

The Effect of Latitude on Carbon, Nitrogen and Oxygen Stable Isotope Ratios in Foliage and in Nitric- oxide ions of Aerosols

Katsura, H.^{1,2*}

¹The United Graduate School of Agricultural Science, Tokyo University of Agriculture & Technology, Saiwai-Cho 3-Chome, Fuchu-Shi, Tokyo 183-8509, Japan

²Nihon University Buzan, Otsuka 5-Chome, Bunkyo-Ku, Tokyo 112-0012, Japan

Received 25 Sep. 2011;

Revised 7 June 2012;

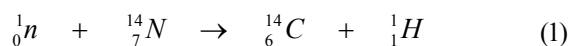
Accepted 14 June 2012

ABSTRACT: Cosmic rays in the upper troposphere (9000 meters to 15000 meters) initiate the following nuclear chemical reaction: $1n + {}^{14}\text{N} \Rightarrow {}^{14}\text{C} + {}^1\text{H}$. Previous research has shown a strong effect of latitude on the abundance of neutrons from cosmic rays. However, to date, there has been little exploration of the relationship between the latitude effect for cosmic-ray neutrons and latitudinal variations of stable isotope ratios in aerosols and foliage. In this study, aerosol samples (PM 4.5) and foliage samples were collected in Singapore in November 2009, February and July 2010 and in Fairbanks, Alaska, U.S.A. in January, April and September 2010. The average value of $\delta 15/14\text{N}$ in foliage in Fairbanks was -1.84 [per mil], whereas the average value in Singapore was -1.3 [per mil]. These results show a clear latitude effect on $\delta 15/14\text{N}$ in foliage. Furthermore, the average value of $\delta 15/14\text{N}$ in the nitric-oxide substances in the aerosol samples in Fairbanks was -2.70 [per mil], whereas the average value in Singapore was +7.61 [per mil], demonstrating that $\delta 15/14\text{N}$ in nitric-oxide substances from aerosol samples also experiences a latitude effect. In both Singapore and Fairbanks, it was observed that values of $\delta 15/14\text{N}$ in nitric-oxide substances from aerosols were correlated with declination. The value of $\delta 15/14\text{N}$ in nitric-oxide substances from aerosols in Fairbanks increased with increasing declination due to more active conversions from ${}^{14}\text{N}$ to ${}^{14}\text{C}$ by neutron bombardment.

Key words: Latitude, Effect, Stable, Isotope, Ratio

INTRODUCTION

The analysis of stable isotopes can provide important insights into many aspects of the global environment, including mechanisms relevant to climate change, such as the geochemical behaviour of aerosols and the functions of foliage (Fang 2011; Samuel 2008; Baechmann 1996; Kapdan *et al.*, 2011; Katsura, 2012; Gallardo and Aoki, 2012). Relatively little is known about latitudinal differences in stable isotopes of nitrogen and oxygen occurring in nitric-oxide substances from aerosols, or about the concentration of anions in aerosols. Knowledge about the latitudinal effects of carbon and nitrogen in foliage on stable isotopes is also limited. Cosmic rays provide a mechanism for such latitudinal effects. In the upper troposphere (30000 feet to 50000 feet; 9000 meters to 15000 meters), cosmic rays initiate the following nuclear chemical reaction (Libby 1946):



Previous research has shown a strong effect of latitude on the abundance of neutrons from cosmic rays (Bieber 2010). However, to date there has been little exploration of the relationship between the latitude effect for cosmic-ray neutrons and latitudinal variations of stable isotope ratios in aerosols and foliage. This study aimed to investigate this relationship by comparing aerosol and foliage samples from an equatorial region (Singapore) and a sub-polar region (Fairbanks, Alaska, U.S.A.). Aerosol samples (PM 4.5) and foliage samples were collected at the National University of Singapore (NUS) in Singapore (latitude: 1°18' N; longitude: 103°46' E; altitude: 67.0 meters) from 16 to 22 November 2009, 08 to 20 February 2010 and 16 to 28 July 2010 and at Fairbanks International Airport in Fairbanks, Alaska, U.S.A. (latitude: 64°50.116'; longitude: 147°49.747' W; altitude: 143.3 meters) from 02 to 05 January 2010, 01 to 09 April 2010 and 21 to 27 September 2010. Anion concentrations in the aerosol samples were analysed by ion chromatography, and nitrogen and oxygen stable isotope ratios were analysed by gas chromatography-mass spectrometry. Carbon and

*Corresponding author E-mail: hi@katsura.dk

nitrogen stable isotope ratios in foliage were analyzed by element mass spectrometry. The objective of this study was to elucidate how the latitude effect on neutron bombardment from cosmic rays affects the isotopic characteristics of aerosols and foliage.

MATERIALS & METHODS

Aerosol samples (PM 4.5) were taken at the rooftop of Building E2 at the National University of Singapore in Singapore (latitude: 1°18' N; longitude: 103°46' E; altitude: 67.0 meters) from 15:27 SGT (Singapore Time = SGT = UTC +7 hours) 16 to 15:27 SGT 17 November 2009; from 15:39 SGT 17 to 15:39 SGT 18 November 2009; from 15:42 SGT 18 to 15:42 SGT 19 November 2009; from 18:18 SGT 19 to 18:18 SGT 20 November 2009; from 18:35 SGT 20 to 18:35 SGT 21 November 2009; from 19:08 SGT 21 to 19:08 SGT 22 November 2009; from 17:09 SGT 08 to 17:09 SGT 12 February 2010; from 17:12 SGT 12 to 17:12 SGT 16 February 2010; from 17:13 SGT 16 to 17:13 SGT 20 February 2010; from 13:22 SGT 16 to 13:22 SGT 20 July 2010; from 13:25 SGT 20 to 13:25 SGT 24 July 2010; and from 13:25 SGT 24 to 13:25 SGT 28 July 2010. A sampling pump (Model SP 250, GL Science) was operated at the rooftop for sample extraction and the samples were collected onto polyamide filters (NX047100, Pall Corporation) at a flow rate of 5 litres per minute.

Aerosol samples (PM 4.5) were taken at Fairbanks International Airport in Fairbanks, Alaska, U.S.A.

