

Article

Source Identification of PM_{2.5} during a Smoke Haze Period in Chiang Mai, Thailand, Using Stable Carbon and Nitrogen Isotopes

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Abstract: Open biomass burning (BB) has contributed severely to the ambient levels of particulate matter of less than 2.5 μm diameter (PM_{2.5}) in upper northern Thailand over the last decade. Some methods have been reported to identify the sources of burning using chemical compositions, i.e., ions, metals, polycyclic aromatic hydrocarbons, etc. However, recent advances in nuclear techniques have been limited in use due to their specific instrumentation. The aims of this study were to investigate the sources of ambient PM_{2.5} in Chiang Mai city using stable carbon (δ¹³C) and nitrogen isotopes (δ¹⁵N). The mean concentrations of total carbon (TC) and total nitrogen (TN) in PM_{2.5} were 12.2 ± 5.42 and 1.91 ± 1.07 μg/m³, respectively, whereas δ¹³C and δ¹⁵N PM_{2.5} were −26.1 ± 0.77‰ and 10.3 ± 2.86‰, respectively. This isotopic analysis confirmed that biomass burning was the source of PM_{2.5} and that C₃ and C₄ plants contributed about 74% and 26%, respectively. These study results confirm that the stable isotope is an important tool in identifying the sources of aerosols.

Keywords: biomass burning; PM_{2.5}; stable isotope; C₃ plants



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1. Introduction

Biomass burning (BB) aerosol plumes with combustion from forest fires or agriculture burns have caused severe air pollution [1–6]. BB is the second largest source of trace gases and the largest source of primary fine carbonaceous particles. BB can happen naturally or anthropogenically (e.g., forest fires, burning of crop residues). BB is a major source of several ambient particles and trace gases that influence the concentration of ozone at ground levels [7]. Previous studies of emissions from biomass burning have used several chemical compositions in aerosol as a tracer such as organic carbon, elemental carbon, stable carbon and nitrogen isotopes, ion species, etc. [7–13]. Chiang Mai city is one of most productive agricultural areas in northern Thailand. There have been few studies that have reported the chemical and isotopic compositions of biomass burning aerosols in this region, even though burning agricultural residues is a very common practice during the harvest in dry season from February to May [14–19] as shown by the Moderate Resolution Imaging Spectroradiometer (MODIS) fire count data. The stable carbon (δ¹³C) and nitrogen (δ¹⁵N) composition of bulk aerosols have been used as geochemical markers to identify aerosol sources in a variety of environments [8–10,20,21]. Likewise, the studies of δ¹³C and δ¹⁵N in bulk aerosols have provided more information on the multiple sources and atmospheric processing of organic aerosols, such as biomass and biofuel burning [12,22–25]. δ¹³C and δ¹⁵N in ambient particulate matter have been used to great effect in previous studies to distinguish the aerosols emitted from the burning of C₃ and C₄ plants and evaluate the total contribution of sources from biomass and biofuel combustion [10–13,26–28].

Biomass burning is responsible for more than 90% of organic carbon (OC) in the atmosphere [29]. Previous studies concluded that the dominant source of global black carbon (BC) was BB, and recent studies reported that BB was an important source of brown carbon [29]. Biomass burning particles could impact the climate as well as human health [30,31]. Previous laboratory studies [32] reported that the concentrations of organic aerosol from wood burning increased by a factor of 1.5 to 2.8 after several hours of exposure to UV light, indicating a large amount of secondary organic aerosol (SOA) generated from BB emission. Several tracers have been used in the BB research [13,27,28], such as anhydro-sugars, resin acids [33,34], and water-soluble non-sea-salt potassium (nss-K^+) [35–37].

Air pollution is a major environmental problem in Chiang Mai province. Open biomass burning in agricultural and forest areas causes significant smoke and haze from February to April, according to satellite hotspot data. During this period of time, the maximum concentrations of $\text{PM}_{2.5}$ are 2–4 times higher than the standard and may pose a health risk to residents due to over-exposure to airborne particulates and their chemicals [38,39]. The Thailand-Myanmar northwest border, extending from Tak to Chiang Mai, and the Thailand-Laos border have a high density of hotspots. There is a high density of active fire hotspots near the Thailand-Myanmar border, especially in the southwest direction where Mae Hong Son province extends to Chiang Mai province [15–18]. Several previous methods, such as those involving ions, metals, and polycyclic aromatic hydrocarbons (PAHs), have been reported to identify the burning sources, although recent advances in nuclear techniques are still limited to report due to their specific instrumentation. The aims of this study were to investigate and identify the possible sources of ambient $\text{PM}_{2.5}$ in Chiang Mai city using stable carbon and nitrogen isotopes.

2. Materials and Methods

2.1. Sampling Site

The study site was located at the Research Institute for Health Sciences (RIHES) on the main campus of Chiang Mai University (CMU; $18^\circ 47' \text{ N}$, $98^\circ 57' \text{ E}$), Chiang Mai city (Figure 1). The $\text{PM}_{2.5}$ samples were collected from the rooftop of the four-story building, which was about 30 m above the ground. Daily samples were collected on quartz filters (Pall Life Sciences, Port Washington, NY, USA) using a medium air volume sampler (KC-6120, Qingdao Laoying Ltd., Qingdao, China) with a flow rate of 100 L/min. The $\text{PM}_{2.5}$ samples were collected from January to May 2017. Before collecting the samples, the quartz filters were heated in the oven for 6 h at 450°C to remove carbon contamination. The quartz filters were wrapped in aluminum foil and stored in desiccators filled with silica gel before and after the sampling. The condition of the filter before and after sampling was considered in a controlled room at a temperature of $25^\circ \text{C} \pm 2^\circ \text{C}$ and a relative humidity of less than 40%. Before analysis, the filters were kept in the freezer ($\leq 20^\circ \text{C}$).

