SUPPORTING INFORMATION

Nitrate, nitrite, and ammonium contamination in $^{15}N_2$ gas

Materials and Methods

$^{15}N_2$ gas supplied by SI Science Co., Ltd was used during all of the cruises. We performed the following experiments to examine possible contamination of nitrate, nitrite, and ammonium in the $^{15}N_2$ gas (Dabundo et al., 2014).

Experiment 1: Ten 250 mL PC bottles were filled with aged subtropical surface water (ASSW), and were sealed with thermoplastic elastomer caps without headspaces. 2 mL 98+ at% $^{15}N_2$ gas (batch #11059) was added to five of these bottles and mixed well. All of the bottles were kept at room temperature for at least 12 h, and the water was subsampled to a 30 mL polypropylene tube.

Experiment 2: Degassed seawater was mixed from the ASSW using a Sterapore membrane unit (20M1500A: Mitsubishi Rayon Co., Ltd., Tokyo, Japan) (Shiozaki et al., 2015), and stored in three 1 L Tedlar bags without headspaces. A different batch number of $^{15}N_2$ gas (#11059 and #11143) was added to two of these bags. The $^{15}N_2$ gas was injected at a ratio of 10 mL per 1 L seawater, and dissolved completely using a ruler (Großkopf et al., 2012). The $^{15}N_2$-enriched and non-$^{15}N_2$-enriched seawater samples

were subsampled to five 30 mL polypropylene tubes from each Tedlar bags.

We determined the concentrations of nitrate, nitrite, and ammonium at the nanomolar levels using supersensitive colorimetric systems (Hashihama et al., 2009, 2015). The detection limits of nitrate, nitrite, and ammonium were 3, 2, and 4, respectively.

Results and Discussion

In both experiments, there were no significant differences between the control and $^{15}N_2$-added samples or among different batch numbers of $^{15}N_2$ gas, with one exception; the nitrite concentration in the $^{15}N_2$ (#11143)-added samples was significantly lower than that of the control in Experiment 2 (Fig. S1a and b). Because we did not determine the $^{15}N/^{14}N$ ratios of nitrate, nitrite, and ammonium, the contamination at isotope level was not evaluated. The contamination of nitrate, nitrite, and ammonium in the 250 ml of seawater with 2 ml $^{15}N_2$ gas was undetectable (<nM level) in experiment 1. During the cruise experiments, we added 2 ml $^{15}N_2$ gas into 4.5 L seawater, and hence, the contamination level would be one order lower than that in experiment 1, indicating that ^{15}N-labeled substrates in the seawater were at most 10^{-2} nM. When the substrate concentration in the seawater was 3 nM (the detection limit of our analysis), the

concentration of ^{15}N-labeled substrate would be <1/100 lower than that of seawater. In this case, the ^{15}N concentration would be too low to detect the uptake rate (Shiozaki et al., 2009).

Dabundo et al. (2014) found contamination of $^{15}N_2$ gas supplied from Sigma-Aldrich and from Cambridge Isotopes and suggested that the contamination could be caused by the production method of the $^{15}N_2$ gas. The $^{15}N_2$ gas of the two companies is produced by the catalytic oxidation of $^{15}NH_3$ gas with cupric oxide (Dabundo et al., 2014), whereas the SI Science $^{15}N_2$ gas is produced by oxidation of ^{15}N-labeled ammonium sulfate with potassium hypobromite (Nakane, 1963). Surplus potassium hypobromite is added to avoid ammonia and NO_x gas generation, and the $^{15}N_2$ gas is purified using a molecular sieve and liquid nitrogen. This different production process from that used by Sigma-Aldrich and Cambridge Isotopes probably explains why there is little contamination of nitrate, nitrite, and ammonium in the $^{15}N_2$ gas produced by SI Science.