(latitude: 64°50.116' N; longitude: 147°49.747' W; altitude: 143.3 meters) from 14:14 AST (Alaska Standard Time = AST = UTC -9 hours) 02 to 14:14 AST 03 January 2010; from 14:17 AST 03 to 14:17 AST 04 January 2010; from 14:21 AST 04 to 14:21 AST 05 January 2010; from 17:11 ADT (Alaska Daylight Saving Time = ADT = UTC -8 hours) 01 to 17:10 ADT 03 April 2010; from 17:12 ADT 03 to 17:12 ADT 05 April 2010; from 17:14 ADT 05 to 17:14 ADT 07 April 2010; from 17:16 ADT 07 to 17:16 ADT 09 April 2010; from 10:10 ADT 21 to 10:10 ADT 23 September 2010; from 10:12 ADT 23 to 10:12 ADT 25 September 2010 and from 10:14 ADT 25 to 10:14 ADT 27 September 2010. A sampling pump (Model SP 250, GL Science) was operated at the 2nd floor terrace of a lodge (Golden North Motel; 4888 Old Airport Road, Fairbanks, Alaska 99709, U.S.A.) at Fairbanks International Airport in Fairbanks, Alaska, U.S.A. and the samples were collected onto polyamide filters (NX047100, Pall Corporation) at a flow rate of 5 litres per minute.

Each polyamide filter was then transferred into 20 mL of ultrapure water and shaken for approximately 40 minutes. The extracts were filtered and analysed using an ion chromatograph (DX 120/AS, Dianex Inc.).

The $^{15}\text{N}/^{14}\text{N}$ and $\delta^{18}\text{O}/^{16}\text{O}$ isotope ratios in NO_3^- were measured using the denitrifier method (Casciotti *et al.*, 2002; Takebayashi *et al.*, 2010). The NO_3^- was converted to N_2O using a denitrifier (*Pseudomonas aureofaciens*; ATCC 13985) lacking N_2O reductase. The N_2O was then introduced into a Delta XP isotope ratio mass spectrometer coupled to an HP6890 gas chromatograph (Hewlett-Packard Co., Palo Alto, CA, U.S.A.) equipped with a PorapLOT column and a GC interface III (Thermo Fisher Scientific). Anion concentrations and isotope ratios were measured at the Laboratory of Social Biogeochemistry (Laboratory of Professor Muneoki YOH & Associate Professor Keisuke KOBAYASHI), Tokyo University of Agriculture & Technology (TUAT), Building #2, Rooms 328 & 2N-101, 5-8, Saiwai-Cho 3-Chome, Fuchu-Shi, Tokyo 183-8509, Japan. The calibration curves for these isotopic analyses were constructed using the international standards USGS32, USGS34, USGS35 and IAEA. The stable isotope ratio δ was calculated with the following equation (Richet 1977).

(2)

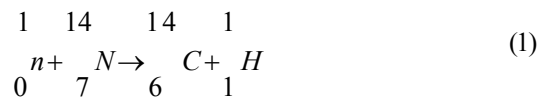
$$\left[\frac{R_{\text{SAMPLE}} - R_{\text{STANDARD}}}{R_{\text{STANDARD}}} \right] \times 1000^\circ / \text{‰} = \delta = \text{delta}$$

Abies firma (Momi or Japanese fir) was selected as the source of foliage samples in Fairbanks, Alaska, U.S.A. This species is a typical forestry tree in the Fairbanks region. *Samanea saman* (rain tree) was selected as the source of foliage samples in Singapore; it is a typical forestry tree in Singapore. Sampling was performed on 05 January 2010, 07 April 2010, 22 September 2010, 23 September 2010 and 25 September 2010 in Fairbanks Alaska, U.S.A. and on 16 February 2010, 18 February 2010, 24 July 2010 and 27 July 2010 in Singapore.

In the laboratory, collected foliage was dried at 80°C to constant weight. All foliage samples were ground using a ball mill (MM200; Retsch GmbH and Co. KG, Haan, Germany). Ground samples were analyzed for delta 15/14N and delta 13/12C using an elemental analyzer (EA1112; Thermo Fisher Scientific K.K., Yokohama, Japan) coupled with a Delta-XP isotope ratio mass spectrometer (Thermo Fisher Scientific K.K.). Calibrated DL-alpha-alanine (delta 13/12C = -23.45 [per mil]; delta 15/14N = -1.66 [per mil]), glycine (delta 13/12C = -34.89 [per mil]; delta 15/14N = 10.04 [per mil]) and histidine (delta 13/12C = -9.94 [per mil]; delta 15/14N = -7.96 [per mil]) were used as internal standards. The stable isotope ratio δ was calculated using equation (1) (Richet 1977).

RESULTS & DISCUSSION

Table 1 shows the statistics of the inorganic anion concentrations and the stable isotope ratios in the nitric-oxide substances within the aerosol samples for all the samples collected in Singapore during this study. Table 2 shows the same data for Fairbanks, Alaska, U.S.A. For both sampling locations, Table 3 shows the following: sampling place, date and time; declination; maximum and minimum temperature; day length; stable isotope ratios in the nitric-oxide substances the aerosol samples; and carbon and nitrogen stable isotope ratios in the foliar samples. It is generally known that delta 15/14N in foliage decreases with increasing latitude (Craine *et al.*, 2009). It was found that the average value of delta 15/14N in foliage in Fairbanks (latitude 64.84° N) was -1.84 [per mil] whereas the average value of delta 15/14N in foliage in Singapore (latitude 1.3° N) was -1.3 [per mil] on Table 3), clearly following the expected trend. However, no such latitudinal trend was apparent for delta 13/12C in foliage: in Fairbanks the average value was -23.18 [per mil] and in Singapore the average value was -24.1 [per mil] (Table 3). Trends for delta 15/14N in aerosols were similar to those in foliage: the average value of delta 15/14N in the nitric-oxide substances within the aerosol samples in Fairbanks was -2.70 [per mil], whereas in Singapore the average value was +7.61 [per mil] (Table 3), showing a clear increase with decreasing latitude. However, delta 18/16O in aerosol samples did not show a latitudinal trend: the average value of delta 18/16O in the nitric-oxide substances within the aerosol samples in Fairbanks was +42.97 [per mil], very similar to the average value in Singapore, which was +46.36 [per mil] (Table 3). In summary, comparison of the Fairbanks samples and the Singapore samples only showed a latitude effect for nitrogen stable isotopes (both in foliage and in nitric-oxide substances within the aerosol samples). In order to understand the effects of neutron bombardment on stable isotopes, it is necessary to refer to the laws governing the binding of the atomic nucleus (Haxel Otto 1949). There are certain nucleons (2, 8, 20, 28, 50, 82, 126) that are more tightly bound than others; this is the origin of the shell model. Even numbers of nucleons are more tightly bound than odd numbers of nucleons. Hence ^{14}N is more easily broken than ^{15}N . The proton number and the neutron number of ^{14}N are both 7. The proton number of ^{15}N is also 7, but its neutron number is 8 (Table 4). Thus a nuclear chemical reaction in which ^{14}N and a neutron (n) produce the ^{14}C radioisotope is known to occur in the atmosphere, especially at altitudes between 30,000 feet and 50,000 feet (9000-15,000 m) (Feather 1932; FERMI 1934; FERMI 1938; Harkins 1933; Libby 1946; Ramsey 2008; Rutherford 1920).



