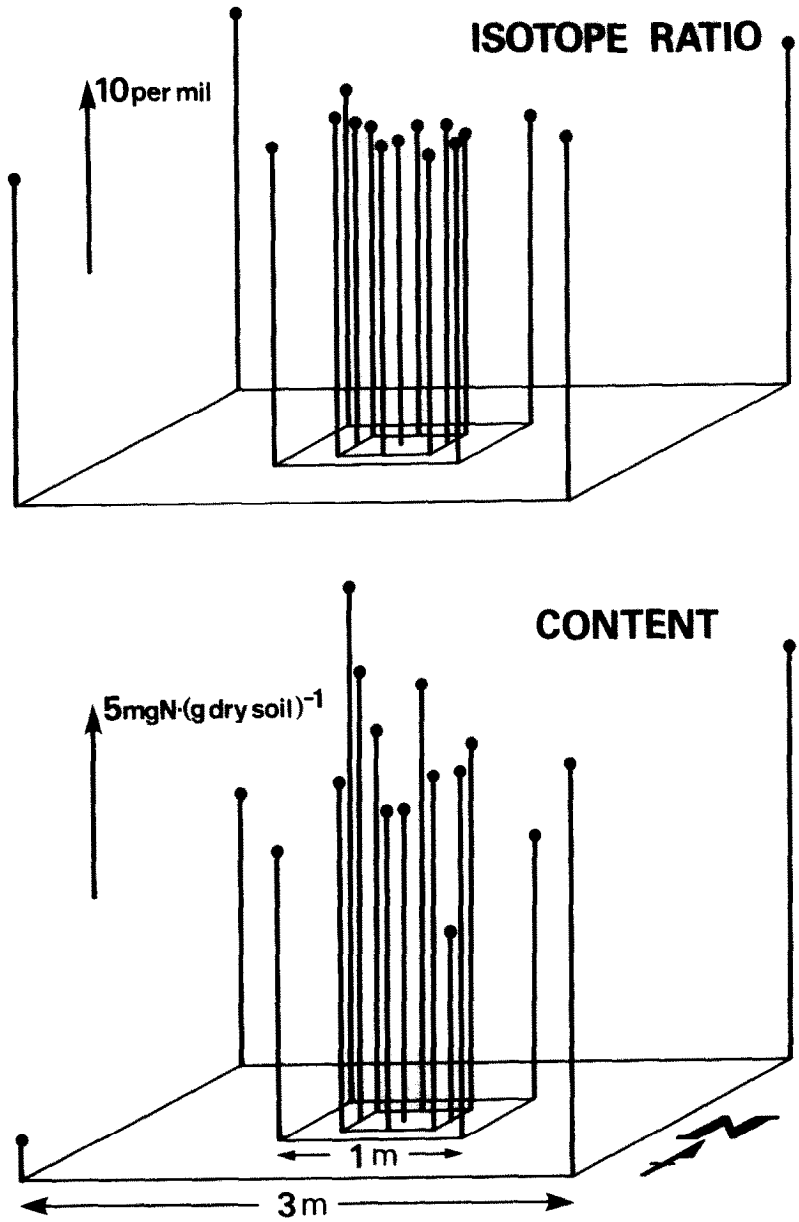

Figure 1. Variation of Kjeldahl nitrogen content and its nitrogen isotope ratio of soils from Fumishima rookery. Surface soil (0–5 cm) was collected by a stainless-steel core sampler (I.D. 20.5 mm). The upper half shows the variation of nitrogen isotope ratio and the lower half that of Kjeldahl nitrogen content. The full length of the arrow represents the value given next to it. The plane of the three flat squares is the origin; i.e., zero per mil for the isotope ratio and zero for the content.