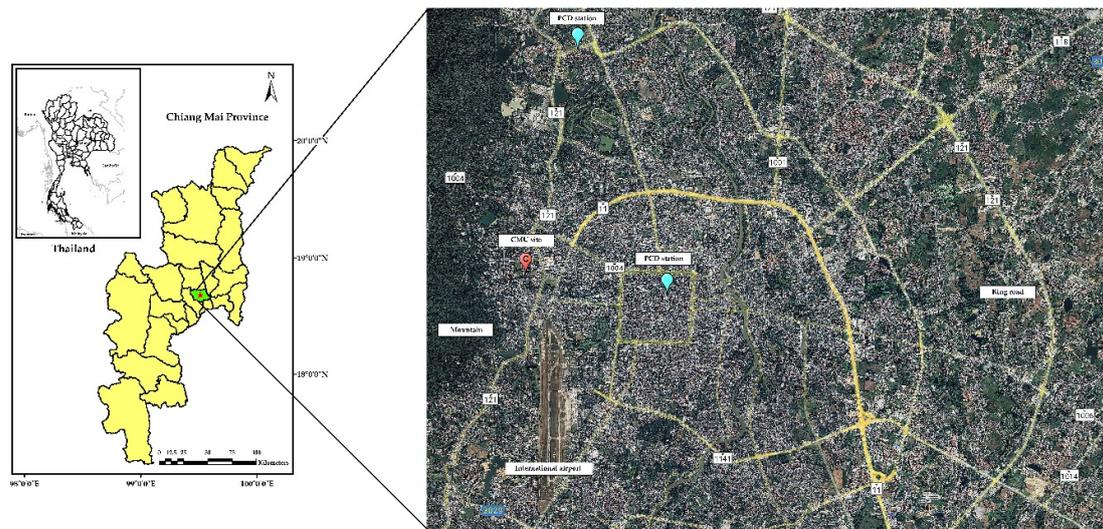


Figure 1. The location of the sampling site in Chiang Mai city.

2.2. Measurement of Stable Isotopes

The stable carbon and nitrogen isotopes were measured by the elemental analyzer (EA, FLASH, 2000; Thermo Fisher Scientific, Waltham, MA, USA) coupled with an isotope ratio mass spectrometer (MAT253, Thermo Fisher Scientific, Waltham, MA, USA) at Yale-NUIST Center on Atmospheric Environment, NUIST, Nanjing, China. The detection limit of this method is 5 μg to 500 μg . The punched-out quartz filter (with a diameter of 17 mm) was folded and placed into a tin cup. After the tin cup was folded up, the quartz filter was tightly sealed in a relatively clean room. Then, the loaded tin cup was placed into a reaction tube combusted at 1000 $^{\circ}\text{C}$ by an autosampler, wherein conversion to CO_2 and TN 's conversion to N_2 occurred. Separated by a gas chromatographic column, N_2 and CO_2 were transferred for IRMS by Con-Flo. Ratios were reported in parts per thousand (‰) written in delta (δ) notation, related to the international standard Vienna Peedee belemnite (V-PDB) for atmospheric carbon and nitrogen, and defined as:

$$\delta^{13}\text{C} (\text{‰}) = \left[\frac{(^{13}\text{C}/^{12}\text{C})_{\text{sample}}}{(^{13}\text{C}/^{12}\text{C})_{\text{standard}}} - 1 \right] \times 1000 \quad (1)$$

$$\delta^{15}\text{N} (\text{‰}) = \left[\frac{(^{15}\text{N}/^{14}\text{N})_{\text{sample}}}{(^{15}\text{N}/^{14}\text{N})_{\text{standard}}} - 1 \right] \times 1000 \quad (2)$$

The program transformed the ion beam intensity (Mv) of sample gas into an isotope ratio by comparing it to reference gases with known isotope values (CO_2 : $\delta^{13}\text{C} = -31.9\text{‰}$; N_2 : $\delta^{15}\text{N} = -1.17\text{‰}$). This study's analytical precision of measurement (reported as relative standard deviation) was 0.02‰ for $\delta^{13}\text{C}$ and 0.14‰ for $\delta^{15}\text{N}$, which was acceptable [40]. Before each batch of testing, a leak test and three empty tests (baseline scan with no sample) were performed. Three on/off tests (continuous tests of reference gas in one scanning period) were performed to ensure the instrument's stability. Three blank tests (tests with an empty tin cup) were used along with each batch of tests to correct for impurity interferences in the analysis. As external standards, three standards with known isotope ratios (acetanilide#1: $\delta^{13}\text{C} = -29.5\text{‰}$, $\delta^{15}\text{N} = 1.18\text{‰}$; acetanilide#2: $\delta^{13}\text{C} = -29.5\text{‰}$, $\delta^{15}\text{N} = 19.6\text{‰}$; sucrose: $\delta^{13}\text{C} = -12.2\text{‰}$) were employed.