In contrast, there is no known nuclear chemical reaction between ^{15}N and a neutron (n) because ^{15}N is more tightly bound than ^{14}N (due to the fact that ^{15}N has a neutron number of 8). Therefore only ^{14}N is affected by neutron bombardment from the sun and from cosmic rays. Thus ^{14}N is converted into the ^{14}C radioisotope and the relative amount of ^{15}N in the stable isotope ratio is increased. The latitude effect on nitrogen stable isotope ratios (both in the foliage and in the nitric-oxide substances within the aerosol samples) should be affected by these principles. Lower latitudes such as Singapore tend to have higher atmospheric temperatures than do higher latitudes such as Fairbanks, Alaska. Higher atmospheric temperatures generate stronger updrafts, allowing aerosols to climb to higher altitudes. At these higher altitudes, neutrons are more abundant than at lower altitudes. Consequently, at lower latitudes there are more active conversions from ^{14}N to ^{14}C by neutron bombardment. This is the mechanism by which nitrogen stable isotope ratios (delta 15/14N), both in foliage and in nitric-oxide substances within the aerosol samples, are increased at lower latitudes.

In Singapore, day length as well maximum and minimum temperature remain nearly constant throughout the year. If delta 15/14N was correlated with these factors only, it should not show substantial change throughout the year (Table 3, Fig. 1, Fig. 3, Fig. 4, Fig. 5). However, delta 15/14N in Singapore does vary considerably over the course of a year. These variations in delta 15/14N in nitric-oxide substances within Singapore aerosol samples were clearly correlated with declination. delta 15/14N in nitric-oxide substances within the aerosol samples reached maximum values at declinations approximately 20° N and 20° S. Minimum values were reached at approximately 0° N and S (Fig. 2). Singapore is located at 1.3° N, almost at the equator (0° N and S). Therefore, similar declination values indicate similar positions of the sun from Singapore. This allowed us to confirm the correlation between declination and delta 15/14N in nitric-oxide substances within aerosols. Another mechanism by which latitude affects stable isotope ratios concerns the deflection of cosmic rays. Cosmic radiation decreases with decreasing latitude, reaching a minimum at the equator. This is because the Earth's geomagnetic field (specifically the Van Allen radiation belt) deflects cosmic rays most effectively at the equator (Van Allen 1959). When declination approaches 0° N and S, cosmic radiation to Singapore decreases.

Table 1. Stable Isotope Ratios and Anion Concentration of Aerosol (PM=4.5) (5L/min.) at National University of Singapore in Singapore, from 16 NOV 2009 to 28 JUL 2010

Sampling Date & Time (Singapore Time = SGT = UTC + 8 hours)	$\delta^{15}\text{N}/^{14}\text{N}$ [per mil]	$\delta^{18}\text{O}/^{16}\text{O}$ [per mil]	NO_3^- [Micro Mol/L]	NO_2^- [Micro Mol/L]	F- [Micro L]	Cl- [Micro Mol/L]	SO_4^{2-} [Micro Mol/L]	PO ₄ ^{3-} [Micro Mol/L]
1527 SGT 16 to 1527 SGT 17 NOV 2009	27.19	56.16	1.18	0.96	N/A	2.28	1.22	N/A
1539 SGT 17 to 1539 SGT 18 NOV 2009	9.02	62.37	1.2	0.54	N/A	2.35	1.75	N/A
1542 SGT 18 to 1542 SGT 19 NOV 2009	21.36	25.54	0.2	N/A	N/A	0.46	N/A	N/A
1818 SGT 19 to 1818 SGT 20 NOV 2009	7.78	21.17	0.23	N/A	N/A	0.7	N/A	N/A
1835 SGT 20 to 1835 SGT 21 NOV 2009	N/A	N/A	N/A	N/A	N/A	0.23	N/A	N/A
1908 SGT 21 to 1908 SGT 22 NOV 2009	16.64	41.91	0.62	0.46	N/A	1.6	0.33	N/A
1709 SGT 08 to 1709 SGT 12 FEB 2010 (Official CHINESE NEW YEAR was from SUN 14 to TUE 16 FEB 2010)	-6.97	56.14	17.89	2.17	2.648	24.23	13.54	2.496
1712 SGT 12 to 1712 SGT 16 FEB 2010	-7.99	56.75	8.18	0.93	0.037	22.17	7.34	N/A
1713 SGT 16 to 1713 SGT 20 FEB 2010	-6.74	59.18	10.48	N/A	0.784	30.48	9.2	N/A
1322 SGT 16 to 1322 SGT 20 JUL 2010	9.74	34.46	6.23	1.44	0.96	7.84	22.65	N/A
1325 SGT 20 to 1325 SGT 24 JUL 2010	8.03	35.11	9.01	N/A	N/A	13.93	20.46	N/A
1325 SGT 24 to 1325 SGT 28 JUL 2010	5.67	61.12	3.73	N/A	N/A	5.54	10.22	N/A
Average	7.781	45.96	5.13	1.08	1.10	9.23	8.00	2.49
Standard Deviation	12.85	15.60	6.14	0.63	1.10	11.69	8.07	N/A
Variance	165.26	243.54	37.70	0.40	1.21	136.71	65.17	N/A
Range	35.18	41.2	17.69	1.71	2.61	30.25	22.32	0
Minimum Value.	-7.99	21.17	0.2	0.46	0.037	0.23	0.33	2.49
Maximum Value	27.19	62.37	17.89	2.17	2.648	30.48	22.65	2.49
Total	70.03	413.68	46.21	6.5	4.429	92.34	56.03	2.49
Number of Sample	9	9	9	6	4	10	7	1

Table 2. Stable isotope ratio and anion concentration of aerosol (PM_{4.5}) (5 L/Minute) at Fairbanks International Airport (N: 64 Degree 50.116 Minute, W: 147 Degree 49.747 Minute, Altitude: 143.4 meter) in Fairbanks Alaska U.S.A

