

Temporal distribution of atmospheric nitrogen deposition and correlation with biomass burning, Chiang Mai (Thailand) during 2008-2012

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Abstract

Biomass burning is the main source of air pollution in the dry season in the Southeast Asian region including Chiang Mai, Thailand. The research aims to assess the spatial and temporal distribution of N deposition and to find out the source of N deposition link with biomass burning in Chiang Mai Province during 2008 - 2012. The main concentrations of N species concentration were NO_2 and NO_3^- . The deposition velocity (V_d) is important for calculation deposition flux. The average V_d values of N_g in descending order were $\text{HNO}_3 > \text{NH}_3 > \text{NO}_2$. While the V_d values of NO_3^- and NH_4^+ in forest area were higher than grass area in particulate phase. The average V_d values during 5 years (2008 - 2012), it can be set for other study to calculate deposition flux of N species. The N deposition trend in gas phase and particulate phase were measured for 5 years. The NH_3 and NH_4^+ were estimated the main deposition for gas and particulate phase, respectively. The good correlations between N_t deposition including N_g and N_p and PM_{10} and hotspot number were found in whole periods. High N deposition was originated from west direction of Chiang Mai Province. Therefore it was supported by correlation and Principal component analysis (PCA) between N species deposition and the number of hotspots. Moreover, the main source of nitrogen deposition was accepted as biomass burning for this area in term both of local source and long range transport in dry season periods.

Key words: Nitrogen deposition, Biomass burning, Backward trajectory, Wind rose deposition, hotspot

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1. Introduction

The global increase in nitrogen (N) emission has led to concerns that chronic elevated atmospheric concentration which has many caused negative effects (e.g., eutrophication, losing biodiversity and acidification) (Cornelissen *et al.*, 2001; Krupa, 2003; Prenni *et al.*, 2014; Karlsson *et al.*, 2013). However, there are few dry N deposition studies in Asia which were many sources of N deposition (agricultural activities, fertilizer production, combustion of fuels, and biomass burning). There are not many studies of linking pollution of reduced nitrogen with biomass burning which are increased for many countries.

Biomass burning was a big problem for Asia especially South East Asia. Thailand, Myanmar, Lao PDR and Indonesia have been annually facing air pollution (biomass burning) during the dry season (Kim Oanh *et al.*, 2011). There have been several large-scale fires for biomass burning in Northern Thailand which were many areas (Kim Oanh and Leelasakultum, 2011). This pollution has been recorded as a serious problem for well over 10 years. The burning area can be represented by hotspots which can be detected by remote sensing and GIS technologies to deliver global MODIS (Moderate Resolution Imaging Spectroradiometer). The number of hotspots in Chiang Mai Province is presented in Figure 1, based on the data from FIRMS (Fire Information for Resource Management System) during 2008-2012. Recently, the problem has become worse than ever in the dry season period (January - April). Chantara *et al.* (2009) determined particulate ions of airborne in Chiang Mai and Lamphun, Thailand from June 2005 to June 2006. Both anions (SO_4^{2-} , NO_3^- , and Cl^-) and cations (NH_4^+ , Na^+ , K^+ , Mg^{2+} and Ca^{2+}) were significantly higher in the dry period (December - March) and transition period I (October - November) than those in other seasons. The dominant anion and cation were SO_4^{2-} and NH_4^+ , respectively. Moreover NO_3^- and NH_4^+ were high in the dry season in Northern Thailand which has been a large scale of biomass burning.

Therefore the objectives of the research are: To assess the spatial and temporal distribution of N species in dry deposition (NO_2 , HNO_3 , NH_3 in gas phase and NO_3^- , NH_4^+ in particulate phase) and to correlate biomass burning and nitrogen species concentrations in Chiang Mai Province. In this study, sampling of dry deposition has been carried out using the filter pack method at a rural site in Chiang Mai Province.

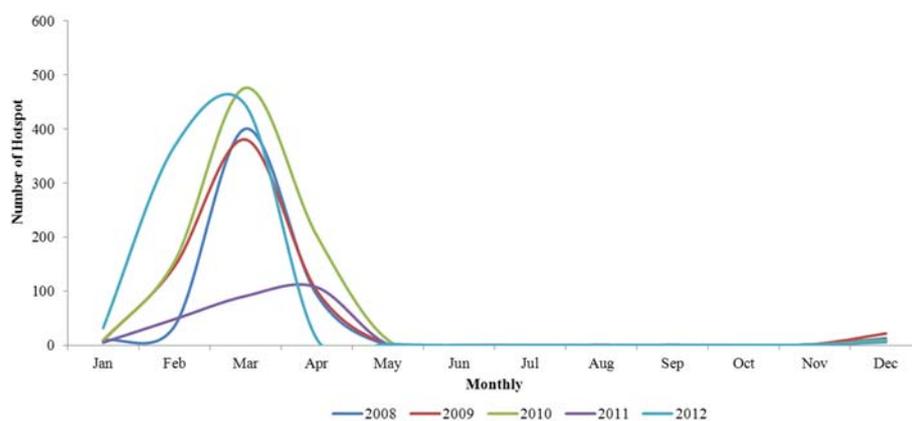


Figure 1. The Monthly hotspot numbers in Northern Thailand (Chiang Mai Province) during 2008-2012.