Table 1. Kjeldahl nitrogen content and its isotope ratio of surface soils

Location	Breeding seabirds	Number of studied sites	Content (mg N · (g dry soil)$^{-1}$)			Isotope ratio (per mil)		
			No.	Mean ± S.D.	Range	No.	Mean ± S.D.	Range
Inside rookery								
Kabushima	gull	9	33	12.4 ± 9.2	0.8–30.6	32	19.0 ± 2.7	13.6–24.8
Tsubakishima	gull and heron	2	3	12.3 ± 2.4	10.7–15.0	2[c]	16.3 ± 0.3	16.1–16.5
Fumishima	gull	2	19	9.4 ± 3.2	1.1–15.4	18	17.2 ± 1.2	15.8–19.8
Bentenshima	gull	2	4	18.7 ± 3.2	16.0–23.2	4	18.3 ± 1.1	17.3–19.6
Yunotsu	gull	4	4	4.7 ± 3.5	1.9–9.6	4	11.7 ± 2.9	9.1–15.5
Torishima[a]	albatrosses	3	8	0.43 ± 0.21	0.10–0.73	7	17.0 ± 2.7	13.3–21.2
Cape Bird	penguin	5	6	27.2 ± 10.8	12.7–44.2	6	32.2 ± 3.5	27.4–37.9
Intermediate area[b]								
Kabushima	none	1	7	4.7 ± 0.9	3.5–5.8	6	10.8 ± 0.5	10.0–11.3
Aomatsushima	none	1	2	1.1 ± 0.1	1.0–1.1	2	14.1 ± 8.1	8.3–19.8
Cape Bird	none	1	1	0.11	–	1	12.2	–
Outside rookery								
Kabushima	none	1	3	1.5 ± 1.1	0.72–2.8	2	7.3 ± 1.8	6.0–8.6
Fumishima	none	1	1	0.91	–	1	8.6	–
Bentenshima	none	1	2	0.41 ± 0.00	0.41–0.41	2	7.0 ± 0.2	6.8–7.1
Yunotsu	none	2	2	0.13 ± 0.04	0.10–0.16	2	−4.4 ± 0.4	−4.6–4.1
Torishima	none	5	17	0.05 ± 0.08	0.006–0.31	2[c]	8.0 ± 0.5	7.6–8.3
Cape Bird	none	1	1	0.004	–	0	–	–

[a] Soils are from nesting areas of the Short-tailed Albatross.
[b] Intermediate area is the area where the seabirds are sighted to casually come nearby and on which some influence of the breeding activity appears quite likely.
[c] Samples from one site were analyzed.

Table 2. Ammonium nitrogen content and its isotope ratio of surface soils

Location	Breeding season	Ammonium content (mg N · (g dry sample)$^{-1}$)				Isotope ratio (per mil)			
		No. of sites	No. of samples	Mean ± S.D.	Range	No. of sites	No. of samples	Mean ± S.D.	Range
Inside rookery									
Kabushima	IN	4	42	1.5 ± 1.7	0.24–7.5	3	18	31.1 ± 8.4	13.9–44.8
Tsubakishima	IN	2	3	0.87 ± 0.13	0.75–1.0	1	2	26.8 ± 3.0	24.7–28.9
Bentenshima	IN	2	4	0.77 ± 0.40	0.34–1.3	2	4	26.8 ± 14.5	13.9–40.0
Yunotsu	IN	4	4	0.54 ± 0.34	0.21–0.92	4	4	21.7 ± 8.3	10.7–29.1
Torishima[a]	IN	3	3	0.032 ± 0.037	0.006–0.074	1	2	8.3 ± 3.0	6.2–10.4
Cape Bird	IN	2	2	6.5 ± 1.1	5.7–7.2	2	2	44.5 ± 7.2	39.4–49.6
Kabushima	OFF	1	4	0.22 ± 0.08	0.11–0.28	0	0	–	–
Fumishima	OFF	2	4	0.10 ± 0.12	0.025–0.27	1	3	7.1 ± 3.2	3.9–10.3
Intermediate area									
Aomatsushima	IN	1	2	0.12 ± 0.03	0.095–0.14	1	2	11.4 ± 8.0	5.7–17.0
Cape Bird	IN	1	1	0.034	–	1	1	16.8	–
Outside rookery									
Kabushima	IN	1	1	0.030	–	0	0	–	–
Bentenshima	IN	1	2	0.029 ± 0.001	0.028–0.030	1	2	6.4 ± 0.4	6.1–6.7
Yunotsu	IN	2	2	0.032 ± 0.018	0.019–0.045	2	2	−0.3 ± 1.3	−1.2–0.6
Cape Bird	IN	1	1	0.0040	–	0	0	–	–

[a]Soils are from nesting areas of the Short-tailed Albatross.

paper. No considerations were paid to the difference in site and in sampling date.