REFERENCES

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Table S1 Locations and obtained data at each station

Station	Area	Longitude	Latitude	Date (LST)	start time (LST)**	*Trichodesmium** [filaments l^{-1}]	N_2 fixation* [nmolN L^{-1} d^{-1}]	Σ N_2 fixation [μmolN m^{-2} d^{-1}]	Temperature* [ºC]	MLD [m]	NO_3^-+NO_2^-* [nM]	PO_4^{3-}* [nM]	N/P ratio*	DFe* [nM]
T0601	East China Sea	129.1	30.9	10-Sep-06	11:30	6	2.18	n.a.	29.3	32	3	9	0.33	0.89
T0602	East China Sea	129.35	30.71	10-Sep-06	14:00	6	6.2	305	29.4	39	10	32	0.31	n.a.
T0606	Kuroshio	131.17	28.92	11-Sep-06	11:45	4	5.61	377	30.5	21	<3	19	0.16	0.6
T0622	East China Sea	127.31	30.39	13-Sep-06	12:00	0	6.35	306	29	40	21	12	1.75	0.83
T0625	East China Sea	127.76	30.39	14-Sep-06	8:45	0	1.41	77.2	28.7	40	19	11	1.73	n.a.
T0702	East China Sea	128.38	30.82	6-Sep-07	13:30	4	0.97	38.4	28.7	13	9	25	0.36	0.55
T0706	Kuroshio	130.97	28.72	7-Sep-07	9:30	68	1.05	88.1	29.4	27	9	15	0.60	0.36
T0711	Philippine Sea	133.17	26.25	8-Sep-07	9:30	16	0.86	67.3	29.5	26	10	<3	3.33	0.39
T0715	Philippine Sea	133.16	23.81	9-Sep-07	14:20	0	0.63	77.7	29.3	21	5	<3	1.67	0.8
T0718	Philippine Sea	130.22	24.44	10-Sep-07	9:10	8	0.29	29.9	29.5	23	10	36	0.28	0.35
T0723	Kuroshio	129.85	29.05	12-Sep-07	2:25	26	3.58	251	29.1	39	3	4	0.75	0.39
GW-1	East China Sea	128.61	33.84	23-Jul-07	5:45	0	0.54	29.5	25.1	17	42	17	2.47	n.a.
GN-3	East China Sea	128.57	33.38	23-Jul-07	15:10	12	14	n.a.	27.6	14	16	22	0.73	n.a.
NW-8	East China Sea	129.25	32.24	25-Jul-07	5:25	0	1.77	113	28.6	12	17	5	3.40	n.a.
B	East China Sea	127.85	32.25	25-Jul-07	20:15	0	5.15	171	28.6	17	32	5	6.40	n.a.
CK-10	East China Sea	127.5	31.75	27-Jul-07	6:15	194	28	418	29.4	13	17	16	1.06	n.a.
T0902	Miyako Islands	125.67	25.13	10-Sep-09	9:50	2	0.54	62.7	29.2	51	15	<3	5.00	0.19
T0903	Miyako Islands	125.43	25.12	10-Sep-09	n.a.	6	n.a.	n.a.	29.2	24	15	<3	5.00	0.35
T0904	Miyako Islands	125.2	25.07	10-Sep-09	16:25	2	0.64	58.9	29	48	374	23	16.3	0.39
T0905	Miyako Islands	125.42	24.93	10-Sep-09	17:35	102	4.37	n.a.	29.7	31	6	4	1.50	0.2
T0906	Miyako Islands	125.73	25.19	10-Sep-09	23:50	>20000	62	753	29.6	60	3	14	0.21	0.25
T0907	Miyako Islands	125.57	24.67	11-Sep-09	11:10	2	1.66	49.1	29.2	27	53	9	5.89	0.89
T0908	Miyako Islands	125.61	24.92	11-Sep-09	14:05	10	1.04	105	29.4	40	4	5	0.80	0.4
T1001	East China Sea	129.28	30.66	4-Sep-10	9:45	6	3.69	70	29.1	28	21	9	2.33	n.a.
T1004	Kuroshio	126.9	28.4	9-Sep-10	7:15	72	3.94	79	28.6	22	19	20	0.95	n.a.
T1007	Miyako Islands	125.75	24.6	11-Sep-10	6:50	24	4.29	179	29.2	40	15	<3	5.00	n.a.