2. Methodology

2.1 Sampling site

The sampling site was at the meteorological station in the area of Mae Hia Research Center, Chiang Mai University, Muang District, Chiang Mai Province (Figure 2). This site was classified as rural site based on Acid Deposition Monitoring Network in East Asia (EANET) criteria since 2000. It is located at latitude 18° 45' 40.3" N and longitude 98° 55' 54.3" E. The sampling had been done within 5 year from 1st January, 2008 to 31st December, 2012.

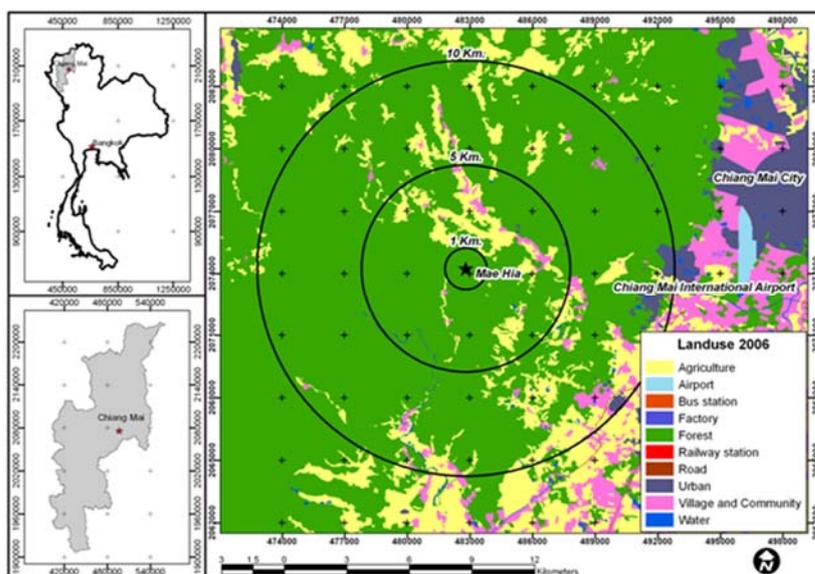


Figure 2. Location of the meteorological station in the area of Mae Hia Research Center, Chiang Mai University.

2.2 Sample collection

N species sample in gas phase such as HNO_3 , NH_3 and in aerosol (NO_3^- , NH_4^+) were collected by a four-stage filter pack. The first two layers are polytetrafluoro ethylene filter; F0 and polyamine filter; F1. The other two layer filters are cellulose acetate filter (F2, F3), which impregnated with potassium carbonate and phosphoric acid and glycerin solution. Prior to sampling, a filters pack was connected to a sampling set at the monitoring site. Flow rate of air input was 1 L/min. Sampling was operated for 10 days long, 3 times a month during 2008 - 2012.

2.3 Samples Analysis

The filters of a four-stage filter pack, F0, F2 and F3 filter papers were extracted with 20 mL Milli Q water, while F1 was extracted with 20 mL 0.05% H_2O_2 by ultrasonication for 30 minutes. After extraction process, insoluble matters were filtered by cellulose acetate membrane (EANET, 2002). Ion chromatograph (Metrohm, Switzerland) was used for determination of major ions (NH_4^+ and NO_3^-). Analytical columns for anions and cations were Metrosep A Supp 5 (5×250 mm) and Metrosep C2 150 (4×150 mm), respectively.

2.4 Hotspot data

The hotspot data are provided by FIRMS that integrates remote sensing and GIS (Geographic Information System) technologies to deliver global MODIS. FIRMS was developed by the University of

Maryland with funds from NASA (National Aeronautics and Space Administration). The hotspots are detected using data from the MODIS instrument, on board NASA's Aqua and Terra satellites. The hotspots represented fire location the center of a 1 km pixel (approximately) flagged as containing one or more actively burning hotspot/fires within that pixel (NASA/University of Maryland, 2002).

2.5 Meteorological data

Thai Meteorological Department (TMD) provided data on temperature, relative humidity, radiation, rain precipitation, wind speed and cloud cover, which was used for this study. Most of the data were hourly data set. The data were used for calculation of velocity deposition (V_d). There were calculated follow method of the technical manual on dry deposition flux estimation in East Asia which was run by EANET (EANET, 2010).

2.6 Trajectory analysis

Backward trajectories arriving at the receptor (sampling site) were calculated using the HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) model. The 3 days backward trajectories are available online at <http://ready.arl.noaa.gov/HYSPLIT.php>. Air mass trajectories for each individual day were calculated only the highest N deposition period (10 days) for each year during 2008 - 2012.

2.7 Data analysis

PCA and Pearson correlation (r) was implemented to identify the relationships between number hotspot, PM_{10} and N deposition species in Chiang Mai Province. SPSS program was used for determination the factors underlying the inter-correlations between the measured species.

