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

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ABSTRACT: Cosmic rays in the upper troposphere (9000 meters to 15000 meters) initiate the following nuclear chemical reaction: $1n + 14N \Rightarrow 14C + 1H$. 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 15/14N 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 15/14N in foliage. Furthermore, the average value of delta 15/14N in the nitric-oxide substances in the aerosol samples in Fairbanks was -2.70 [per mil], whereas the average value in Singapore and Fairbanks, it was observed that values of delta 15/14N in nitric-oxide substances from aerosols were correlated with declination. The value of delta 15/14N in nitric-oxide substances from aerosols were correlated with declination. The value of delta 15/14N in nitric-oxide substances from aerosols were correlated with increasing declination due to more active conversions from ¹⁴N to ¹⁴C 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):

$${}^{1}_{0}n + {}^{14}_{7}N \rightarrow {}^{14}_{6}C + {}^{1}_{1}H$$
 (1)

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

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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 SST 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 ${}^{15}N/{}^{14}N$ and $\delta^{18}O/{}^{16}O$ isotope ratios in NO, were measured using the denitrifier method (Casciotti et al., 2002; Takebayashi et al., 2010). The NO, was converted to N₂O using a denitrifier (*Pseudomonas aureofaciens*; ATCC 13985) lacking N₂O reductase. The N₂O 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 KOBA), 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)

$$\left[\frac{RSAMPLE-RSTANDARD}{RSTANDARD}\right] \times 1000^{\circ} / ^{\circ\circ} = \delta = delta$$

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 nitricoxide 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 18/160 in the nitricoxide 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 ¹⁴N is more easily broken than ¹⁵N. The proton number and the neutron number of ¹⁴N are both 7. The proton number of ¹⁵N is also 7, but its neutron number is 8 (Table 4). Thus a nuclear chemical reaction in which ¹⁴N and a neutron (n) produce the ¹⁴C 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 ¹⁵N and a neutron (n) because ¹⁵N is more tightly bound than ¹⁴N (due to the fact that ¹⁵N has a neutron number of 8). Therefore only ¹⁴N is affected by neutron bombardment from the sun and from cosmic rays. Thus ¹⁴N is converted into the ¹⁴C radioisotope and the relative amount of ¹⁵N 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 ¹⁴N to ¹⁴C 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.

Sampling Date & Time	815N/14N n	er 8180/1601 per	NO3- Micro	NO2-1 Micro	F-1 Micro Mol / Ch	- Micro Mol	SO4 2-1 Micro	PO43-1 Micro
(Singapore Time= SGT = UTC + 8 hours)	mil	mil]	Mol/L]	Mol / L]	L]	/L]	Mol/L]	Mol/L]
1527 SGT 16 to 1527 SGT 17 NOV	27.19	56.16	1.18	0.96	N/A	2.28	1.22	N/A
2009 1539 SGT 17 to 1539 SGT 18 NOV 2000	9.02	62.37	1.2	0.54	N/A	2.35	1.75	N/A
2002 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	7.78	21.17	0.23	N/A	N/A	0.7	N/A	N/A
2009 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	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
Stand ard 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 Total	27.19 70.03	62.37 413.68	17.89 46.21	2.17 6.5	2.648 4.429	30.48 92.34	22.65 56.03	2.49 2.49
Number of Sample	6	6	6	9	4	10	L	1