*values in the surface water

**start time of incubation for nitrogen fixation

n.a. = no data available

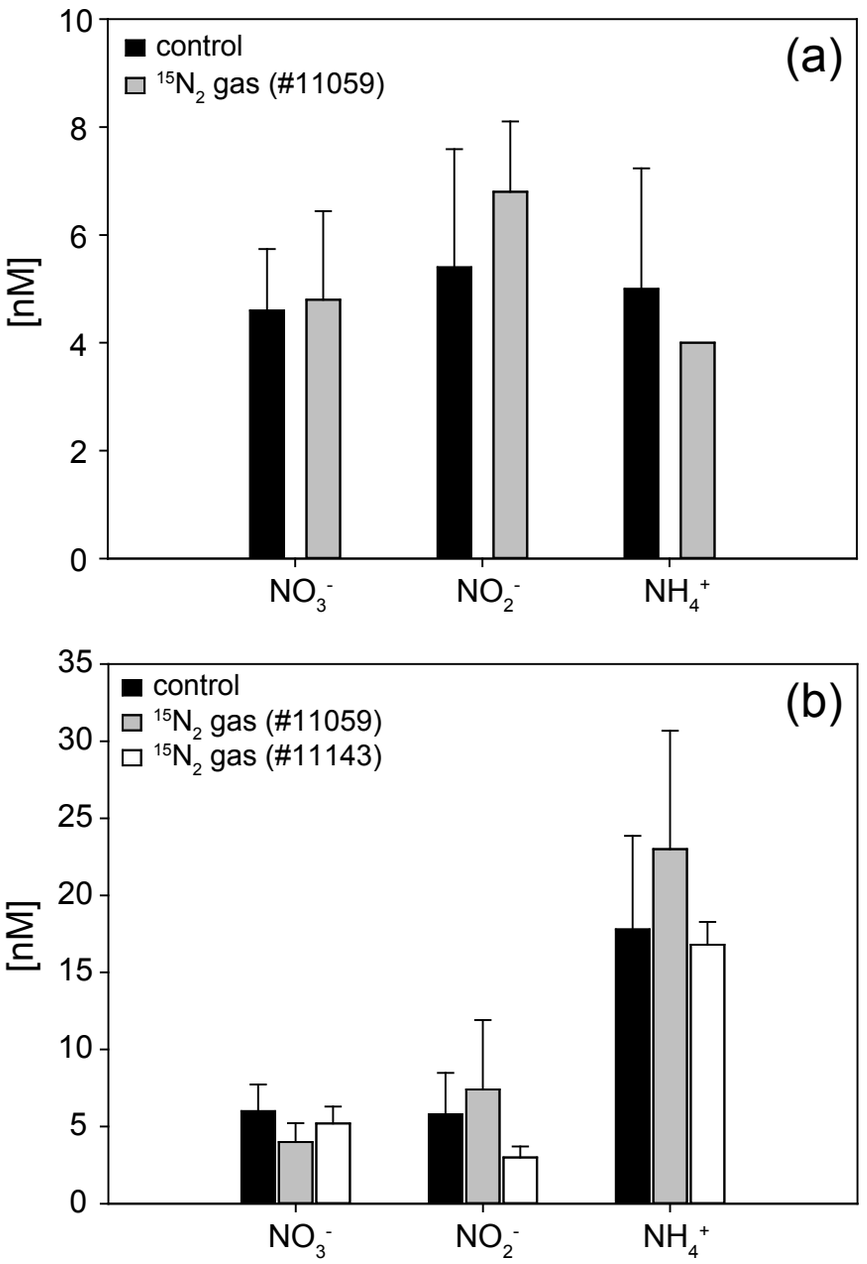

Fig. S1 Nitrate, nitrite, and ammonium concentration in control and $^{15}N_2$-gas added waters in experiment (a) 1 and (b) 2