The generally high value is found for the isotope ratio of Kjeldahl nitrogen in all of the seabird rookeries (Table 1). The ratio for the soil outside the five rookeries (Kabushima, Fumishima, Bentenshima, and Yunotsu of gulls, and Torishima of albatrosses) averages 5.3 per mil, whereas the average ratio of the soil inside the same five rookeries is 16.6 per mil. This ^{15}N enrichment of the rookery soil becomes more significant, if one recalls that the range of the nitrogen isotope ratio for various soils reported by others is mostly from -5 to 14 per mil (Sweeney et al., 1978; Mizutani et al., 1985b). The rookery soils, therefore, appear exceptionally enriched in ^{15}N.

Another point of interest in Table 1 is that all the five gull rookeries and the one albatross rookery gave similar isotope ratios, though the nitrogen content at the albatross rookery was nearly two orders of magnitude lower than that at the gull rookeries. In contrast with these rookeries, the rookery in Cape Bird exhibited a much higher ratio. This uniquely higher ratio for Kjeldahl nitrogen of the soils of the penguin rookery may suggest that the climate is responsible for the observation. The five gull rookeries and the one albatross rookery are located at about middle latitude (30 °N to 41 °N), whereas the penguin rookery is at a high latitude (77 °S) In this context, it would be interesting to study the ratio of soils from a rookery near the equator. This aspect of ^{15}N enrichment will be further discussed later under a separate subheading.

Though it varies, the ammonium content in soils inside rookeries is, on the average, higher than that in the other soils (Table 2). In Cape Bird, the content changes from 6.5 (inside rookery) to 0.0040 mg N · (g dry soil)$^{-1}$ (outside rookery). The same trend is found for all the other rookeries where such a comparison can be made, i.e., Kabushima, Tsubakishima-Aomatsushima, Bentenshima, and Yuonotsu. Furthermore, as to be shown later in Table 4, the ammonium content is the highest at the soil surface. These observations should support the presence of an *in situ* aerobic decomposition of organic matters deposited onto the rookery and the eventual production of ammonium.

A comparison between the isotope ratio for Kjeldahl nitrogen and that for ammonium nitrogen reveals an interesting nature of the nitrogen dynamics in seabird rookeries. The isotope ratio of ammonium nitrogen in soils of four gull rookeries (Kabushima, Tsubakishima, Bentenshima, and Yunotsu) was about 10 per mil higher than that for the corresponding Kjeldahl nitrogens. A similar extent of the difference was also found for the penguin rookery soils. The soils from Torishima rookery and Fumishima rookery, however, show an apparent opposite: the isotope ratio for the ammonium nitrogens is about 10 per mil lower than that for the corresponding Kjeldahl nitrogens. The difference appears to reflect the difference in the source of ammonium nitrogen.

The Fumishima rookery soils were sampled outside the breeding season and the ammonium nitrogen content is nearly one order of magnitude lower than that found in other gull rookeries during the breeding season. This probably indicates that the rate of ammonium consumption is rapid in Fumihsima, and a considerable portion of the ammonium found in its soils are of precipitational origin. Nakai (1962) noted that a dense vegetation quickly covers Fumishima rookery after the breeding season, whereas no plants exist while the gulls are present. The Fumishima soils were sampled in October, and the plants had already withered. The rapidly growing plants in Fumishima rookery must have taken up a considerable portion of the ammonium of avian excretal origin, and it may be one of the causes for the observation. Torishima rookery soils also contained a very small amount of ammonium nitrogen, though they were sampled during the breeding season of the albatrosses. As noted in this paper and elsewhere (Mizutani and Wada, 1985b), the soils of the Torishima rookery is coarse, volcanic ash, and their water permeability is very large. The rookery of the Short-tailed Albatross is located at the upper portion of the slope. Therefore, it is likely that the ash soils are effectively washed with rainwater. This would have resulted in the light isotope ratio for ammonium nitrogen even during the breeding season.

A decrease in the ammonium content during a non-breeding season is found at Kabushima, and this would indicate an exisitence of a certain process through which ammonium is lost from the soil. A sudden, rapid growth of vegetation after the breeding season, however, is not known in Kabushima rookery. The high $\delta^{15}N$ for Kjeldahl nitrogen of the rookery soils and still higher ratio for the ammonium nitrogen must inidate that a major cause for the high ratios is the large fractionation factor associated with the volatilization of ammonia. A significant portion of the ammonium loss must have been caused by volatilization of ammonia. It will be discussed further under the following subheading.