2.3. Data Analysis

MODIS onboard NASA's Aqua and Terra satellites calculated the number of hotspots during the intensive biomass burning season. All data collected from the experiment and analyzed using SPSS statistical program are expressed as mean and standard deviations (S.D.). Moreover, a correlation between $\text{PM}_{2.5}$ concentrations and chemical compositions was also calculated using Pearson correlation analysis.

3. Results and Discussion

3.1. PM_{2.5} Mass Concentration

The daily PM_{2.5} samples were collected from 12 January to 30 May 2017 at the Research Institute for Health Sciences, Chiang Mai University, the Chiang Mai city site. PM_{2.5} concentrations were obtained from the gravimetric method and compared with the values of PM_{2.5} from the air quality monitoring station operated by the Pollution Control Department (PCD) in Chiang Mai city. The PM_{2.5} concentration trends were correlated to the active fire data (number of hotspots) detected by satellite remote sensing imaging from the Moderate Resolution Imaging Spectroradiometer (MODIS); the trends were collected and analyzed with statistical methods to confirm the significant reasons for the air pollution in Chiang Mai city in this study. From January to May 2017, there were 365 hotspots in Chiang Mai province, with a peak in March (250 hotspots) and a highest daily average PM_{2.5} concentration of $45.3 \pm 6.17 \mu\text{g}/\text{m}^3$. In this study, the average PM_{2.5} concentrations ranged from 6.07 to $79.9 \mu\text{g}/\text{m}^3$, averaging $35.8 \pm 16.3 \mu\text{g}/\text{m}^3$. The average values were lower than those collected at the Chiang Mai University campus in 2016 ($64.3 \pm 17.6 \mu\text{g}/\text{m}^3$), monitored during the same period [19], and lower than those at the CMU site in 2016, where the mean was $44.5 \pm 32.1 \mu\text{g}/\text{m}^3$ [14]. We observed a peak in PM_{2.5} on 14 May 2017 ($79.9 \mu\text{g}/\text{m}^3$) after the no-burning policy was implemented from 20 February to 20 April 2017. There was no observed number of hotspots during this period. Moreover, nine out of sixty-three days' daily PM_{2.5} concentrations in 2017 exceeded the Pollution Control Department (PCD) standard ($50 \mu\text{g}/\text{m}^3$). The monthly average PM_{2.5} concentrations from January to May were 19.9, 39.6, 44.5, 51.2, and $23.1 \mu\text{g}/\text{m}^3$, respectively. The monthly concentration of PM_{2.5} reached its maximum in May and then declined at the beginning of the rainy season.

3.2. Temporal Variation in Stable Carbon and Nitrogen Isotopes in PM_{2.5}

The temporal variation in $\delta^{13}\text{C}$ in PM_{2.5} at Chiang Mai city during the sampling period ranged from -27.4‰ to -24.3‰ , with a mean value of -26.1‰ , as reported in Table 1.

Table 1. The concentrations of TC and TN ($\mu\text{g}/\text{m}^3$), their isotope ratios (‰), and TC/TN in PM_{2.5} from Chiang Mai city.

Parameter	January	February	March	April	May	Mean \pm S.D.
PM _{2.5} ($\mu\text{g}/\text{m}^3$)	19.9 ± 5.33	39.6 ± 9.92	44.5 ± 6.65	51.2 ± 11.2	23.1 ± 19.2	35.8 ± 16.3
TC ($\mu\text{g}/\text{m}^3$)	9.90 ± 1.84	14.7 ± 3.13	14.8 ± 4.25	15.3 ± 5.31	6.23 ± 4.91	12.0 ± 5.32
TN ($\mu\text{g}/\text{m}^3$)	1.10 ± 0.29	1.87 ± 0.71	1.90 ± 0.76	2.99 ± 0.76	1.76 ± 1.47	1.83 ± 0.89
TC/TN	9.23 ± 1.46	9.09 ± 4.21	7.47 ± 2.04	5.15 ± 1.64	3.85 ± 1.58	6.82 ± 3.15
$\delta^{13}\text{C}$ (‰)	-25.3 ± 0.46	-25.5 ± 0.79	-26.6 ± 0.53	-26.7 ± 0.39	-26.2 ± 0.42	-26.1 ± 0.77
$\delta^{15}\text{N}$ (‰)	7.77 ± 1.32	9.38 ± 2.25	10.5 ± 1.71	12.7 ± 1.61	11.1 ± 3.89	10.3 ± 0.86