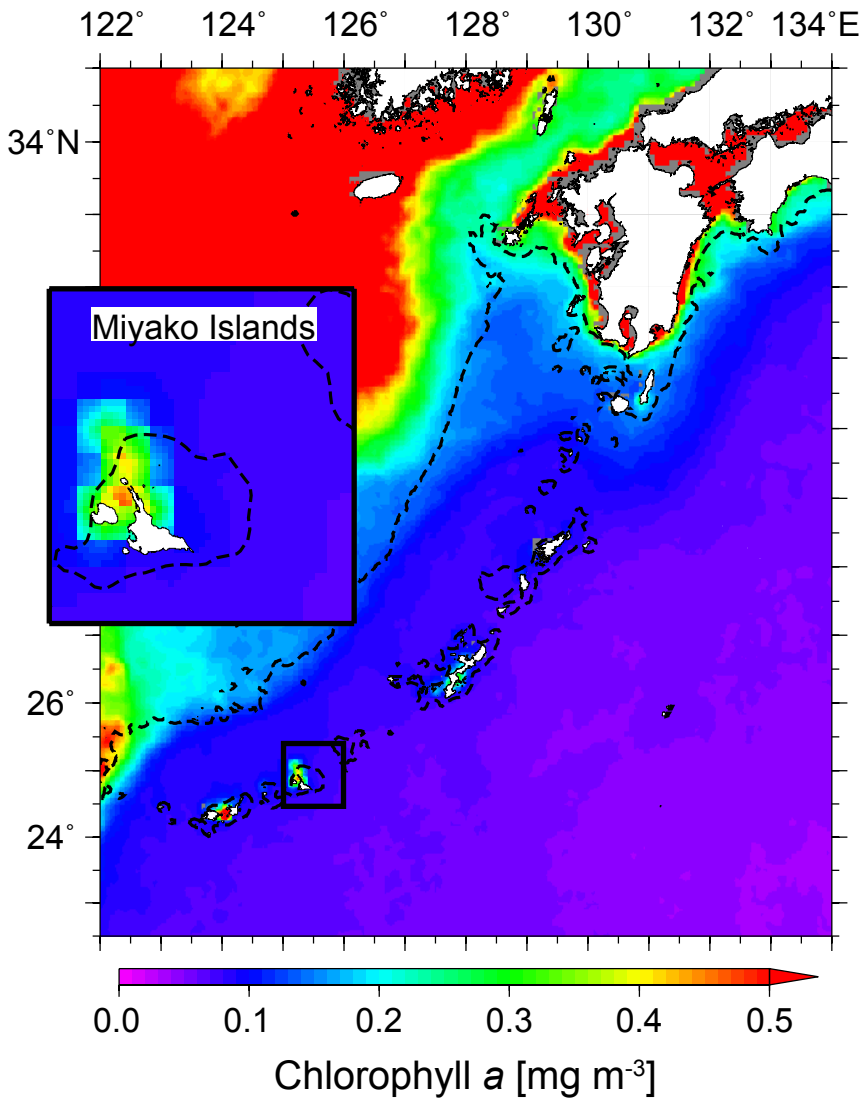

Fig. S2 Satellite-derived average chlorophyll *a* during the summer from July 2003 to September 2009. Dashed lines indicate 200 m isobaths.