Sampling Date & Time (Alaska Standard Time = AST δ ¹⁵ N/14N δ ¹⁸ O/16O per mil)	NO ₃ - [Micro Mol/L]	NO ₂ - [Micro Mol/L]	F- [Micro Mol/L]	Cl- [Micro Mol/L]	SO ₄ 2- [Micro Mol/L]	PO ₄ 3- [Micro Mol/L]
1414 AST 02 to 1414 AST 03 JAN 2010	N/A	N/A	0.18	0.48	N/A	1.2
1417 AST 03 to 1417 AST 04 JAN 2010	0.26	N/A	1.73	N/A	N/A	N/A
1421 AST 04 to 1421 AST 05 JAN 2010	0.22	N/A	N/A	0.72	N/A	1.68
1711 ADT 01 to 1710 ADT 03 APR 2010	2.05	N/A	1.33	11.07	1.34	N/A
1712 ADT 03 to 1712 ADT 05 APR 2010	0.93	N/A	1.07	11.17	1.72	N/A
1714 ADT 05 to 1714 ADT 07 APR 2010	1.21	N/A	0.0054	5.06	N/A	N/A
1716 ADT 07 to 1716 ADT 09 APR 2010	N/A	N/A	0.021	3.15	0.53	N/A
1010 ADT 21 to 1010 ADT 23 SEP 2010	N/A	N/A	N/A	1.1	N/A	N/A
1012 ADT 23 to 1012 ADT 25 SEP 2010	N/A	N/A	N/A	0.52	N/A	N/A
1014 ADT 25 to 1014 ADT 27 SEP 2010	N/A	N/A	N/A	N/A	N/A	N/A
Average	0.934	N/A	0.72273	4.15875	1.19667	1.44
Standard Deviation	0.7559	N/A	0.74907	4.57801	0.60781	0.33941
Variance	0.5714	N/A	0.5611	20.9582	0.36943	0.1152
Range	1.83	0	1.7246	10.69	1.19	0.48
Minimum Value.	0.22	0	0.0054	0.48	0.53	1.2
Maximum Value	2.05	0	1.73	11.17	1.72	1.68
Total	4.67	0	4.3364	33.27	3.59	2.88
Number of Sample	5	0	6	8	3	2

Table 3. Stable isotope ratios in aerosol (PM 4.5) (5 L/Minute) and foliage at National University of Singapore (N: 1 degree 18 minute; E: 103 degree 46 minute; Altitude: 67.0 meter), Singapore and Fairbanks International Airport (N: 64 Degree 50.116 Minute, W: 147 Degree 49.747 Minute, Altitude: 143.4 meter) Fairbanks Alaska U.S.A. -Continues

Sampling Place, Date & Time (Singapore Time = SGT + 8 hours) (Alaska Standard Time = UTC - 9 hours; Alaska Daylight Saving Time = ADT = UTC - 8 hours;)	Date	Declination [degree]	Maximum Temperature in Sampling Place [degree Celsius]	Minimum Temperature in Sampling Place [degree Celsius]	Length of day in Aerosol in Sampling Place [hour]	$\delta 15N/14N$ [per mil]	$\delta 15O/16O$ [per mil]	$\delta 13C/12C$ [per mil]	$\delta 15N/14N$ [per mil]	$\delta 13C/12C$ [per mil]	$\delta 15N/14N$ [per mil]	in foliage of Samanea saman (Rain tree) in Singapore
Singapore	16-Nov-09	S 18.70	27.03	23.93	12.05	N/A	N/A	N/A	N/A	N/A	N/A	N/A
1527 SGT 16 to 1527 SGT 17	17-Nov-09	S 18.95	30.17	24.16	12.05	N/A	N/A	27.19	56.16	N/A	N/A	N/A
NOV 2009 in Singapore												
1539 SGT 17 to 1539 SGT 18	18-Nov-09	S 19.20	29.79	25.38	12.05	N/A	N/A	9.02	62.37	N/A	N/A	N/A
NOV 2009 in Singapore												
1542 SGT 18 to 1542 SGT 19	19-Nov-09	S 19.43	28.4	22.86	12.05	N/A	N/A	21.36	25.54	N/A	N/A	N/A
NOV 2009 in Singapore												
1818 SGT 19 to 1818 SGT 20	20-Nov-09	S 19.67	27.62	23.33	12.05	N/A	N/A	7.78	21.17	N/A	N/A	N/A
NOV 2009 in Singapore												
1908 SGT 21 to 1908 SGT 22	22-Nov-09	S 20.10	29.37	24.4	12.05	N/A	N/A	16.64	41.91	N/A	N/A	N/A
NOV 2009 in Singapore												
1414 AST 02 to 1414 AST 03 JAN 2010 in Fairbanks	3-Jan-10	S 22.85	-31.1	-36.7	4.13	-24.11	35.63	N/A	N/A	N/A	N/A	N/A

Table 3 . Stable isotope ratios in aerosol (PM 4.5) (5 L/Minute) and foliage at National University of Singapore (N: 1 degree 18 minute; E: 103 degree 46 minute; Altitude: 67.0 meter), Singapore and Fairbanks International Airport (N: 64 Degree 50.116 Minute, W: 147 Degree 49.747 Minute, Altitude: 143.4 meter) Fairbanks Alaska U.S.A. -Continues

I417 AST 03 to I417 AST 04 JAN 2010 in Fairbanks	4-Jan-10	S 22.75	-22.2	-32.2	4.2	-9.28	26.87	N/A	N/A	N/A	N/A	N/A	N/A
I421 AST 04 to I421 AST 05 JAN 2010 in Fairbanks	5-Jan-10	S 22.63	-16.1	-22.8	4.28	-11.26	43.66	N/A	N/A	-17	-7.1	N/A	N/A
I709 SGT 08 to I709 SGT 12 FEB 2010 in Singapore; (Official CHINESE NEW YEAR was from SUN 14 to TUE 16 FEB 2010)	12-Feb-10	S 13.78	31.74	24.92	12.07	N/A	N/A	-6.97	56.14	N/A	N/A	N/A	N/A
I712 SGT 12 to I712 SGT 16 FEB 2010 in Singapore	16-Feb-10	S 12.43	30.53	25.57	12.07	N/A	N/A	-7.99	56.75	N/A	N/A	-15.2	2.6
I713 SGT 16 to I713 SGT 20 FEB 2010 in Singapore	18-Feb-10	S 11.73	30.52	25.87	12.07	N/A	N/A	N/A	N/A	N/A	N/A	-15.8	-5.1
I714 ADT 01 to I714 ADT 03 APR 2010 in Fairbanks	20-Feb-10	S 11.03	29.01	25.81	12.07	N/A	N/A	-6.74	59.18	N/A	N/A	N/A	N/A
I715 ADT 03 to I715 ADT 05 APR 2010 in Fairbanks	3-Apr-10	N 5.18	7.8	-6.7	13.85	2.43	29.93	N/A	N/A	N/A	N/A	N/A	N/A
I716 ADT 05 to I716 ADT 07 APR 2010 in Fairbanks	5-Apr-10	N 5.93	11.1	-6.1	14.07	5.1	38.77	N/A	N/A	N/A	N/A	N/A	N/A
I717 ADT 07 to I717 ADT 09 APR 2010 in Fairbanks	6-Apr-10	N 6.32	6.1	-2.2	14.18	N/A	N/A	N/A	N/A	-14.3	-6.4	N/A	N/A
I718 ADT 09 to I718 ADT 11 APR 2010 in Fairbanks	7-Apr-10	N 6.70	1.7	-3.9	14.3	1.76	28.57	N/A	N/A	-13.9	1.8	N/A	N/A