The temporal variations in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, as well as the meteorological data, are shown in Figure 2. The meteorological data were obtained from the Chiang Mai meteorological station, located in Chiang Mai city, about 2 km away from the sampling site. Generally, the trend varied within a narrow range. The mean value of $\delta^{13}\text{C}$ was comparable to the reported $\delta^{13}\text{C}$ value ($-25.8 \pm 0.52\text{‰}$) measured in Brazil, where C₃ plant burning is predominant [12], and was similar to the values reported from Chiang Mai (Doi Ang Kang) during the biomass burning season in 2015 (mean value of $\delta^{13}\text{C}$: $-26.0 \pm 1.2\text{‰}$) [10]. Kawichai et al. (2020) reported a mean $\delta^{13}\text{C}$ value of $-27.9 \pm 0.67\text{‰}$ (collected from February to August 2016), although the $\delta^{13}\text{C}$ value of this study was $-26.1 \pm 0.77\text{‰}$ (collected from January to May 2017). These two studies confirm that biomass burning is the major source of ambient PM_{2.5} in Chiang Mai city [14]. On the other hand, the $\delta^{13}\text{C}$ values were greater than the values measured at Prague-Suchdol in winter 2016 (mean values of -27.2‰) [41]. Furthermore, Martinsson et al. (2017) observed a mean value of -26.3‰ for $\delta^{13}\text{C}$, attributable to biogenic emissions from terrestrial plants [42]. Previous investigations have indicated significant differences. The $\delta^{13}\text{C}$ values in this study were lower than those

recorded at residential, commercial, and industrial locations in Mexico (-25.1‰ [43]) and slightly lower than the mean value of the $\delta^{13}\text{C}$ of TC in Taiyuan, China ($-24.3 \pm 1.0\text{‰}$). The different emission sources could explain these findings. In this study, the isotope abundance (-24.2‰), which is a signature of biomass burning, and emissions from C_3 plant combustion were compared and the values of $\delta^{13}\text{C}$ varied from -34.0‰ to -23.0‰ , with the highest $\delta^{13}\text{C}$ values in $\text{PM}_{2.5}$. Biomass burning has been observed to range from -22.0‰ to -8.0‰ [12,28,44,45].

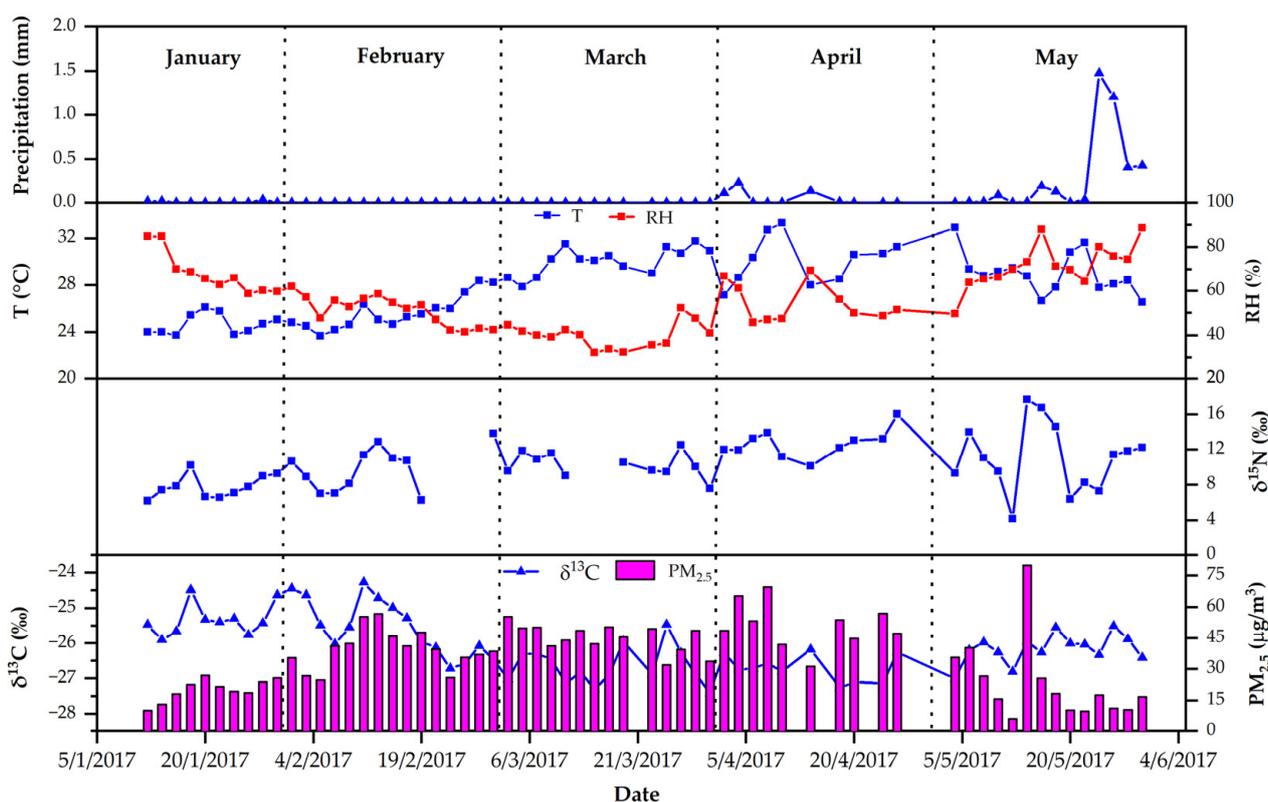


Figure 2. Temporal variation in stable isotopic and meteorological parameters during the sampling period in Chiang Mai city.