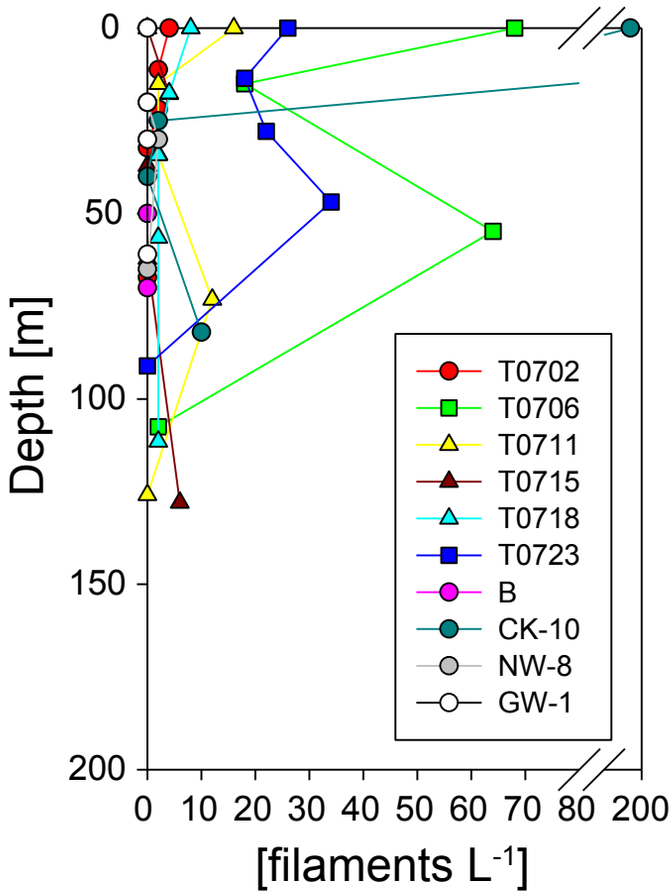

Fig. S3 Vertical profiles of *Trichodesmium* spp. during the *Nagasaki-maru* 242 and KT-0721 cruises