Table 3 . Stable isotope ratios in aerosol (Continuation)

1322 SGT 16 to 1322 SGT 20 JUL 2010 in Singapore	20-Jul-10	N 20.71	29.33	23.71	12.18	N/A	N/A	34.46	N/A	N/A	N/A	N/A
1325 SGT 20 to 1325 SGT 24 JUL 2010 in Singapore	24-Jul-10	N 19.91	27.99	23.9	12.18	N/A	N/A	35.11	N/A	N/A	-32.7	-1.1
1325 SGT 24 to 1325 SGT 28 JUL 2010 in Singapore	27-Jul-10	N 19.27	28.74	25.9	12.16	N/A	N/A	N/A	N/A	N/A	-32.7	-1.6
1325 SGT 28 JUL 2010 in Singapore	28-Jul-10	N 19.05	27.56	23.5	12.16	N/A	N/A	61.12	N/A	N/A	N/A	N/A
Fairbanks	22-Sep-10	N 0.43	9.4	0	12.26	N/A	N/A	N/A	-28.8	-0.5	N/A	N/A
1010 ADT 21 to 1010 ADT 23 SEP 2010 in Fairbanks	23-Sep-10	N 0.05	8.3	-2.8	12.17	2.38	69.99	N/A	-28.8	-0.6	N/A	N/A
Fairbanks	24-Sep-10	S 0.33	6.1	-2.8	12.05	N/A	N/A	N/A	-29.4	0.1	N/A	N/A
1012 ADT 23 to 1012 ADT 25 SEP 2010 in Fairbanks	25-Sep-10	S 0.72	6.7	-5	11.95	4.11	56.29	N/A	-30.1	-0.2	N/A	N/A
1014 ADT 25 to 1014 ADT 27 SEP 2010 in Fairbanks	27-Sep-10	S 1.50	3.3	-8.9	11.72	4.6	57	N/A	N/A	N/A	N/A	N/A
Average												
Standard Deviation												
Variance												
Range												
Minimum Value												
Maximum Value												
Total												
Number of Sample												
			9	9	11	11	7	11	7	7	4	4
			-2.6967	42.9678	7.61182	46.3555	-23.186	-1.84286	-24.1	9.933445	9.926667	3.150661
			10.0466	15.0538	11.5167	15.1465	7.6693	3.45198	58.8181	11.9162	98.67333	9.926667
			100.935	22.6618	132.633	229.415	16.2	8.9	17.5	7.7	-5.1	2.6
			29.21	43.12	35.18	21.17	-30.1	-7.1	-32.7	-15.2	-96.4	-5.2
			-24.11	26.87	-7.99	62.37	-13.9	1.8	-15.2	-96.4	-5.2	4
			5.1	69.99	27.19	509.91	-162.3	-12.9	-96.4	-5.2	4	4
			-24.27	386.71	83.73	509.91	-162.3	-12.9	-96.4	-5.2	4	4

Table 4. Characterisations of Carbon (C), Nitrogen (N) and Oxygen (O) Stable Isotopes for this study

Stable Isotope	12C	13C	14N	15N	16O	18O
Atomic Number = Proton Number	6	6	7	7	8	8
Mass Number	12	13	14	15	16	18
Neutron Number	6	7	7	8	8	10

Therefore conversion from ^{14}N into ^{14}C decreases, and thus the relative amount of ^{15}N in the stable isotope ratio also decreases. On the other hand, as declination approaches 20°N and S, cosmic radiation to Singapore increases, and accordingly the relative amount of ^{15}N in the stable isotope ratio also increases. The results of this study were closely aligned with these explanations of the relationship between declination and values of delta $15/14\text{N}$ in nitric-oxide substances within the aerosol samples.

Fig. 2 shows a significant positive linear relationship between declination and delta $15/14\text{N}$ in nitric-oxide substances from the aerosol samples in Fairbanks. However, unlike Singapore, Fairbanks also

showed distinct trends for maximum and minimum atmospheric temperature and day length. In higher-latitude regions such as Fairbanks, declination has a greater effect on maximum and minimum atmospheric temperature and day length than it does in lower-latitude regions such as Singapore. Longer day lengths in Fairbanks contribute to higher atmospheric temperatures, which in turn propel nitric-oxide substances within the aerosol to higher altitudes where they are exposed to greater numbers of neutrons. Consequently, it was observed that the value of delta $15/14\text{N}$ in nitric-oxide substances within the aerosol samples in Fairbanks increased with increasing declination, due to more active conversions from ^{14}N to ^{14}C by neutron bombardment.