Food chain effect and ammonia volatilization

The high isotope ratio for both Kjeldahl and ammonium nitrogens in soils of seabird rookeries must result from a combination of various factors that govern the distribution of nitrogen isotopes in nature. Among these factors, two appear to be of prominent importance. They are: 1) Relatively high nitrogen isotope ratio for incoming nitrogen to a seabird rookery due to the ^{15}N enrichment along a food chain, and 2) A large fractionation factor during an escape of nitrogen through ammonia volatilization.

It is known that an enrichment of ^{15}N takes place along a food chain (Miyake and Wada, 1967; Minagawa and Wada, 1984). As seabirds feed mostly on fish and squids, they are in a sense at the top end of a food chain in an oceanic ecosystem. It, therefore, results in a relatively high ratio for nitrogenous matters that enter the rookery.

Common sources of nitrogen to a terrestrial ecosystem other than the direct inflow of organic nitrogen are nitrate and ammonium in precipitation and *in situ* nitrogen fixation. These forms of incoming nitrogen normally give an isotope ratio of about zero per mil or less (Wada et al., 1975; Peters et al., 1978). The majority of nitrogen in penguin excreta is in the form of uric acid, and the uric acid in penguin excreta collected at the rookery in Cape Bird had the ratio of 6.2 per mil (Mizutani et al., 1985b) and that in gull excreta sampled at Kabushima rookery 9.1 per mil (Mizutani et al., 1985a).

Furthermore, bodies of seabirds are another source of incoming nitrogen to the rookeries, and they have still higher ratio. This is expected because of the enrichment of ^{15}N along a food chain, and, in fact, various feathers of the gull collected at Kabushima and Fumishima rookeries gave as their nitrogen isotope ratio 12.4 per mil with the standard deviation of 0.4 per mil (9 analyses). A feather of another gull species (*Larus schistisagus*) collected in Hokkaido showed the ratio of 15.2 per mil. The ratio for five feathers of Common Cormorants (*Pharacrocorax carbo*) sampled once nearly every two months from 1984 to 1985 in Tokyo averaged 14.4 per mil with the standard deviation of 1.8 per mil. Two feathers of the Short-tailed Albatross obtained at Torishima each on a different visit yielded 15.7 per mil and 15.2 per mil. The rather large standard deviation of the isotope ratio for the cormorant feathers may be caused by the fact that the cormorants go for their food to coastal areas during summer and to rivers during winter (Fukuda, 1985).

Table 3 shows the nitrogen content and its isotope ratio for various organs of the penguin chick and the juvenile in Cape Bird. In case of the chick, the isotope ratio for its organs other than the tail feather averaged 9.4 per mil with the standard deviation of 0.5 per mil. The organs of the juvenile other than the body feather gave the mean of 11.5 per mil with the standard deviation of 1.2 per mil. In the both two instances, feathers showed the ratio slightly lower than the average. Hence, the ratio of the bodies of seabirds may be at least as high as that for feathers.

The fractionation during the evaporation of ammonia that was produced by the decomposition of organic matters further enriches in ^{15}N the remaining nitrogen in soil (Kirshenbaum et al., 1947). Kreitler (1975) reported that there was a 38 per mil difference in the isotope ratio between the evolved ammonia from the remaining inorganic nitrogen in barnyard and feedlot soils that had been soaked in animal urine, and a study on soils from the Kabushima rookery gave a similar fractionation (Mizutani et al., 1985a). Therefore, the volatilization of ammonia from rookery soils must contribute largely to the enrichment of ^{15}N for remaining nitrogen in rookery soils, if the volatilization process accounts for a significant portion of ammonium loss from the rookeries.