It was found that the $\delta^{13}\text{C}$ value was highest in January ($-25.3 \pm 0.45\text{‰}$), followed by February ($-25.5 \pm 0.81\text{‰}$), May ($-26.2 \pm 0.39\text{‰}$), March, and April, as shown in Figure 3. The values of $\delta^{13}\text{C}$ were more stable and showed very small differences in this study. The highest $\delta^{13}\text{C}$ values were found in January and February, significantly higher than in the other months. This indicates C_3 plant combustion to be the main contributor to fine aerosol [12]. In a previous study, high values of $\delta^{13}\text{C}$ (-26.0‰ to -23.2‰ , with an average of -25.2‰) were observed in wheat residue burning aerosols collected from Mt. Tai, China [22]. During the sampling period, it was confirmed that biomass burning from agriculture, such as wheat and corn residue on high land, is a dominant source in the region. According to observation, the highest value (-24.3‰) of $\delta^{13}\text{C}$ was recorded on 11 February 2017. The mean value was -25.2‰ from 12 January 2017 (the start of sampling) to 20 April 2017 (the end of the no-burning policy). After the local government ceased burning (after 20 February 2017), the lowest value of $\delta^{13}\text{C}$ was recorded on 31 March 2017, at -27.2‰ . The average value of $\delta^{13}\text{C}$ was lower than the previous period, which was -26.5‰ . The change in $\delta^{13}\text{C}$ in this study can be explained by emissions and a changing aerosol source, observed from meteorological reports.

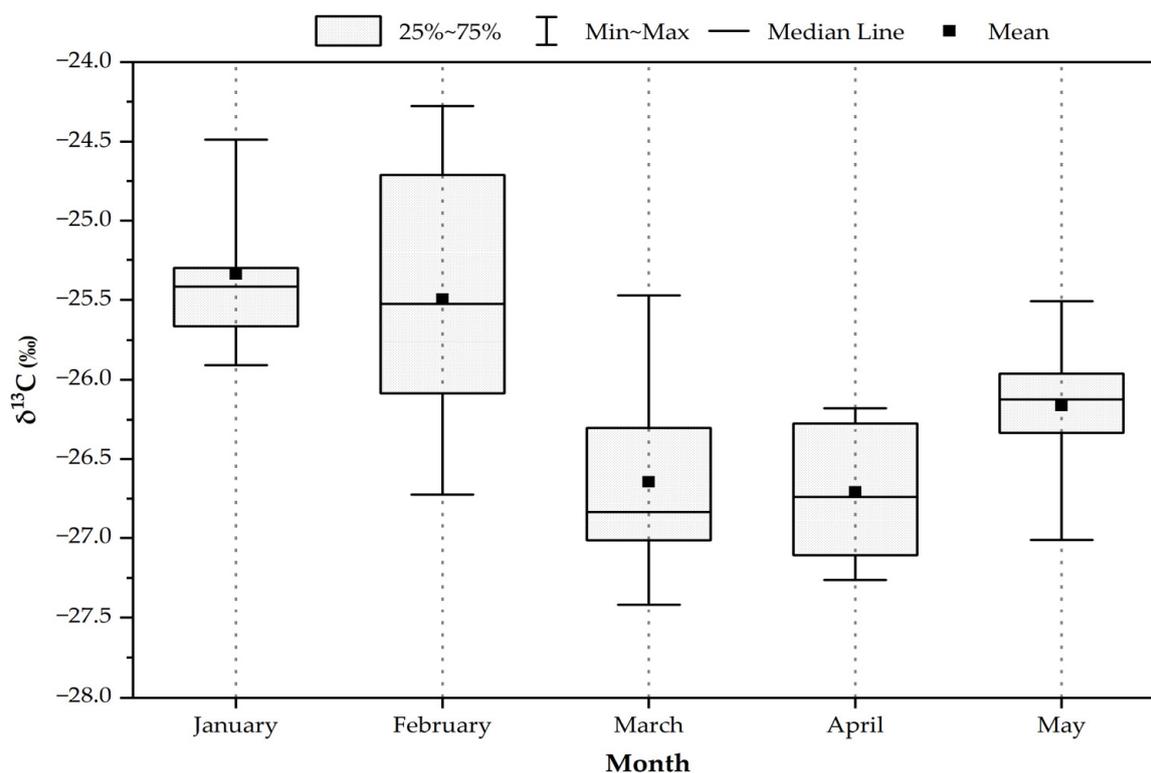


Figure 3. The box plots of stable carbon isotopes during the sample period in Chiang Mai city.

In this study, the $\delta^{15}\text{N}$ value ranged from 4.15‰ to 17.7‰ (mean: $10.3 \pm 2.86\text{‰}$), which was similar to that of aerosols collected from the Santarem region [15] ($10.4 \pm 1.86\text{‰}$), an area with a high density of primary tropical rainforest. In January, February, March, April, and May, the mean \pm S.D. $\delta^{15}\text{N}$ values were $7.76 \pm 1.32\text{‰}$, $9.37 \pm 2.24\text{‰}$, $10.5 \pm 1.71\text{‰}$, $12.7 \pm 1.61\text{‰}$, and $11.0 \pm 2.24\text{‰}$, respectively. The highest $\delta^{15}\text{N}$ values found in April are similar to those in Okinawa, Japan ($12.2 \pm 2.2\text{‰}$) [46], which are slightly lower than those ($15.1 \pm 3.4\text{‰}$) of Gosan aerosol [26]. This location is in the tropical rainforest, with a few pasture regions in the northeast of the Amazon basin, and biomass burning may have an impact. On the other hand, the $\delta^{15}\text{N}$ values of TN from Cape Hedro, similar to the aerosol samples from Piracicaba, Brazil, were 10.6‰ [12], and the $\delta^{15}\text{N}$ value in these studies was slightly higher than the $\delta^{15}\text{N}$ value ($4.6 \pm 0.8\text{‰}$) in TN reported by Widory (2007), implying that the aerosol was derived from diesel engine combustion, with diesel oil varying from 3.9‰ to 5.4‰ [25].