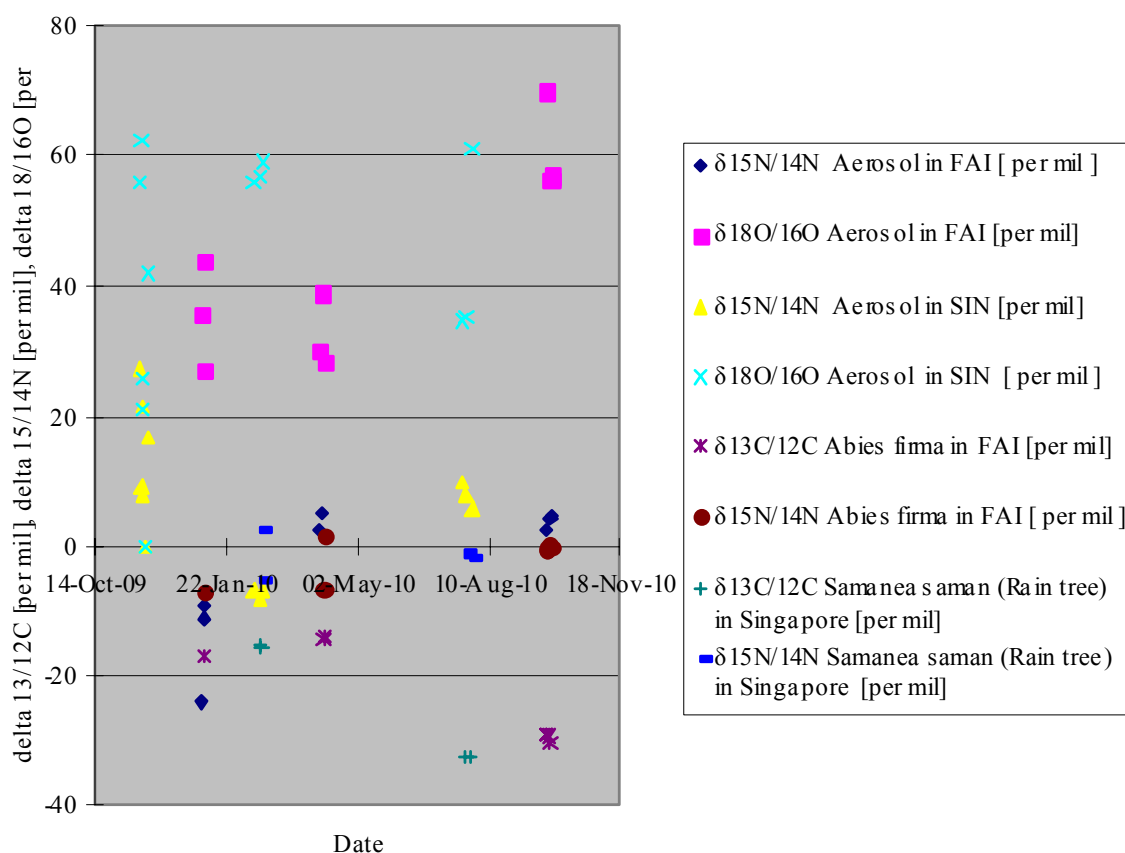


Fig.1. Date Vs. Isotope Ratios (FAI: Fairbanks, Alaska; Latitude: 64.84 degree North) (SIN: Singapore; Latitude: 1.3 degree North)

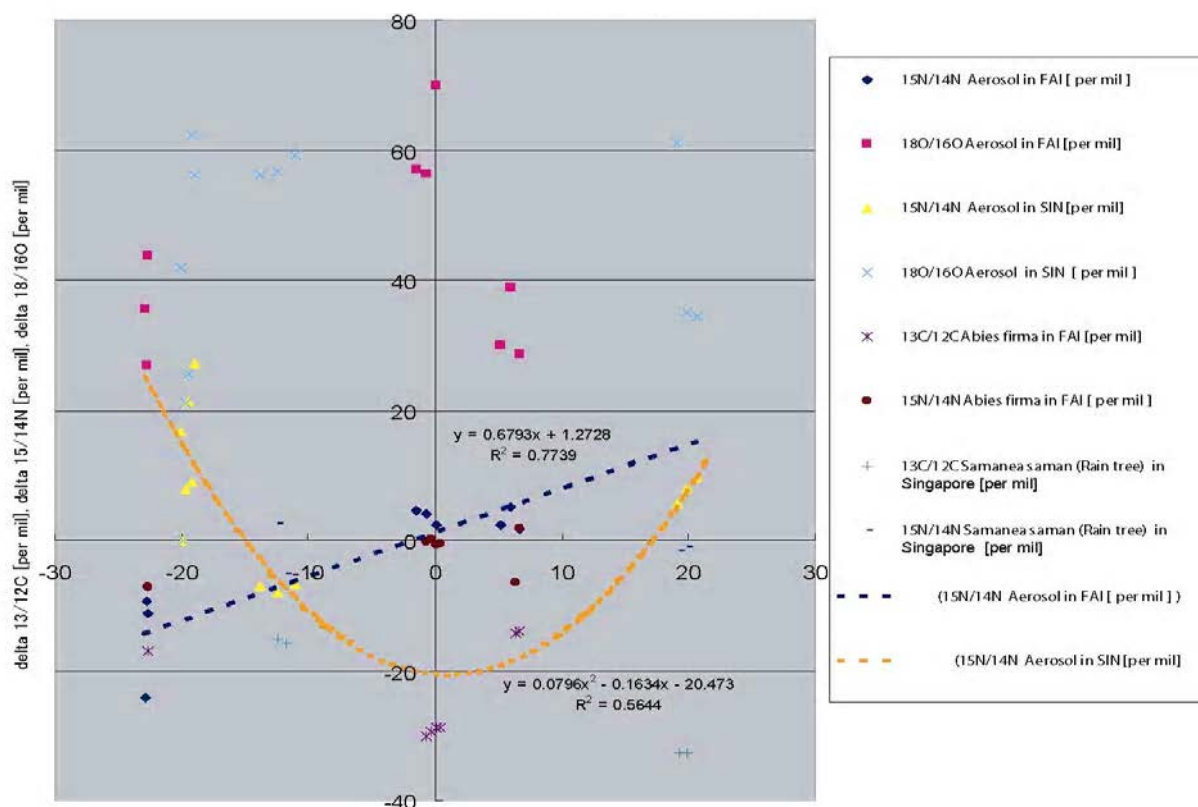


Fig. 2. Declination vs. Isotope Ratios(FAI: Fairbanks, Alaska; Latitude: 64.84 degree North)
(SIN: Singapore; Latitude: 1.3 degree North)