A laboratory experiment on rookery soils of a King Penguin (*Aptenodytes patagonicus*) rookery and of a Macaroni Penguin (*Eudyptes chrysolophus*) rookery in Marion Island (46°54′S, 37°44′E) showed that nitrification and

Table 3. Kjeldahl nitrogen content and its isotope ratio of various organs of a chick and a juvenile of the Adelie Penguin

Organ	Kjeldahl nitrogen	
	Content (mg N · (g dry sample)$^{-1}$)	Isotope ratio (per mil)
Chick		
Feather, tail	111	8.8
Spine with surrounding muscle	104	9.7
Muscle, heart	100	9.6
Muscle, shoulder	88	9.5
Muscle, thigh	150	8.7
Juvenile		
Feather, body	n.d.	10.8
Skin, abdomen	128	11.5
Skin, tarsus	121	10.9
Bone, tarsus	37	9.5
Toe	104	12.3
Muscle, back	109	13.0
Brain	n.d.	11.8

n.d.: not determined.
Dietary differences might cause the generally lower isotopic ratios observed in chick organs relative to juvenile organs. Tarsus bone of the juvenile may retain the ratio as it was when the bird was a chick.

denitrification are relatively unimportant processes, and that the degradation of uric acid into ammonia and its subsequent volatilization are quantitatively the most important processes (Lindeboom, 1984). And, in fact, Smith (1978) wrote about the distinct smell of ammonia above a penguin rookery in Marion Island.

Although the extent of ammonia volatilization may depend on many factors, chemical analysis on soils from Kabushima rookery and from Tsubakishima rookery showed the aerobic decomposition of uric acid and the resultant formation of ammonium (Mizutani and Wada, 1985a), and a laboratory experiment with Kabushima rookery soils demonstrated the ammonia evaporation from the soil (Mizutani et al., 1985a). The nitrification of ammonia must also take place in the gull and the albatross rookeries that are located in a middle latitudinal region. Indeed, our unpublished laboratory study on the decomposition of uric acid in gull rookery soils shows that the soil ammonium is lost through the nitrification as well as the volatilization, though the most nitrogen is lost from the soil through ammonia volatilization.

In the field conditions, the contribution of the processes of nitrification, leaching, and denitrification to the ^{15}N enrichment seems small. The decomposition of uric acid must be mostly aerobic and the production of ammonia occurs at the surface of the soil (Mizutani and Wada, 1985a). The distinct smell of ammonia prevails in the air over the gull rookeries too. Table 4 gives the vertical profile of Kjeldahl, ammonium, and uric acid nitrogen contents and their isotope ratios found in the Tsubakishima rookery soil. The high content of and the high isotope ratio for ammonium nitrogen of the surface soil in the top 5 cm layer in contrast with the low values for the other layers clearly demonstrate that the ammonia volatilization is the predominant process that accounts for the loss of ammonium from the soil. The examination of the top 1 cm of the same soil sample yielded a still higher ratio of 39.4 per mil for the ammonium nitrogen. In view of these facts, the volatilization must, indeed, contribute significantly to the observed enrichment.

Table 4 also shows other points of interest. Among them are 1) an apparent, relative constancy of the ratio of Kjeldahl nitrogen, 2) a similar constancy of the ratio for ammonium nitrogen in the soils other than the top 5 cm layer, and 3) a depletion of ^{15}N in the ammonium nitrogen of the lower layers relative to the coexisting Kjeldahl nitrogen. The relative constancy of the ratio for Kjeldahl nitrogen was also observed in soils from Kabushima rookery. And the same sort of the depletion of ^{15}N was also found in case of Torishima and Fumishima rookery soils. Tsubakishima rookery is densely covered with vegetation during the breeding season, and the rapid loss of ammonium by plant uptake seems likely. It may, therefore, prevent the ammonium from penetrating down into the soil. Even if it is the case, however, the source of the isotopically light ammonium nitrogen in the lower layers must be identified; unlike cases of Torishima and Fumishima,

Table 4. Vertical distribution of Kjeldahl nitrogen, ammonium nitrogen, and uric acid nitrogen, and their isotope ratio in soils of Tsubakishima rookery

Depth (cm)	Kjeldahl nitrogen		Ammonium nitrogen		Uric acid nitrogen	
	Content	Isotope ratio (per mil)	Content	Isotope ratio (per mil)	Content	Isotope ratio (per mil)
0–5	15.0	16.5	1.0	28.9	2.1	8.9
5–10	4.2	14.7	0.20	5.9	0	–
10–15	8.7	16.8	0.22	5.9	0	–
15–20	3.5	16.1	0.21	5.4	0	–

Nitrogen content is expressed in unit of mg N · (g dry soil)$^{-1}$.

isotopically light nitrogen in rainwater is unlikely to be responsible for the observations. Instead, a regeneration of ammonium from soil organic matters might be playing a significant role in the lower layers. Whatever the case, a further study of these observations would reveal an interesting aspect on the nature of nitrogen dynamics along the vertical axis.