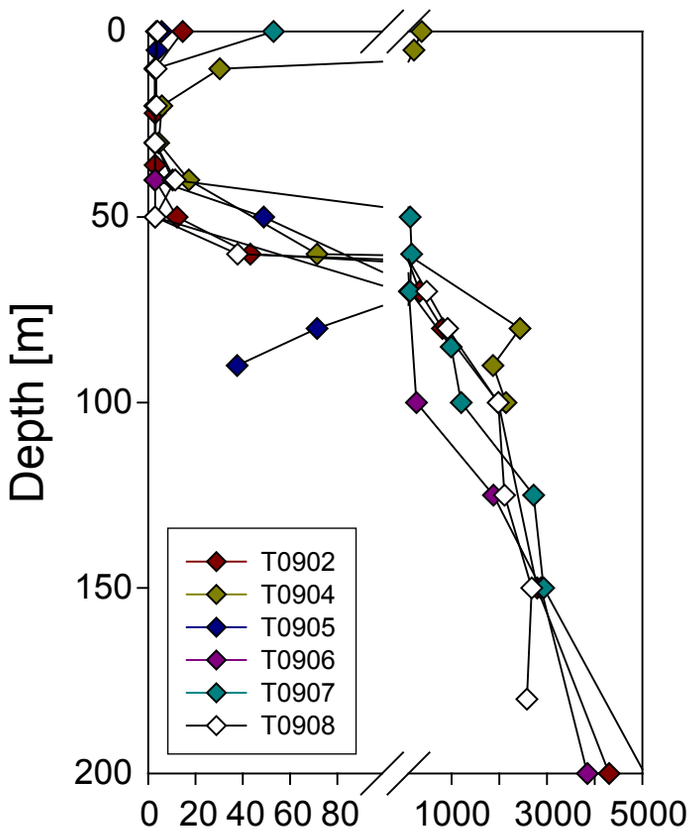

Fig. S4 Vertical profiles of nitrate during the KT-09-17 cruise

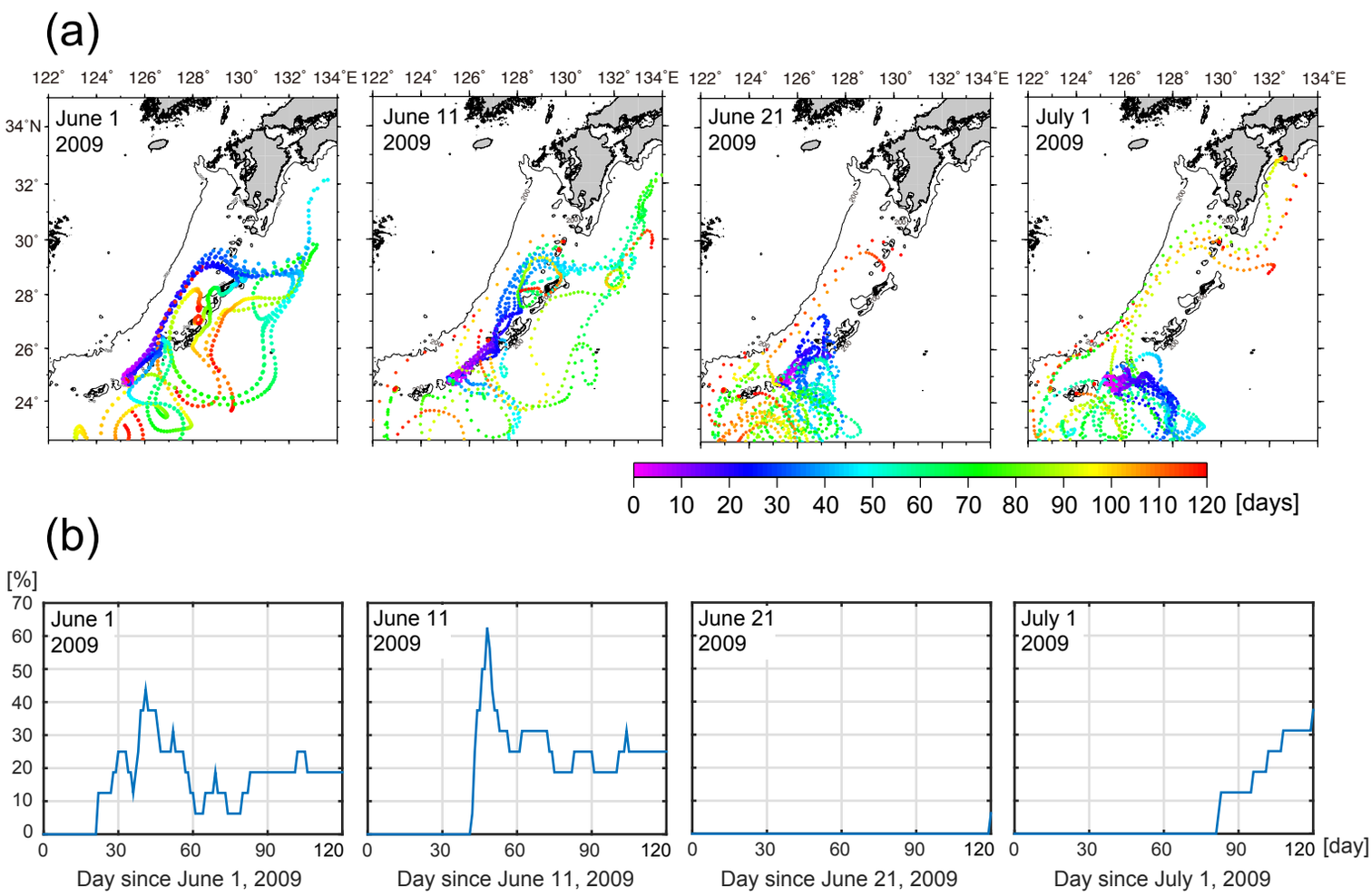

Fig. S5 (a) Particle trajectries released from points aroud the Miyako Islands on June 1, June 11, June 21, and July 1, 2009. (b) The ratio of particles delivered to Area K to the total released particles.