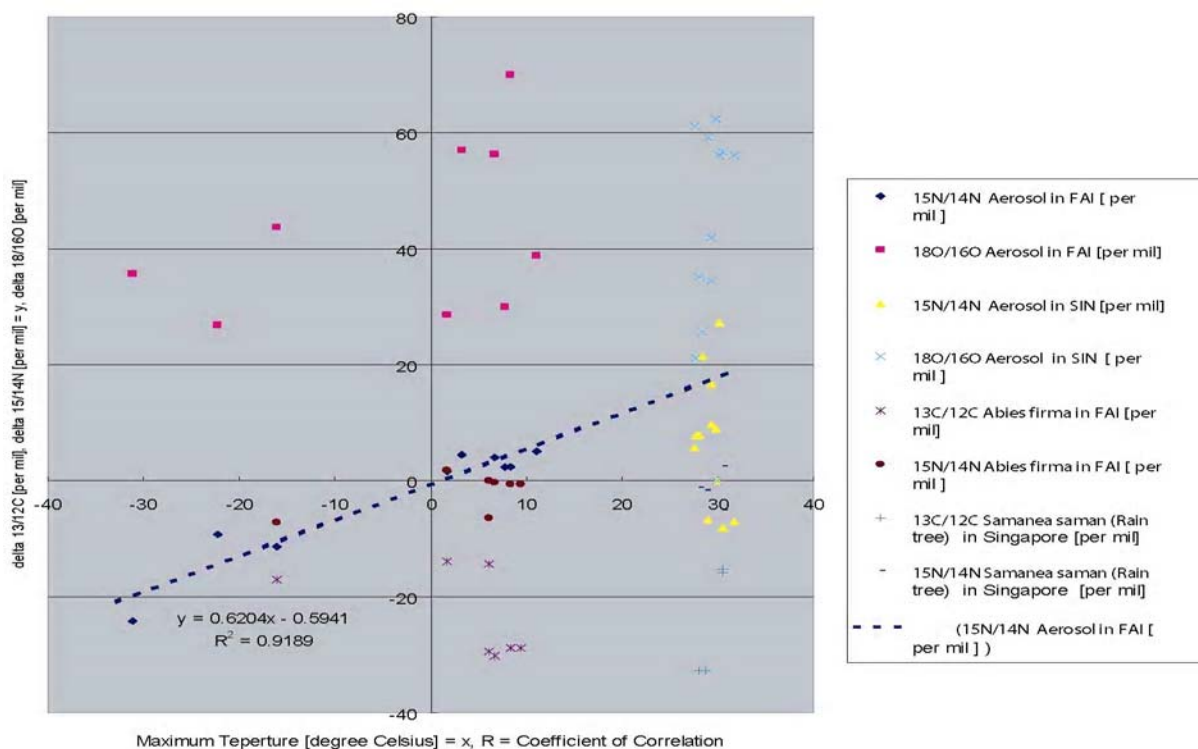


Fig. 3. Maximum Temperature vs. Isotope Ratios (FAI: Fairbanks, Alaska; Latitude: 64.84 degree North)
(SIN: Singapore; Latitude: 1.3 degree North)

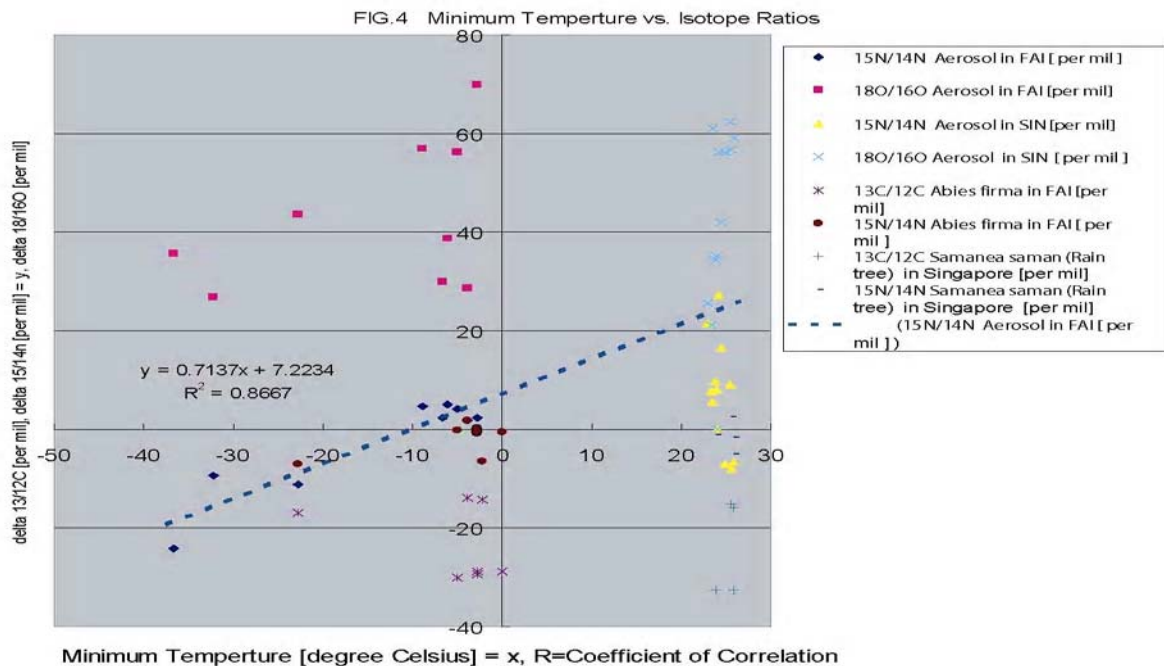


Fig. 4. Minimum Temperature vs. Isotope Ratios (FAI: Fairbanks, Alaska; Latitude: 64.84 degree North) (SIN: Singapore; Latitude: 1.3 degree North)

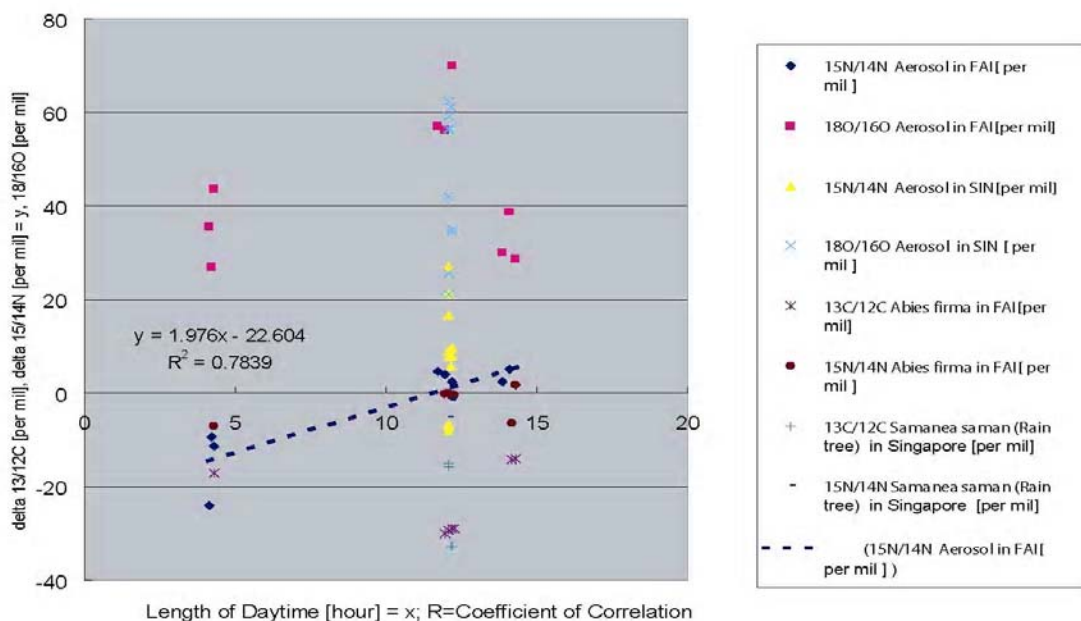


Fig. 5. Length of Daytime vs. Isotope Ratios (FAI: Fairbanks, Alaska; Latitude: 64.84 degree North) (SIN: Singapore; Latitude: 1.3 degree North)