Effect of Stable Isotope Ratios in Foliage

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Sampling Date & Time (Alaska Standard Time = AST & = UTC - 9 hours; Alaska Daylight Saving Time = ADT = UTC - 8 hours;)	315N/14N [per mil]	8180/160 [permil]	N O3- [M icr o M ol/L]	NO2- [Micro MoVL]	F- [M icr o M ol/L]	Cl- [Micro Mol/L]	SO 4 2- [M icro M oVL]	PO4 3- [Micro MoVL]
1414 AST 02 to 1414 AST 03 JAN 2010	-24.1	35.6	\mathbf{N}/\mathbf{A}	N/A	0.18	0.48	N/A	1.2
1417 AST 03 to 1417 AST 04 JAN 2010	-9.3	26.9	0.26	N/A	1.73	N/A	N/A	N/A
1421 AST 04 to 1421 AST 05 JAN 2010	-11.3	43.7	0.22	N/A	N/A	0.72	N/A	1.68
1711 ADT 01 to 1710 ADT 03 APR 2010	2.4	29.9	2.05	N/A	1.33	11.07	1.34	N/A
1712 ADT 03 to 1712 ADT 05 APR 2010	5.1	38.8	0.93	N/A	1.07	11.17	1.72	N/A
1714 ADT 05 to 1714 ADT 07 APR 2010	1.8	28.6	1.2.1	N/A	0.0054	5.06	N/A	N/A
1716 ADT 07 to 1716 ADT 09 APR 2010	N/A	N/A	N/A	N/A	0.021	3.15	0.53	N/A
1010 ADT 21 to 1010 ADT 23 SEP 2010	2.4	70	\mathbf{N}/\mathbf{A}	N/A	N/A	1.1	N/A	N/A
1012 ADT 23 to 1012 ADT 25 SEP 2010	4.1	56.3	\mathbf{N}/\mathbf{A}	N/A	N/A	0.52	N/A	N/A
1014 ADT 25 to 1014 ADT 27 SEP 2010	4.6	57	N/A	N/A	N/A	N/A	N/A	N/A
A ve rage	-2.7	42.977778	0.934	N/A	0.72273	4.15875	1.19667	1.44
Standard Deviation	10.05062	15.053885	0.7559	N/A	0.74907	4.57801	0.60781	0.33941
Variance	101.015	226.61944	0.5714	N/A	0.5611	20.9582	0.36943	0.1152
Range	29.2	43.1	1.83	0	1.7246	10.69	1.19	0.48
Minim um Value.	-24.1	26.9	0.22	0	0.0054	0.48	0.53	1.2
Maximum Value	5.1	70	2.05	0	1.73	11.17	1.72	1.68
Total	-24.3	386.8	4.67	0	4.3364	33.27	3.59	2.88
Number of Sample	6	6	S	0	9	8	3	2

minute; Altit	tude: 67.0 me	ter), Singar	oore and Fa	irbanks Interna meter) F	utional Ai airbanks	trport (N	: 64 Degre 1.S.ACoi	e 50.116] ntinues	Minute, ¹	W: 147 D(egree 49.747	Minute, Alt	titude: 143.4
Sampling Place, Date & Time (Sing apore Time = SGT = UTC+8 hours) (Alaska Standard Time = AST = UTC - 9 hours; Alaska Dayli ght Saving Time = ADT = UTC - 8 hours;)	Date	[degræ]	Maximum Temperature in Sampling Place [degree Celsius]	Minimum Temperature in Sampling Place [degree Celsius]	Length & Of of daytime i daytime i in / in / in / Place Place [hour]	si 5N/14N8 [per mil][in NOx of i Aerosol in/ Farbank s]	180/1608 per mil] [n NOx ofir Aerosol inA Farbanks S	ISN/14N8 per mil][iNOx of in erosol in S S S	I80/1608 per mil][n NOx of ii Aerosol in ingapore F	13C/12C a needles (foliage) of Abics A ifrma in airbanks	ber mil] in] per mil] in] needles (foliage) of bies firma in Fairbanks	 ðl 3C/12C[per mil] in foliage of Samanea s saman (Rain tree) in Singapore 	õl 5N/14N [per mil] in foliage of Samanca aman (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	NA	N/A
1527 SGT 16 to 1527 SGT 17 NOV 2009 in Singapore	1 7-Nov-09	S 18.95	30.17	24.16	12.05	NA	N/A	27.19	56.16	N/A	N/A	N/A	N/A
1539 SGT 17 to 1539 SGT 18 NOV 2009 in Singapore	1 8-Nov-09	S 19.20	29.79	25.38	12.05	NA	N/A	9.02	62.37	N/A	N/A	N/A	N/A
1542 SGT 18 to 1542 SGT 19 NOV 2009 in Singapore	1 9-Nov-09	S 19.43	28.4	2.2.86	12.05	NA	N/A	21.36	25.54	N/A	N/A	N/A	N/A
1818 SGT 19 to 1818 SGT 20 NOV 2009 in Singapore	20-Nov-09	S 19.67	27.62	23.33	12.05	NA	N/A	7.78	21.17	N/A	N/A	N/A	N/A
1908 SGT 21 to 1908 SGT 22 NOV 2009 in Singanore	22-Nov-09	S 20.10	29.37	24.4	12.05	NA	N/A	16.64	41.91	N/A	N/A	N/A	N/A
1414 AST 02 to 1414 AST 03 JAI 2010 in Fairbanks	3-Jan-10 N	S 22.85	-31.1	-36.7	4.13	-24.11	35.63	N/A	N/A	N/A	N/A	N/A	N/A