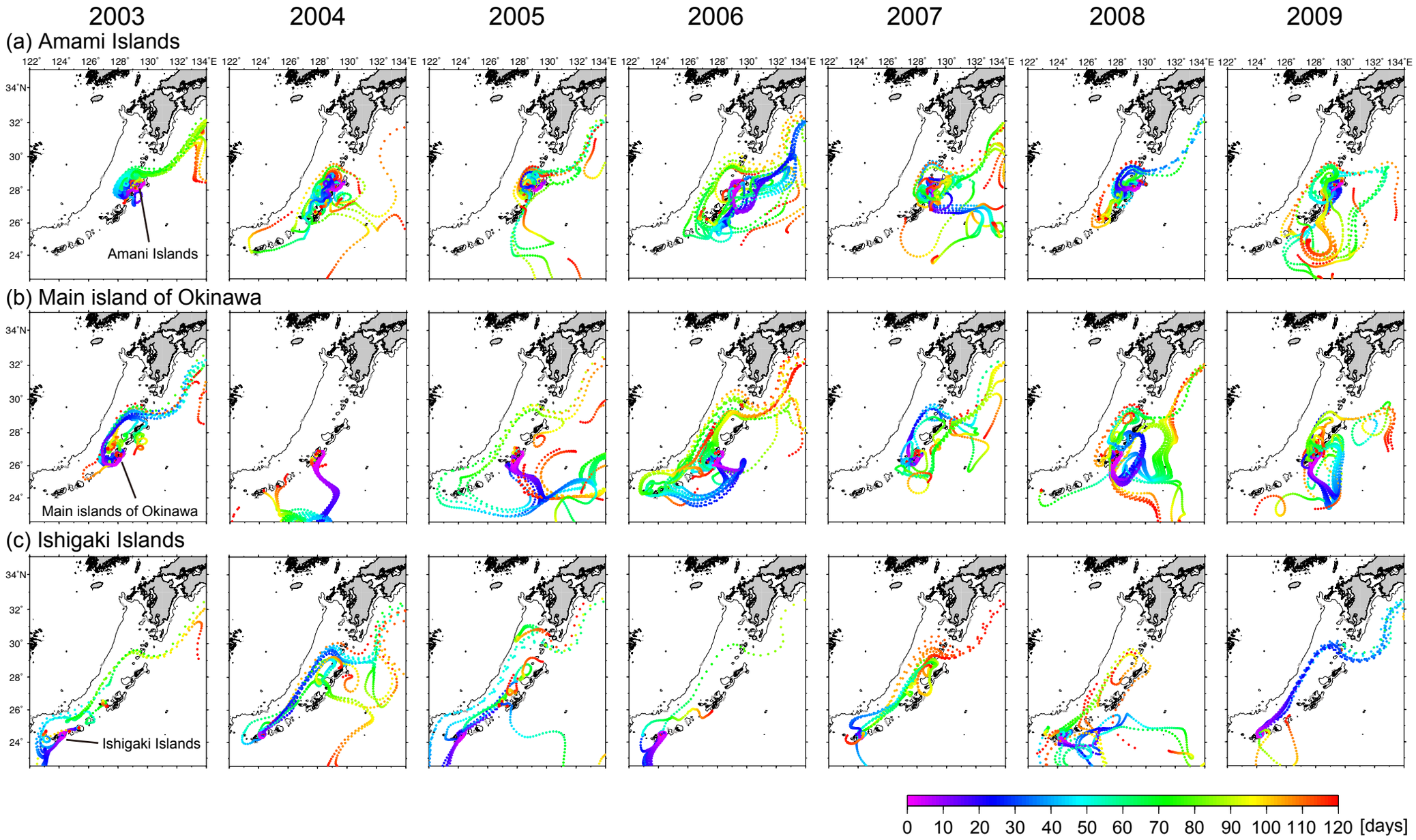

Fig. S6 Particle trajectories from the release points around (a) Amami Islands, (b) Main island of Okinawa, and (c) Ishigaki Islands. The released points of particles were selected at model grid points around the coastal waters of each island