3. Results and discussion

3.1 Seasonal variability of N species concentrations

The concentrations of atmospheric N species had been done from 2008 - 2012. Total number of samples was 36 samples per year. The N species in gas phase (N_g), it was consist of NO_2 , HNO_3 and NH_3 while N species in particulate phase (N_p) was NH_4^+ and NO_3^- . The total nitrogen species (N_t) was included all of N_g and N_p . All nitrogen species was analysed by IC except NO_2 was used chemiluminescence. The NO_2 data was provided by Pollution Control Department, Thailand (PCD). The 5 years average N_g concentration of NO_2 , HNO_3 and NH_3 were $21.08 \pm 11.52 \mu g/m^3$, $3.12 \pm 1.48 \mu g/m^3$ and $3.25 \pm 3.87 \mu g/m^3$, respectively. While N_p concentration NH_4^+ and NO_3^- were $1.72 \pm 0.67 \mu g/m^3$ and $4.17 \pm 1.05 \mu g/m^3$, respectively. The main concentration of N_g was NO_2 and N_p was NO_3^- .

Figure 3 illustrated mean concentrations of hotspot number PM_{10} and N species (N_g , N_p and N_t) in periods during 2008 - 2012. Seasonal variations of hotspot number, PM_{10} and N species concentrations were almost the same. Their concentrations were highest in dry season decreasing in wet season. The important source was open burning including forest fire, agricultural waste and garbage burning in communities in dry season due to PM_{10} was very high concentration in the same period. They were assumed to be the main sources N species in this period. In addition, high frequency of forest fire was observed via number of hotspots detected by FIRM. Therefore May 2010, the concentration of N_p was high in rainy period which was abnormal situation. Moreover, in year 2011 was high amount of rain in the sampling area, which was diluted to low N_t concentration in dry season period (low number of hot spots and PM_{10} concentration).

3.2 N species deposition (2008 - 2012)

The amount of acid deposition per unit area per time or we know in term of deposition flux which is importance data to assess the impact by acid deposition. The products of air concentration and deposition velocity are necessary to be known. This study was followed the process from EANET method (EANET, 2010). The deposition velocities depend on many factors such as wind speed, humidity, temperature, net radiation etc.

Deposition velocity (V_d) of gases and particulates phase N species in forest and grass area in Chiang Mai Province are shown in Table 1. The deposition velocities are presented in the unit of cm^2/s . V_d of hourly data were calculated and then the V_d were averaged in the time resolution of N species concentration. The average values of V_d were estimated for gas phase (NO_2 , HNO_3 and NH_3) and particulate phase (NO_3^- , NH_4^+) in forest and grass area during 2008 - 2012. The V_d values were high for HNO_3 in forest area ($2.26 - 3.09 \text{ cm}^2/\text{s}$) while the V_d values of NO_2 ($0.08 - 0.10 \text{ cm}^2/\text{s}$) and NH_3 ($0.32 - 0.20 \text{ cm}^2/\text{s}$) were small variance in both of forest and grass area. NO_3^- and NH_4^+ were $0.40 - 0.50 \text{ cm}^2/\text{s}$ and $0.31 - 0.40 \text{ cm}^2/\text{s}$, respectively in forest. While the V_d values of NO_3^- and NH_4^+ were used the same values ($0.12 - 0.15 \text{ cm}^2/\text{s}$).

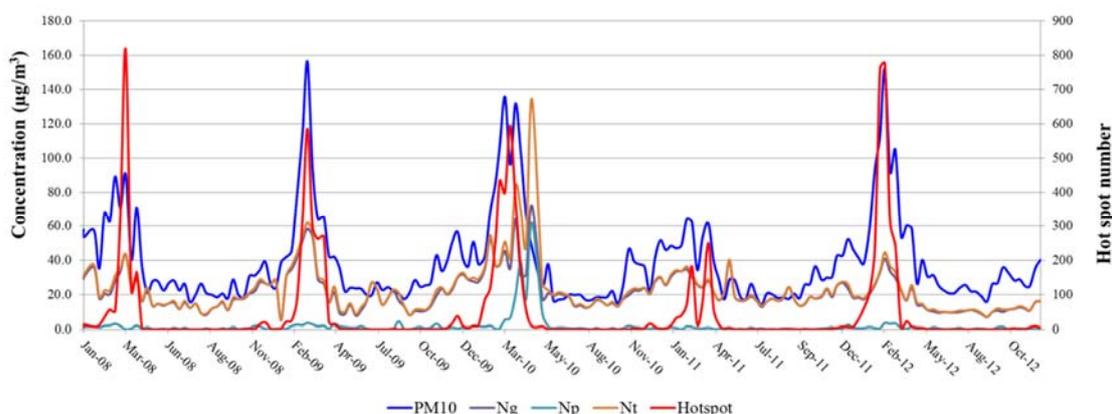


Figure 3. Ten days averaged concentration of N species, PM_{10} and hotspot number during 2008 - 2012.

Table 1. Deposition velocity of gases and particulates phase N species in forest and grass area in Chiang Mai (Unit: cm^2/s) .