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Table 3. Stable isotope ratios in aerosol (PM 4.5) (5L/Minite) and foliage at National University of Singapore (N: 1 degree 18 minute; E: 103 degree 46

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minute; Altitu	de: 67.0 meter),	Singapore and	Fairbanks Inter meter	rnational Ai) Fairbanks	rport (1 Alaska	V: 64 D U.S.A.	egree 50. -Continu	116 Minut les	e, W: 147]	Degree '	49.747 Mi	inute, Altitud	le: 143.4
1417 AST 03 to 1417 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	NA	N/A	N/A
1421 AST 04 to 1421 AST 05 JAN 2010 in Fairbarks	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
1709 SGT 08 to 1709 SGT 12 FEB 2010 in Singapore; (Official CHINESE NEW YEAR was from SUN 14 to TUE	12.Feb.10	S 13.78	31.74	24.92	12.07	N/A	N/A	-6.97	56.14	N/A	MA	N/A	N/A
16 HEB 2010) 1712 SGT 12 to 1712 SGT 16 FEB 2010 in	16-Feb-10	S 12.43	30.53	25.57	12.07	N/A	N/A	66. <i>T</i> -	56.75	N/A	NA	-15.2	2.6
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
1713 SGT 16 to 1713 SGT 20 FEB 2010in Sincerood	20-Feb-10	S 11.03	29.01	25.81	12.07	N/A	N/A	-6.74	59.18	N/A	NA	N/A	N/A
Augatore 1711ADT 01 to 1710 ADT 03 APR 2010 in Fairbarks	3-Apr-10	N 5.18	7.8	-6.7	13.85	2.43	29.93	N/A	N/A	N/A	NA	ΝΑ	N/A
1712 ADT 03 to 1712 ADT 05 APR 2010 in Fairbarks	5-Apr-10	N 5.93	1.11	-6.1	14.07	5.1	38.77	N/A	NA	N/A	NA	ΝΑ	N/A
Fairbanks	6-Apr-10	N 6.32	6.1	-2.2	14.18	N/A	N/A	N/A	NA	-14.3	-6.4	N/A	N/A
1714 ADT 05 to 1714 ADT 07 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