The linear regression between $\delta^{13}\text{C}$ and temperature had a moderate correlation, with a correlation coefficient of -0.6747 ($p = 0.000$), whereas a low correlation was found between $\delta^{15}\text{N}$ and temperature, with a correlation coefficient of 0.3284 ($p = 0.0145$) (shown in Figure 4). The results for $\delta^{13}\text{C}$ were more scattered than those for $\delta^{15}\text{N}$, indicating that the isotopic composition of $\delta^{13}\text{C}$ tended to be temperature-dependent [41,47]. The lowest temperatures in Chiang Mai city were 24.7 ± 0.86 and 25.4 ± 1.27 °C in January and February, respectively, whereas the highest $\delta^{13}\text{C}$ values occurred during this period. This discovery indicated that the combustion of C_4 plants was responsible for the highest $\delta^{13}\text{C}$ values [12]. In contrast, over the same period, the $\delta^{15}\text{N}$ values were lower than those from March to May, when the temperature was higher (26.5 to 33.2 °C), suggesting that the high temperature may have driven the $\delta^{15}\text{N}$ increases.

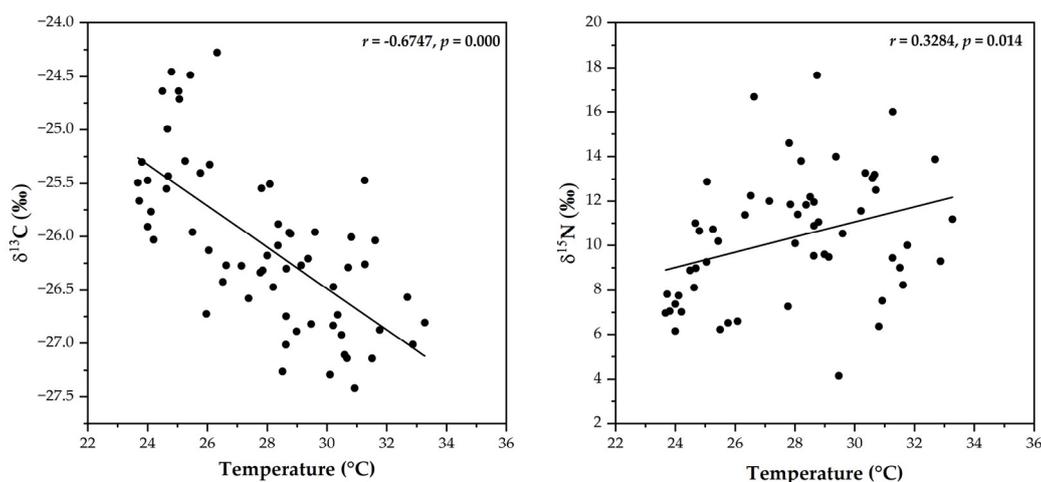


Figure 4. Relationship between stable isotopes and temperature in Chiang Mai city.

Furthermore, the mean TC concentration in Chiang Mai was $12.6 \pm 3.54 \mu\text{g}/\text{m}^3$ with a range from $5.42 \mu\text{g}/\text{m}^3$ to $22.9 \mu\text{g}/\text{m}^3$, which is equivalent to that reported for biomass burning by Boreddy et al. in DAK, Thailand, where biomass burning emissions varied from $1.5 \mu\text{g}/\text{m}^3$ to $80.3 \mu\text{g}/\text{m}^3$ [10]. In addition, on March 13, an extremely high concentration of TC was recorded, with a value of $25.1 \mu\text{g}/\text{m}^3$. There were 54 hotspots monitored on this day, implying that BB could be the potential source of TC throughout the sampling period. The correlation between $\text{PM}_{2.5}$ concentrations and TC was 0.8153 ($p < 0.001$). There was no correlation found between $\delta^{13}\text{C}$ and TC concentrations in these studies. Moreover, the concentrations of TN varied from 0.64 to $6.32 \mu\text{g}/\text{m}^3$ (mean: $1.91 \pm 1.07 \mu\text{g}/\text{m}^3$). These results show that the higher contributions in this study are lower than those of Varanasi and Kolkata, India (range: $14.9 \pm 10.8 \mu\text{g}/\text{m}^3$). The correlation between $\text{PM}_{2.5}$ concentrations and TN was moderate ($r = 0.6984$, $p < 0.001$). The results suggest that the sources of $\text{PM}_{2.5}$ were emissions from biomass burning. This result was supported by the moderate correlation between TC and TN ($r = 0.6109$, $p < 0.001$), as shown in Figure 5. The ratio of TC/TN in $\text{PM}_{2.5}$ ranged from 1.88 to 19.9, with a mean of 6.75. The higher TC/TN ratios in East Africa implied a higher organic aerosol contribution from biomass/biofuel combustion [20].