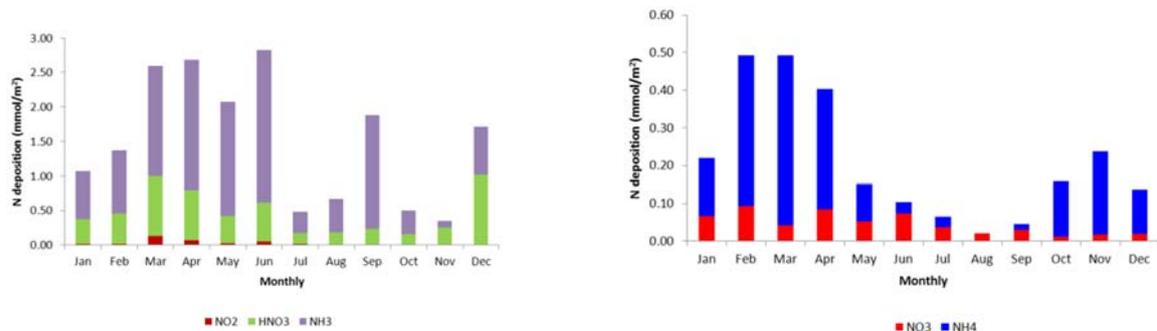
years	Gas Phase						Particulate Phase		
	forest			grass			forest		grass
	NO_2	HNO_3	NH_3	NO_2	HNO_3	NH_3	NO_3^-	NH_4^+	$\text{NO}_3^-, \text{NH}_4^+$
2008	0.09	2.26	0.32	0.08	0.76	0.22	0.40	0.31	0.12
2009	0.10	2.96	0.24	0.10	0.99	0.22	0.48	0.38	0.15
2010	0.09	3.09	0.22	0.10	1.04	0.21	0.50	0.40	0.15
2011	0.09	2.67	0.23	0.10	0.89	0.21	0.45	0.35	0.12
2012	0.09	2.66	0.21	0.10	0.89	0.20	0.44	0.34	0.13
Average	0.09	2.73	0.24	0.10	0.91	0.21	0.45	0.36	0.13

The dry deposition flux of N species were calculated from concentration of N species and V_d :

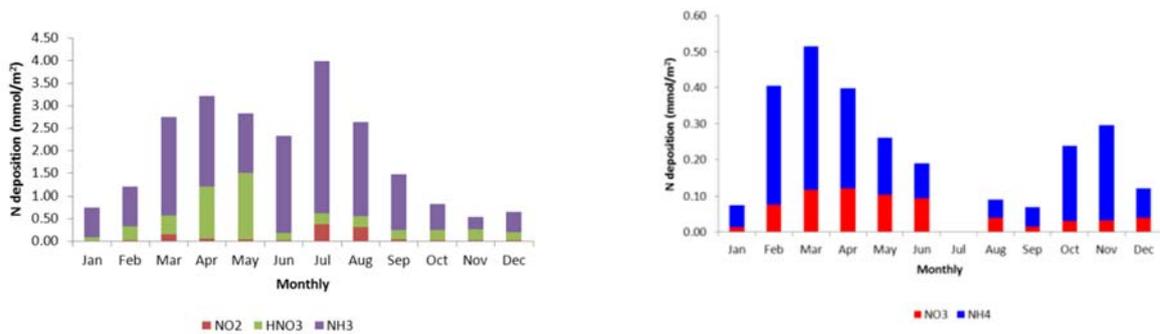
$$F_i = V_d^i \times C_i \quad (1)$$

where F_i is flux of i species and C_i is concentration of i species. The N species deposition fluxes were calculated in gas and particulates phase in both of area (forest and grass). The percentage of forest area (85 %) and grass (15 %) were estimated by Arc-GIS land use program.

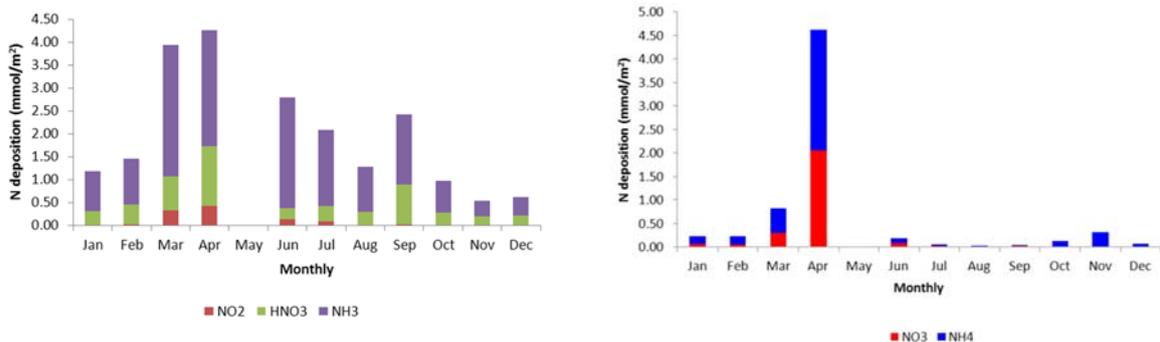
The profile of N species deposition in gas phase and particulate phase were illustrated in Figure 4 during 2008 - 2012. The main N species depositions were NH_3 and NH_4^+ for gas and particulate phase. Furthermore deposition in gas phase higher than in particulate phase. The particulate N depositions were high in the dry season and always highest in March - April. The most important factors are the amount of rain precipitation and scale and frequency of open burning. The gas N depositions were high in March - June while quite low for about 5 months (October - February). Then the concentrations increased again at in dry season (March) due to lower precipitation and higher burning activities in agricultural areas including forest area. Therefore in 2011 was low deposition for particulate phase due to the high amount of rain precipitation.