322 SGT 16 to 322 SGT 20 JUL	010 in Singapore 325 SGT 20 to 325 SGT 24 JUL	o io in Singapore Singapore	325 SGT 24 to 325 SGT 28 JUL 010 in Sing apore	⁷ ai rbanks	010 ADT 21 to 010 ADT 23 SEP 010 in Fairbanks	⁷ ai rbanks 012 ADT 23 to 012 ADT 25 SEP 010 in Faibanks	1014 ADT 25 to 014 ADT 27 SEP 010 in Fairbanks								
20-Jul-10	24-Jul-10	27-Jul-10	28-Jul-10	22-Sep-10	23-Sep-10	24-Sep-10 25-Sep-10	27-Sep-10								
N 20.71	N 19.91 N	N 19.27	N 19.05	N 0.43	N 0.05	S 0.33 S 0.72	S 1.50								
29.33	27.99	28.74	27.56	9.4	8.3	6.1 6.7	3.3								
23.71	23.9	25.9	23.5	0	-2.8	-2.8 -5	-8.9	A	ΝЦ	~	R	47		Ξ	20
12.18	12.18	12.16	12.16	12.26	12.17	12.05 11.95	11.72	Average	Standard Deviation	Variance	Range	/linimum /ahre	Maximum Zalue	Total	Number of Namole
N/A	N/A	N/A	N/A	N/A	2.38	N/A 4.11	4.6	-2.6967	10.0466	100.935	29.21	-24.11	5.1	-24.27	6
N/A	N/A	N/A	N/A	N/A	66.69	N/A 56.29	57	42.9678	15.0538	226.618	43.12	26.87	66.69	386.71	6
9.74	8.03	N/A	5.67	N/A	N/A	N/A N/A	N/A	7.61182	11.5167	132.633	35.18	-7.99	27.19	83.73	11
34.46	35.11	N/A	61.12	N/A	N/A	N/A N/A	N/A	46.3555	15.1465	229.415	41.2	21.17	62.37	509.91	11
N/A	N/A	N/A	N/A	-28.8	-28.8	-29.4 -30.1	N/A	-23.186	7.6693	58.8181	16.2	-30.1	-13.9	-162.3	L
N/A	N/A	N/A	NA	-0.5	-0.6	0.1 -0.2	N/A	-1.84286	3.45 198	11.9162	8.9	-7.1	1.8	- 12.9	L
N/A	-32.7	-32.7	N/A	N/A	N/A	N/A N/A	N/A	-24.1	9.933445	98.67333	17.5	-32.7	-15.2	-96.4	4
N/A	-1.1	-1.6	N/A	N/A	N/A	N/A N/A	N/A	-1.3	3.150661	9.926667	T.T	-5.1	2.6	-5.2	4

Table 3. Stable isotope ratios in aerosol (Continuation)

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Effect of Stable Isotope Ratios in Foliage

Stable Isotope	12C	13C	14N	15N	160	180
Atomic Number = Proton Nmber	6	6	7	7	8	8
Mass Number	12	13	14	15	16	18
Neutron Number	6	7	7	8	8	10

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

Therefore conversion from ¹⁴N into ¹⁴C decreases, and thus the relative amount of ¹⁵N 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 ¹⁵N 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 15/14N in nitric-oxide substances within the aerosol samples.

Fig. 2 shows a significant positive linear relationship between declination and delta 15/14N 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 higherlatitude regions such as Fairbanks, declination has a greater effect on maximum and minimum atmospheric temperature and day length than it does in lowerlatitude 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 15/14N in nitric-oxide substances within the aerosol samples in Fairbanks increased with increasing declination, due to more active conversions from ¹⁴N to ¹⁴C 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)



Maximum Teperture [degree Celsius] = x, R = Coefficient of Correlation

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



Minimum Temperture [degree Celsius] = x, R=Coefficient of Correlation

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 18/160 in the nitric-oxide substances within the aerosol samples in Fairbanks was +42.97 [per mil] whereas average value of delta 18/160 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/ 160 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 ¹⁴N to ¹⁴C by neutron bombardment.

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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). Artifical 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 NOx in the Arctic Atmosphere Using Stable Isotopes in Nitrate. Science, **322** (**5902**), 730–732.

Takebayashi, Y. (2010). The natural abundance of 15N in plant and soil-available N indicates a shift of main plant N resources to NO_3 - from NH_4^+ 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.