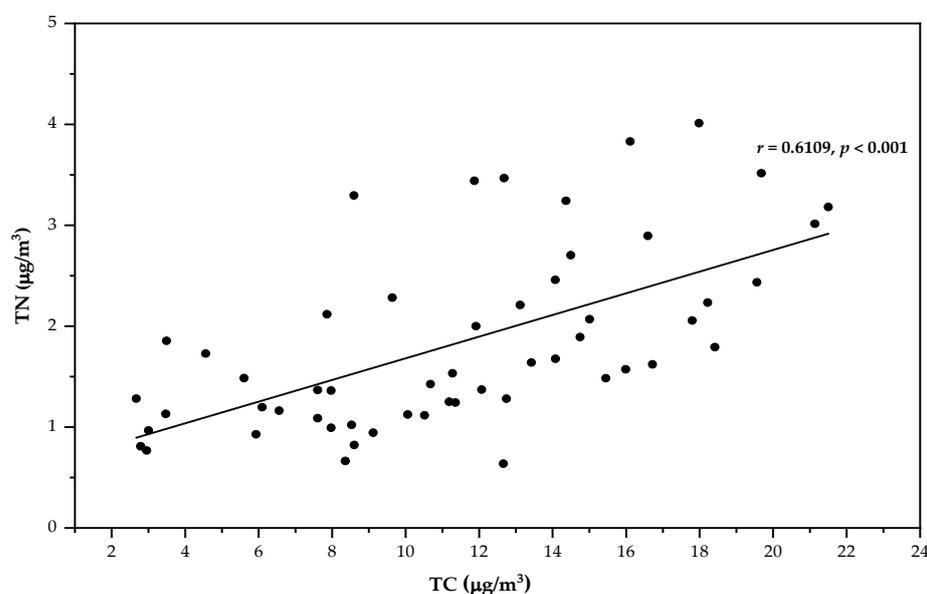


Figure 5. Correlation plot between TC and TN during the sample period from January to May in Chiang Mai city.

3.3. Contribution to PM_{2.5} from Burning C₃ and C₄ Plants

Many atmospheric studies use the value of $\delta^{13}\text{C}$ to distinguish between C₃ and C₄ plants. According to Martinelli et al. [12], a $\delta^{13}\text{C}$ value ranging from -26.9‰ to -24.9‰ suggests the domination of C₃ plants. In South Africa, Mkoma et al. (2013) found that the $\delta^{13}\text{C}$ value of biomass burning was dominated by C₄ plants and ranged from -24.4‰ to -22.4‰ [13]. The C₃ plants are isotopically ($\delta^{13}\text{C}$) different from C₄ plants, as reported by Bender, 1971 [48] and Smith and Epstein, 1971 [49]. The $\delta^{13}\text{C}$ values of C₃ plants are generally lower than those of C₄ plants due to different photosynthetic pathways [12]. The region's TC concentrations were linked to open burning emissions using MODIS fire spots in Chiang Mai city, supporting the emission sources identified in the current study. We estimated relative contributions from the burning of C₃ and C₄ plants to TC using the isotopic mass balance equation as follows:

$$\text{C}_3 \text{ plants} = \frac{(\delta^{13}\text{C}_{\text{aerosol}} - \delta^{13}\text{C}_{\text{C}_4\text{plant}})}{(\delta^{13}\text{C}_{\text{C}_3\text{plant}} - \delta^{13}\text{C}_{\text{C}_4\text{plant}})} \quad (3)$$

$$\text{C}_4 \text{ plants} = [1 - \text{C}_3 \text{ plants}] \quad (4)$$

The TC of aerosol was determined using the above equations, where $\delta^{13}\text{C}$ aerosol has a stable carbon isotope composition in total carbon ($\delta^{13}\text{C}_{\text{TC}}$) in the equation above and where $\delta^{13}\text{C}_{\text{aerosol}}$ is a stable carbon isotope composition of aerosol particles. The $\delta^{13}\text{C}_{\text{C}_3\text{plant}}$ and $\delta^{13}\text{C}_{\text{C}_4\text{plant}}$ are isotope ratios of C₃ and C₄ plants, respectively, whose end-members are defined to be 30.5‰ and 13.5‰ based on the average $\delta^{13}\text{C}$ values determined for leaves of C₃ and C₄ vegetation. A fraction of TC from C₃ and C₄ plants is referred to as $\delta^{13}\text{C}_{\text{C}_3\text{plant}}$ and $\delta^{13}\text{C}_{\text{C}_4\text{plant}}$, respectively. Based on the mean values of $\delta^{13}\text{C}$ found in the leaves of C₃ and C₄ vegetation in the Amazon basin [12], the values of $\delta^{13}\text{C}_{\text{C}_3\text{plant}}$ and $\delta^{13}\text{C}_{\text{C}_4\text{plant}}$ were assumed to be -31.5‰ and -13.5‰ , respectively. The mean contribution of C₃ and C₄ plants in this study was 74% (ranging from 63.4% to 81.9%) and 26% (ranging from 18.1% to 36.6%), respectively. Boreddy et al. (2018) reported the contribution of C₃ (74%) and C₄ (26%) plants to TC in DAK, Fang district, Chiang Mai province, which is about 170 km north of our study site in Chiang Mai city [10].