A (2008)



B (2009)



C(2010)

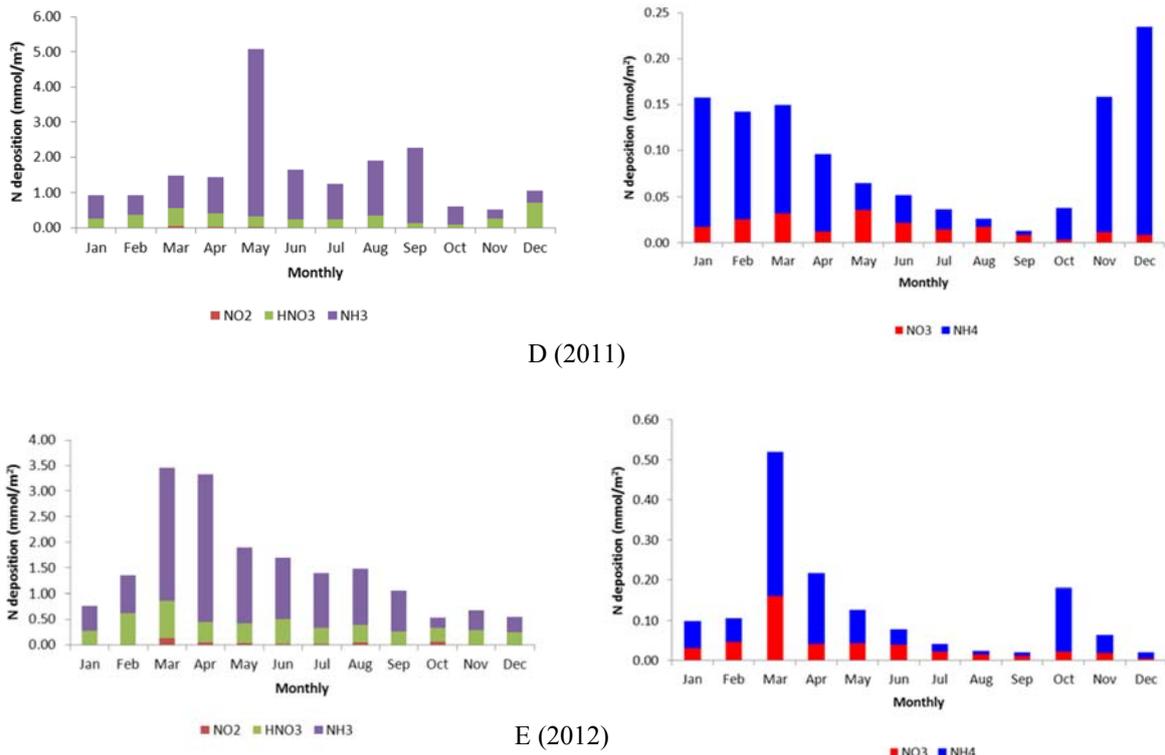


Figure 4. Monthly averaged N depositions in Chiang Mai during 2008 - 2012.

Figure 5 was illustrated the monthly averaged of N species deposition (N_g , N_p and N_t), PM_{10} and hotspot number during 2008 - 2012. The N_t were quite high in the dry season and getting lower in the wet season in every years. In May 2010, the concentration of N species was very high (2-3 times higher than normal) which was contaminated from the method. Therefore the data of May 2010 was deleted for data analysis. In 2011, it was started raining early than others year that was made lower PM_{10} concentrations and hotspot number.

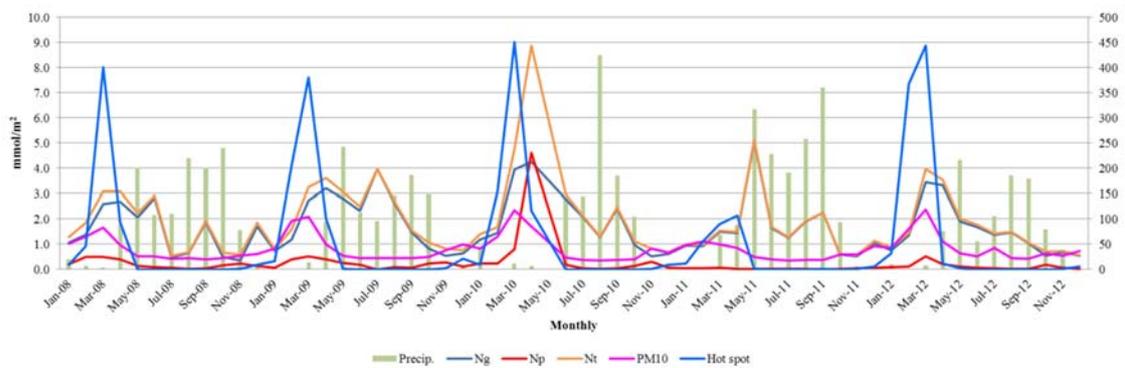


Figure 5. Monthly averaged trend of N species deposition, PM_{10} and hotspot number during 2008 - 2012.