A combination of slow nitrification and rapid decomposition of avian organic matters must prolong the residence time of ammonium in soil, rendering a longer period of time available for ammonia volatilization and thus causing $\delta^{15}N$ values for the remaining ammonium soil to be elevated. The resultant ^{15}N-enriched ammonium must, then, be incorporated into soil organic matters, since not only ammonium in surface soil (Table 2) but also soil organic matters (Table 1) possess a much higher nitrogen isotope ratio than the incoming nitrogen to the rookery. At first sight, then, one may expect a negative correlation between the Kjeldahl nitrogen content and its isotope ratio. This is because a smaller Kjeldahl nitrogen content may reflect a larger extent of ammonia volatilization, which must be responsible for the enrichment of ^{15}N in the remaining nitrogen in the soil. However, it was not the case found from the rookery soils. For instance, the 17 pairs of data on the nitrogen content and the isotope ratio that were used to construct Figure 1 gives the correlation coefficient of 0.059. Possible factors that work against the first sight may include a local heterogeneity of the amount of nitrogen input and of the rate of nitrification. The observed enrichment of ^{15}N in Kjeldahl nitrogen of rookery soils must be an accumulated result of effects of these and other factors for a certain period. In fact, two soil samples from the relatively new Yunotsu rookery gave as the nitrogen isotope ratio of Kjeldahl nitrogen the mean of 11.7 per mil that are distinctly lighter than those found from other rookeries.

Because the ratio observed for rookery soils is extraordinarily high comparing with that normally found for a soil, it may be used as a tool to locate an abandoned rookery. Ishizuka (1966) reported that, about 60 years after its abandonment, the content of water-soluble ammonium in soils from a former rookery was more than six times of that from nearby places where no record of a past avian colonization was known. If it is the case, the isotope ratio of ammonium or Kjeldahl nitrogen of a soil may reveal the past avian activity for even a longer period of time after its abandonment as a seabird rookery, for the present study showed that the exceptionally high ratio is characteristic of seabird rookeries and that the ratio reflects events occuring during a longer period of time than the ammonium content.

Factors that determine the extent of ^{15}N enrichment

As the volatilization of ammonia seems responsible for the enrichment of ^{15}N in the soils, the extent of the enrichment may in turn reflect the extent of the volatilization. Because most incoming nitrogenous substances to a bird rookery must be in the form of avian excreta, and because uric acid accounts for the majority of nitrogen in the excreta and has almost the same nitrogen isotope ratio to that of the whole excreta (Mizutani et al., 1985a and 1985b),

nitrogen isotope ratio for uric acid in excreta may be roughly regarded as the representative isotope ratio for the incoming nitrogen. The overall increase in $\delta^{15}N$ of Kjeldahl nitrogen in soil would be, therefore, 9.9 per mil for Kabushima rookery (soil: 19.0 per mil, uric acid: 9.1 per mil) and 26.0 per mil for the penguin rookery in Cape Bird (soil: 32.2 per mil, uric acid: 6.2 per mil). The extent in the penguin rookery seems about two and a half times as large as that for the gull rookery. As discussed earlier, the apparent relationship between the extent of the enrichment of ^{15}N and the latitude may be at least in part due to a latitudinal difference in rates of soil microbial nitrification and denitrification.

More ammonium is lost in Antarctica through the volatilization partly because the generally cold climate there reduces the rate of nitrification by soil microbiota and allows the volatilization to play a larger role. However, there may be another factor that determines the actual extent of the volatilization. The process of ammonia volatilization is known to occur also for other ecosystems. They are bat guano systems (Poulson, 1972), barnyard and feedlot soils (Kreitler, 1975), prairie grasslands (Woodmansee, 1978), and agricultural areas (Denmead et al., 1974 and 1976). As the nitrogen loss from soil through the ammonia evaporation carries an important implication in terms of fertilizer management to agricultural sciences, there are numerous studies on the extent of the loss and the factors that affect its magnitude (e.g., Faurie and Bardin, 1979; Woodmansee, 1979).