Further reports have indicated that $\delta^{13}\text{C}$ values from the open burning of C₃ plants are lower than those of C₄ plants [15,20,29]. The highest $\delta^{13}\text{C}$ values (from January to February) were associated with C₄ plants, suggesting that the primary combustion source might be related to maize residue burning at our sampling site. The burning of C₃ plants accounted for most of the reduction in $\delta^{13}\text{C}$ in aerosol; however, a similar decline in $\delta^{13}\text{C}$ was reported. Indonesia's forest fire contributed the most C₃ to the aerosol sample [50]. The association between $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values was consistent with that reported in prior investigations, as illustrated in Figure 6. The carbon and nitrogen isotopes in the current study indicated no significant correlation, implying that there are various aerosol C and N sources in Chiang Mai city. The $\delta^{13}\text{C}$ values in TC ranged from -27.4‰ to -24.3‰ in this study, whereas the $\delta^{15}\text{N}$ values in TN ranged from 4.15‰ to 17.7‰. The overlapping data over the sampling period, as shown in Figure 6, indicate that emissions were the primary source of TC. In addition, our region (northern Thailand) was dominated by the combustion of C₃ plants. However, this burning could provide a major source of nitrogen species during our sampling period. On the other hand, an important source, fossil fuel combustion, lowers the $\delta^{15}\text{N}$ levels in the ambient atmosphere [51]. The mean $\delta^{15}\text{N}$ values are lower in particulates from fossil fuel combustion, such as coal ($\sim 5.3\text{‰}$), diesel oil (3.9‰ to 5.4‰), and unleaded gasoline (4.6‰) [25,52]. C₃ plant burning, however, reported average $\delta^{15}\text{N}$ values of $13.2 \pm 4.2\text{‰}$ [12,53].

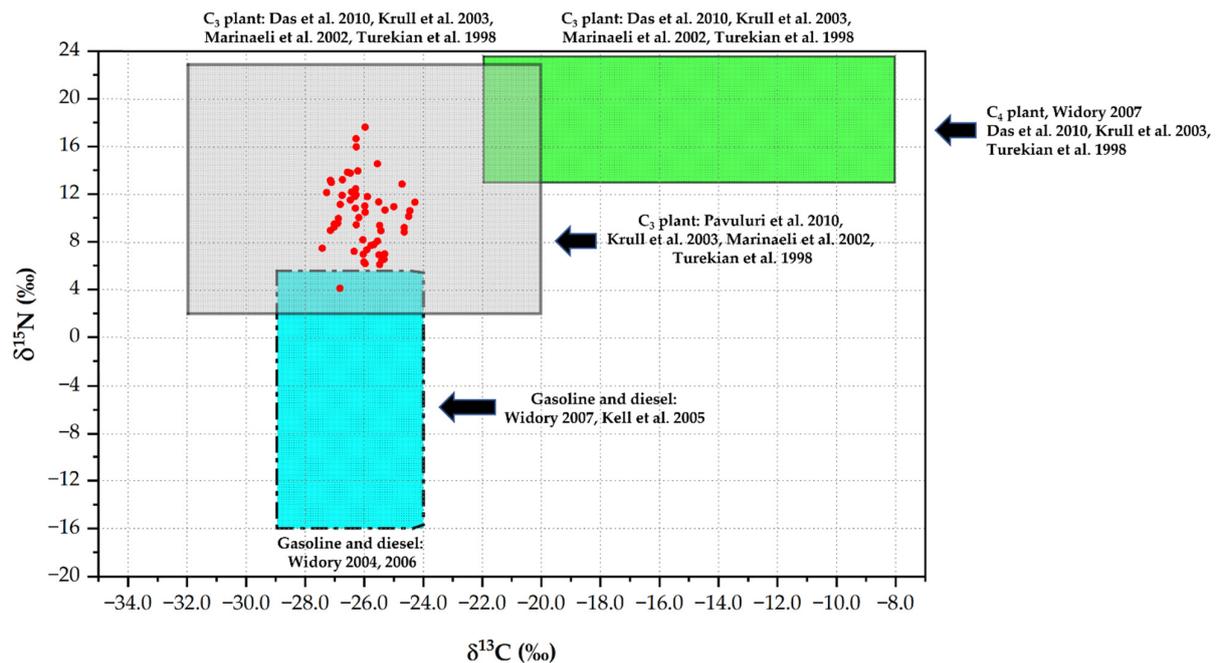


Figure 6. Relationship between $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ with sign level of potential sources in $\text{PM}_{2.5}$ [12,24,25, 28,44,45,51].

4. Conclusions

This study collected $\text{PM}_{2.5}$ samples from Chiang Mai city from January to May 2017 to investigate the $\text{PM}_{2.5}$ sources using stable carbon ($\delta^{13}\text{C}$) and nitrogen isotopes ($\delta^{15}\text{N}$). The mean value of $\delta^{13}\text{C}$ ranged from -27.4‰ to -24.3‰ , with a mean of -26.1‰ , whereas the $\delta^{15}\text{N}$ values ranged from 2.67‰ to 5.71‰ , with a mean of 10.3‰ , indicating that biomass burning was the primary source of $\text{PM}_{2.5}$, whereas fuel combustion was a minor source. Finally, the $\delta^{13}\text{C}$ data, reporting emissions from C_3 plant burning, confirmed that forest fires and agricultural waste were the primary sources of $\text{PM}_{2.5}$. C_4 plants, such as maize residue, were found to have been burned.

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