The comparison annual average of N deposition in this study with others study was shown in Figure 6. The N_p deposition in Chiang Mai (CM) was lowest (5.68 $mmol/(m^2 \text{ y})$) while was highest deposition in EMEP (20.71 $mmol/(m^2 \text{ y})$). For N_g deposition, CM was highest deposition compare with other studies. The

Clean Air Status and Trends Network (CASTNET) in USA and The European Monitoring and Evaluation Programme (EMEP) were not reported N_g deposition. The N_t deposition in descending order were CM (26.10 $\text{mmol}/(\text{m}^2 \text{ y})$) > EANET (24.24 $\text{mmol}/(\text{m}^2 \text{ y})$) > EMEP (20.71 $\text{mmol}/(\text{m}^2 \text{ y})$) > CASTNET (9.09 $\text{mmol}/(\text{m}^2 \text{ y})$) .

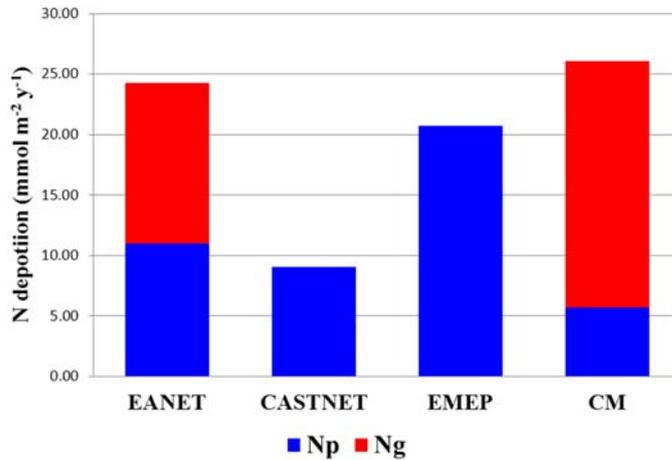


Figure 6. Comparison of N deposition per year with other studies.

3.3 Correlation between N species deposition and biomass burning factor

Correlations between N species deposition, PM_{10} and hotspot number were analysed by Pearson correlation during 2008 - 2012 in whole year (Table 2). The N_g and N_t (N_t+N_g) depositions were very strong correlated ($r = 0.990$). Furthermore, N_g deposition was correlated ($r = 0.348$) with hotspot number. While N_p deposition was moderately correlated with PM_{10} and hotspot number ($r = 0.739$ and $r = 0.6565$, respectively) with $p < 0.01$. Moreover, N_t deposition was weakly correlated ($r = 0.372$ and $r = 0.432$) with PM_{10} and hotspot number with $p < 0.05$.

Table 2. The correlation between N species deposition, PM_{10} and hotspot number during 2008 - 2012.

	N_g	N_p	N_t	PM_{10}	Hotspot	Precipitation
N_g	1.000					
N_p	0.349*	1.000				
N_t	0.990**	0.475**	1.000			
PM_{10}	0.280	0.739**	0.372*	1.000		
Hotspot	0.348*	0.656**	0.423*	0.891**	1.000	
Precipitation	0.077	-0.358	0.019	-0.621**	-0.444**	1.000

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

For assessment the impact of biomass burning was analysed data in dry season periods (January - April) which was demonstrated in Table 3. The N_t deposition was moderately with PM_{10} ($r = 0.680$) and hotspot number ($r = 0.618$) with $p < 0.01$. The strong correlations were found between deposition of N_g and N_t ($r = 0.973$) and N_p and N_t ($r = 0.819$). Moreover, N_g deposition was fairly correlated with PM_{10} ($r = 0.603$) and hotspot number ($r = 0.9597$) while N_p was good correlated with PM_{10} ($r = 0.704$) and hotspot number ($r = 0.569$). The results revealed that biomass burning emitted N species.

Table 3. The correlation between N species deposition, PM₁₀ and hotspot number during 2008 - 2012 in dry season periods.

	N _g	N _p	N _t	PM ₁₀	Hotspot
N _g	1.000				
N _p	0.711**	1.000			
N _t	0.973**	0.819**	1.000		
PM ₁₀	0.603**	0.704**	0.680**	1.000	
Hotspot	0.597**	0.567**	0.618**	0.802**	1.000

* Correlation is significant at the 0.05 level (2-tailed).