A recent study showed that an adequate moisture in the topsoil is one of the factors that contribute most to ammonia volatilization in a semiarid environment (Bouwmeester et al., 1985). It also indicated that a rapid drying of the surface soil reduced the loss. However, the present study showed that the generally dry condition in Antarctica appeared ineffective in retarding the volatilization of ammonia. On the contrary, the process of ammonia volatilization seemed more important in Antarctica than in middle latitudinal regions. As it is likely that a completely dry condition lets ammonium tightly adsorb on soil, consequently reducing the loss of ammonium nitrogen through the volatilization, the results from soils of the penguin rookery must indicate that a certain balance between wet and dry conditions is most effective in volatilizing ammonium in soil and, consequently, in enhancing the nitrogen isotope ratio for soil ammonium and Kjeldahl nitrogens.

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Appendix

Content and isotope ratio of Kjeldahl and ammonium nitrogens in individual surface soil samples (0–5 cm)

Location	Studied site	Date of sampling	Kjeldahl nitrogen		Ammonium nitrogen	
			Content (mg N · (g dry soil)$^{-1}$)	Isotope ratio (per mil)	Content (mg N · (g dry soil)$^{-1}$)	Isotope ratio (per mil)
Inside rookery						
Kabushima						
	site 1	March 1, 1980	18.3	16.9	–	–
	site 2	March 1, 1980	13.6	17.1	–	–
	site 3	March 1, 1980	17.1	17.2	0.58	13.9
			13.2	19.3	–	–
		May 14, 1980	21.1	18.0	–	–
		June 9, 1980	4.8	18.7	–	–
		July 21, 1980	18.4	18.8	–	–
			22.6	17.6	–	–
		July 24, 1981	28.6	–	1.9	–
			16.5	21.2	2.8	33.6
			15.2	21.3	0.90	40.9
			16.6	18.5	0.47	42.0
			12.6	15.8	0.24	22.2
		Sept. 11, 1981	–	–	0.11	–
		Jan. 21, 1982	–	–	0.21	–
			–	–	0.27	–
			–	–	0.28	–
		June 20, 1983	30.6	20.6	6.5	34.6
			23.6	21.0	5.6	32.6
			27.2	19.2	4.6	26.6
			27.2	19.3	4.7	25.1
	site 4	March 1, 1980	21.6	16.6	–	–
	site 5	March 1, 1980	1.4	15.5	–	–
			0.80	13.6		

	June 9, 1980	5.3	23.5	2.5	32.3
		3.5	19.2	2.0	23.8
		3.8	19.0	2.2	23.6
site 7	June 9, 1980	3.3	15.2	–	–
site 8	June 9, 1980	6.9	19.0	1.4	28.4
		3.1	20.4	0.91	28.5
		4.6	19.8	1.1	26.0
	July 21, 1980	7.3	23.6	1.5	40.8
		3.1	24.6	0.70	40.7
		4.7	24.8	0.87	44.8
site 10	May 13, 1981	–	–	0.87	–
		–	–	0.85	–
		–	–	1.1	–
		–	–	0.85	–
		–	–	0.69	–
		–	–	0.70	–
		–	–	0.56	–
		–	–	0.58	–
		–	–	0.87	–
		–	–	0.92	–
		–	–	0.45	–
		–	–	0.44	–
		–	–	0.68	–
		–	–	0.66	–
		–	–	0.50	–
		–	–	0.43	–
		-	–	0.73	–
		–	–	0.52	–
		–	–	0.74	–
		–	–	0.39	–
		–	–	7.5	–
		–	–	0.74	–
		–	–	0.90	–

Location	Studied site	Date of sampling	Kjeldahl nitrogen		Ammonium nitrogen	
			Content (mg N · (g dry soil)$^{-1}$)	Isotope ratio (per mil)	Content (mg N · (g dry soil)$^{-1}$)	Isotope ratio (per mil)
Kabushima	site 11	June 9, 1980	4.9	17.7	–	–
		July 21, 1980	3.3	16.5	–	–
			3.5	16.9	–	–
Tsubakishima						
	site 1	June 5, 1982	15.0	16.5	1.0	28.9
			11.2	16.1	0.87	24.7
	site 6	June 5, 1982	10.7	–	0.75	–
Fumishima						
	site 1	Oct. 4, 1980	8.2	16.1	–	–
			11.2	16.4	–	–
			10.0	16.4	–	–
			11.8	17.1	–	–
			9.1	17.9	–	–
			8.4	16.2	–	–
			9.3	15.8	–	–
			4.9	17.0	–	–
			9.6	15.9	–	–
			6.9	16.2	–	–
			13.5	17.8	–	–
			7.6	16.8	–	–
			9.6	16.9	–	–
			10.7	17.9	–	–
			7.1	19.8	0.050	10.3
			1.1	17.2	0.025	3.9
			10.8	19.2	0.046	7.2
	site 2	Oct. 4, 1980	15.4	18.8	–	–
		July 29, 1981	12.6	–	0.27	–