CONCLUSION

It is known general tendency as a latitude effect that nitrogen stable isotope ratios delta 15/14N in the foliage in lower latitude has higher than that of in higher latitude (Craine 2009). Average value of delta 15/14N in foliage in Fairbanks (Latitude: 64.84 degree North) was -1.84 [per mil] versus average value of delta 15/14N in foliage in Singapore (Latitude: 1.3 degree North) was -1.3 [per mil] on Table 3. It was subjected to

this latitude effect clearly. However average value of delta 13/12C in foliage in Fairbanks was -23.18 [per mil] versus average value of delta 13/12C in foliage in Singapore was -24.1 [per mil] on Table 3. It was unclear tendency for delta 13/12C in foliage between Fairbanks and Singapore. In addition average value of delta 15/14N in the nitric-oxide substances within the aerosol samples in Fairbanks was -2.70 [per mil] versus average

value of delta 15/14N in the nitric-oxide substances within the aerosol samples in Singapore was +7.61 [per mil] on Table 3. It was also subjected to this latitude effect cleanly in spite of samples were not foliage, it was nitric-oxide substances within the aerosol samples. Meanwhile average value of delta 18/16O in the nitric-oxide substances within the aerosol samples in Fairbanks was +42.97 [per mil] whereas average value of delta 18/16O in the nitric-oxide substances within the aerosol samples in Singapore was +46.36 [per mil] on Table 3. It was unclear tendency for 18/16O in the nitric-oxide substances within the aerosol samples between Fairbanks and Singapore. Therefore clearly latitude effect between Fairbanks (Latitude: 64.84 degree North) and Singapore (Latitude: 1.3 degree North) was only observed for nitrogen stable isotopes. both in the foliage and in nitric-oxide substances within the aerosol samples.

[2] In Singapore it was also observed that values of delta 15/14N in nitric-oxide substances within the aerosol samples were clearly correlated with declination.

[3] The value of delta 15/14N in nitric-oxide substances within the aerosol samples in Fairbanks increased with increasing declination due to more active conversions from ^{14}N to ^{14}C by neutron bombardment.

ACKNOWLEDGEMENT

The author i.e. Hidemitsu KATSURA wishes to thank Neutron monitors of the Bartol Research Institute, University of Delaware in Newark, Delaware U.S.A. are supported by the National Science Foundation; Dr. Blake Moore, Alaska Climate Research Center in Fairbanks, Alaska, U.S.A.; Mr. Paul and Ms. Betty BAER, Owners of Golden North Motel in Fairbanks, Alaska, U.S.A. and Dr. Rajasekhar BALASUBRAMANIAN, Professor, National University of Singapore in Singapore; Dr. Ryo FUNADA, Dean, The United Graduate School of Agricultural Science (UGSAS), TUAT; Dr. Masao TAKAYANAGI, Deputy Dean, UGSAS, TUAT for assisting with this study. The international standards used in this study were purchased from Shoko SI Science in Matsudo-Shi, Chiba-Ken, Japan, in May 2008 by Muneoki YOH, Professor, TUAT.

REFERENCES

Baechmann, K. (1996). The chemical content of raindrops as a function of drop radius-I. Field measurement at the cloud base and below the cloud. *Atmos. Environ.*, **30** (7), 1019-1033.

Bieber, J. W. (2010). Cosmic Rays and Earth - A Summary. *Space Science Reviews*, **93** (1-2), 1-9.

Casciotti, K. L. (2002). Measurement of the Oxygen Isotopic Composition of Nitrate in Seawater and Freshwater Using the Denitrifier Method. *Anal. Chem.*, **74** (19), 4905-4912.

Craine, J. M. (2009). Global patterns of foliar nitrogen isotopes and their relationships with climate, mycorrhizal

fungi, foliar nutrient concentrations, and nitrogen availability. *New Phytologist*, **183** (4), 980-992.

Fang, Y. (2011). Anthropogenic imprints on nitrogen and oxygen isotopic composition of precipitation nitrate in a nitrogen-polluted city in southern China. *Atmos. Chem. Phys.*, **11**, 1313-1325.

Feather, N. (1932). The collisions of neutrons with nitrogen nuclei. *Proceedings of the Royal Society of London. Series A.*

FERMI, E. (1934). Radioactivity Induced by Neutron Bombardment. *Nature*, **133**, 757-757.

FERMI, E. (1938). Artificial radioactivity produced by neutron bombardment. *Nobel Lecture*.

Gallardo, A. H. and Aoki, H. (2012). Attitude Toward the Geological Disposal of Radioactive Wastes in Japan: the Opinion of the Youth prior to the Tohoku Earthquake. *Int. J. Environ. Res.*, **6** (2), 399-408.

Harkins, W. (1933). The Disintegration of the Nuclei of Nitrogen and Other Light Atoms by Neutrons. I. *Phys. Rev.* **44**, 529-537.

Haxel, O. (1949). On the "magic numbers" in nuclear structure. *Phys. Rev.* **75**, 1766-1766.

Kapdan, E., Varinlioglu, A. and Karahan, G. (2011). Radioactivity Levels and Health Risks due to Radionuclides in the Soil of Yalova, Northwestern Turkey. *Int. J. Environ. Res.*, **5** (4), 837-846.

Katsura, H. (2012). The Effect of Electrically Charged Clouds on the Stable Nitrogen Isotope Ratio and the Anion Concentrations in Cloud-based Aerosols. *Int. J. Environ. Res.*, **6** (2), 457-466.

Libby, W. F. (1946). Atmospheric helium three and radiocarbon from cosmic radiation. *Physical Review*, **69**, 671-672.

Ramsey, C. B. (2008). Radiocarbon dating: revolutions in understanding. *Archaeometry*, **50** (2), 249-275.

Rutherford, E. (1920). Bakerian Lecture. Nuclear Constitution of Atoms. *Proceedings of the Royal Society of London. Series A.*

Richet, P., Bottinga, Y. and Javoy, M. (1977). A Review of Hydrogen, Carbon, Nitrogen, Oxygen, Sulphur, and Chlorine Stable Isotope Fractionation Among Gaseous Molecules. *Annual Review of Earth and Planetary Sciences*, **5**, 65-110.

Samuel M. (2008). Tracing the Origin and Fate of NO_x in the Arctic Atmosphere Using Stable Isotopes in Nitrate. *Science*, **322** (5902), 730-732.

Takebayashi, Y. (2010). The natural abundance of ^{15}N in plant and soil-available N indicates a shift of main plant N resources to NO₃⁻ from NH₄⁺ along the N leaching gradient. *Rapid communications in mass spectrometry*, **24** (7), 1001-1008.

Van Allen, J. A. and Frank, L.A. (1959). Radiation around the earth to a radial distance of 107,400 km. *Nature*, **183**, 430-434.