** Correlation is significant at the 0.01 level (2-tailed).

PCA has been applied to confirm the source from biomass burning. The factor analysis for N deposition and other factors were performed with two factors with an Eigen value >1, contributing ~85% of the variance. First factor contributes 50.85% of the total variance and high loading with N_p, PM₁₀ and hotspot number (Figure 7). Therefore, loading scatter plots for secondary factors were grouped N_t and N_g which were the same coordinate. These results can be used for identification of the biomass burning source.

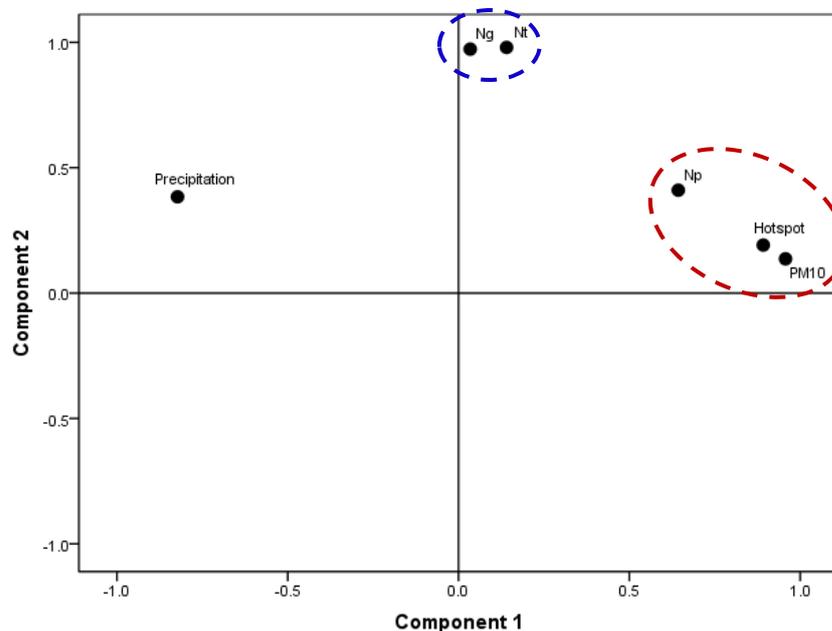


Figure 7. Loading scatter plot of N species deposition, PM₁₀ and hotspot number during 2008 - 2012.

3.4 Wind rose deposition and back ward trajectory

In order to gain more information on N deposition, both in terms of type and position (local or long range transportation), wind rose deposition and three days backward trajectories of the air masses arriving at Chiang Mai were analysed by meteorological data and HYSPLIT model.

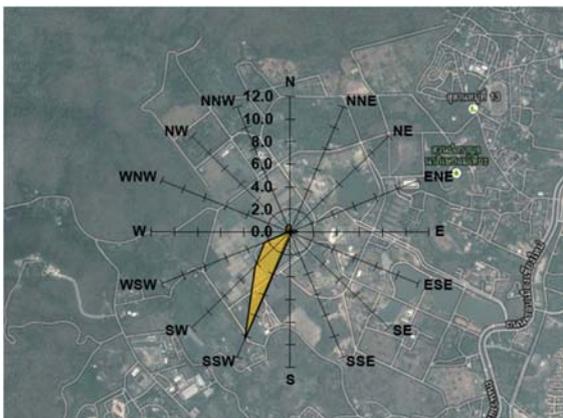
The wind rose deposition was computed by wind direction and N_i deposition (Figure 8) during 2008 - 2012. All of them were recorded from the west direction, generated in western part of Chiang Mai (Mae Cham District). In dry season, high density of hotspots in the western part of Chiang Mai Province was found especially Mae Cham District and Mae Hong Son Province.



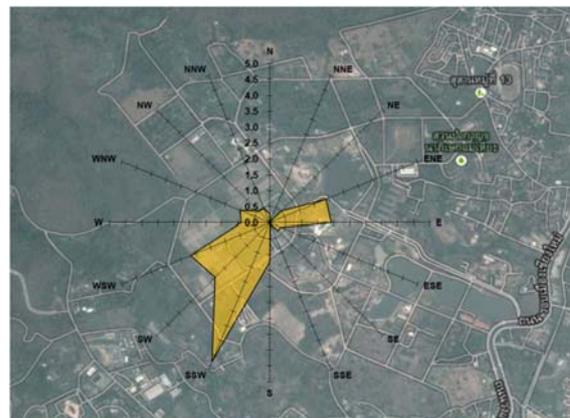
A(2008)



B(2009)



C(2010)



D(2011)



E(2012)

Figure 8. Wind rose N deposition during 2008 - 2012.

Therefore a movement of air mass from air mass west direction could bring pollutants generated from open burning to the receptor (sampling point).

Three days backward trajectories were run on the high Nt deposition one day per each year. The directions of trajectories were shown in Figure 9. Almost of direction came from the west direction, generated in the western region (Myanmar) and then travelling over western Thailand (Mae Hong Son Province), before arriving in Chiang Mai. The hotspots numbers were high in dry season periods which were occurred in the western part of Chiang Mai Province. Therefore a movement of air mass from air mass west direction could bring pollutants generated from open burning to the receptor (Chiang Mai Province). Air mass trajectories pattern were similar to wind rose deposition which were identified the biomass burning source.