Bentenshima						
	site 1	May 14, 1981	18.7	17.3	0.34	13.9
			23.2	17.5	0.71	14.6
	site 2	May 14, 1981	16.9	18.6	0.73	38.8
			16.0	19.6	1.3	40.0
Yunotsu						
	site 1	June 8, 1984	4.8	12.3	0.30	10.7
	site 2	June 8, 1984	2.4	15.5	0.72	27.0
	site 3	June 8, 1984	9.6	9.7	0.92	29.1
	site 4	June 8, 1984	1.9	9.1	0.21	20.0
Torishima						
	site 3	March 25, 1982	0.23	17.1	–	–
			0.73	16.7	–	–
		June 22, 1982	0.37	17.1	0.006	–
			0.38	13.3	–	–
	site 9	Nov. 17, 1982	0.42	14.4	–	–
			0.10	–	–	–
	site 10	Nov. 17, 1982	0.69	21.2	0.074	6.2
			0.52	19.2	0.016	10.4
Cape Bird						
	site 1	Jan. 6, 1981	32.8	37.9	–	–
	site 2	Jan. 6, 1981	24.7	27.4	–	–
	site 3	Jan. 6, 1981	12.7	30.2	–	–
	site 11	Jan. 6, 1981	44.2	31.7	–	–
			20.4*	33.7	7.2	39.4
	site 12	Jan. 6, 1981	28.5*	32.1	5.7	49.6

Location	Studied site	Date of sampling	Kjeldahl nitrogen		Ammonium nitrogen	
			Content (mg N · (g dry soil)$^{-1}$)	Isotope ratio (per mil)	Content (mg N · (g dry soil)$^{-1}$	Isotope ratio (per mil)
Intermediate area						
Kabushima						
	site 6	March 1, 1980	5.4	–	–	–
			3.5	10.0	–	–
			5.5	10.7	–	–
		June 9, 1980	4.4	10.3	–	–
			3.9	11.2	–	–
		July 21, 1980	5.8	11.3	–	–
			4.5	11.0	–	–
Aomatsushima						
	site 4	May 16, 1981	1.0	8.3	0.095	5.7
			1.1	19.8	0.14	17.0
Cape Bird						
	site 50	Jan. 6, 1981	0.11*	12.2	0.034	16.8
Outside Rookery						
Kabushima						
	site 100	June 9, 1980	0.95	8.6	–	–
			0.72	6.0	–	–
		July 24, 1981	2.8	–	0.030	–
Fumishima						
	site 100	Oct. 4, 1980	0.91	8.6	–	–
Bentenshima						
	site 100	May 14, 1981	0.41	7.1	0.030	6.7
			0.41	6.8	0.028	6.1
Yunotsu						
	site 100	June 9, 1984	0.098	−4.1	0.045	−1.2
	site 101	June 9, 1984	0.16	−4.6	0.019	0.6

Torishima						
	site 1	June 22, 1982	0.036	–	–	–
			0.032	–	–	–
			0.045	–	–	–
		Nov. 17, 1982	0.31	7.62	–	–
			0.21	8.29	–	–
	site 4	June 22, 1982	0.032	–	–	–
			0.017	–	–	–
Torishima						
	site 4	Nov. 17, 1982	0.020	–	–	–
			0.012	–	–	–
	site 5	June 22, 1982	0.023	–	–	–
			0.023	–	–	–
	site 6	June 22, 1982	0.017	–	–	–
			0.019	–	–	–
		Nov. 17, 1982	0.013	–	–	–
			0.006	–	–	–
	site 7	June 22, 1982	0.016	–	–	–
			0.033	–	–	–
Cape Bird						
	site 100	Jan. 6, 1981	0.039*	–	0.004	–

A short, horizontal bar indicates that the measurement was not done. Data marked with an asterisk were published in Mizutani et al. (1985b). Although some values are the average of more than one analysis on the same sample, most are obtained by a single analysis.

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