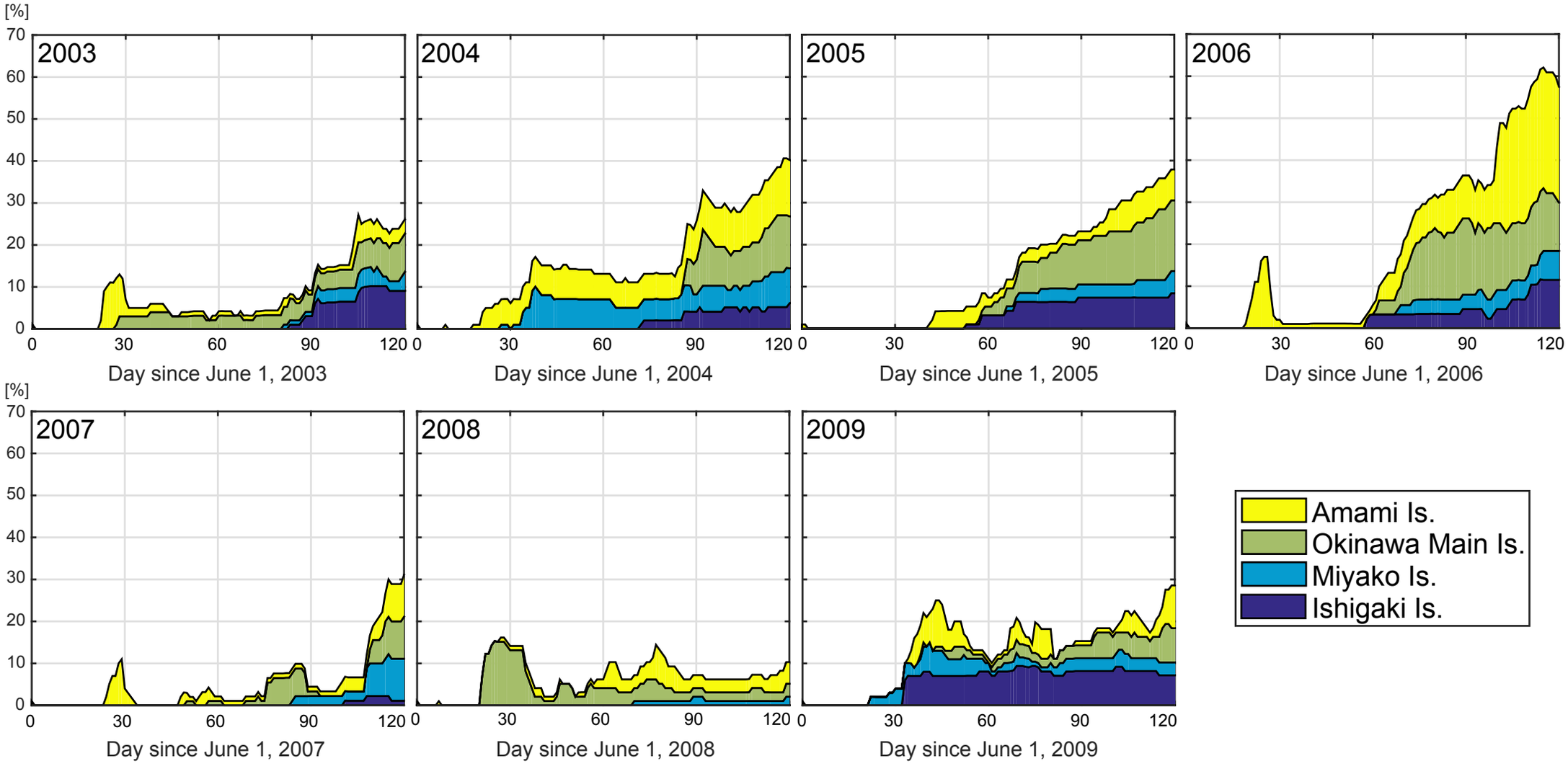

Fig. S7 The ratio of particles delivered to Area K from the Ishigaki Islands, Miyako Islands, Okinawa Main Island, Amami Islands to the total released particles from all islands