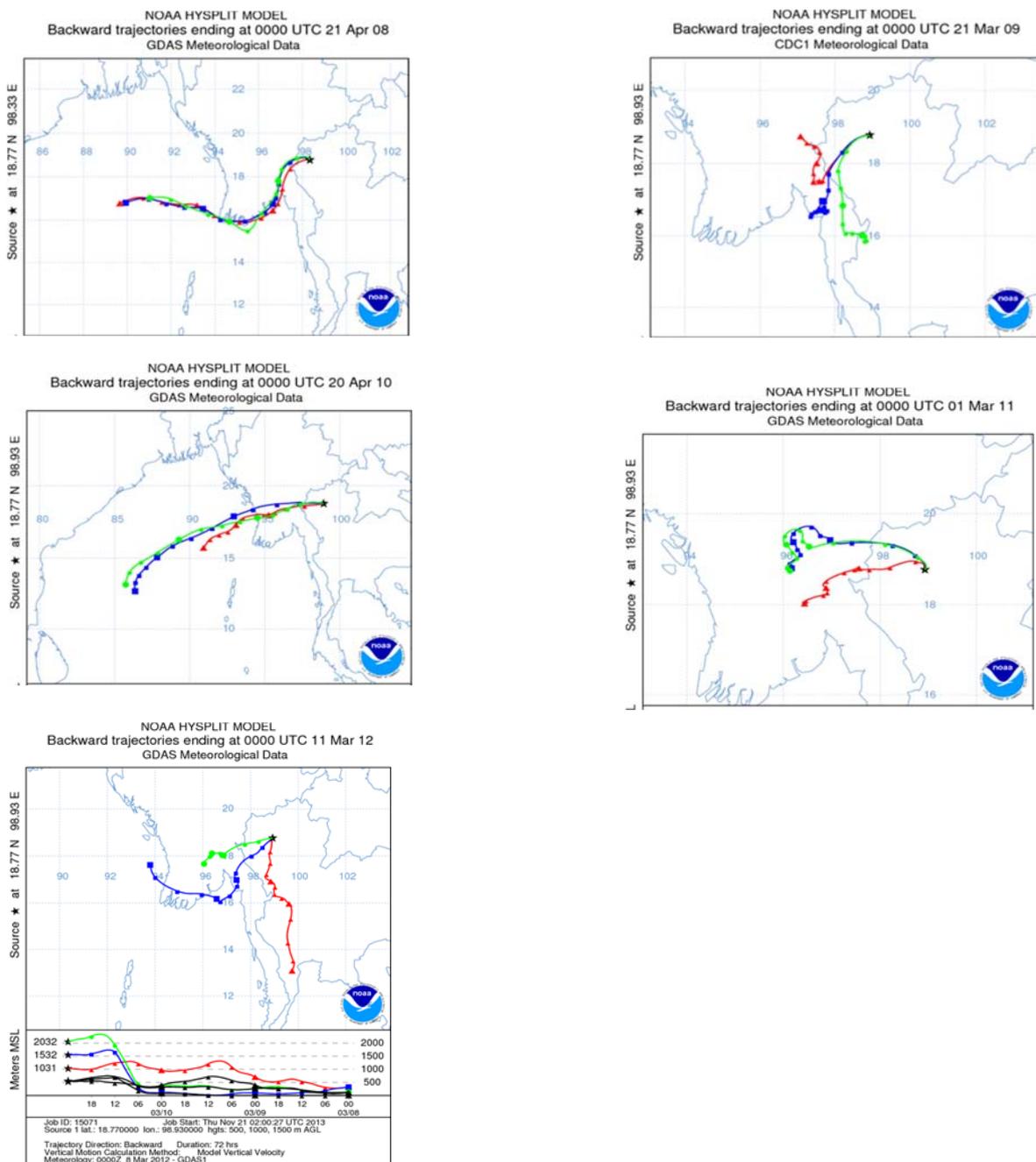


Figure 9. 3-day backward trajectories of the high N deposition periods during 2008 - 2012.

4. Conclusion

Chiang Mai and the northern part of Thailand have been faced with air pollution every dry season. The main air pollution source in this area is open burning especially forest fires. However, there were only few studies on N deposition from biomass burning in Thailand.

The concentrations of atmospheric N species were monitored in 2008 - 2012 in Chiang Mai Province (Chiang Mai University). The 5 years average N_g concentration in disorder were $NO_2 \gg NH_3 > HNO_3$ while N_p were $NO_3^- > NH_4^+$. The dominant of N species concentration for N_g and N_p were NO_2 and NO_3^- , respectively. The importance data were deposition velocity are important for calculation deposition flux. The average V_d values were HNO_3 (2.73 cm^2/s) higher than NH_3 (0.24 cm^2/s) and NO_2 (0.10 cm^2/s) while NO_3^- and NH_4^+ were 0.40 - 0.50 cm^2/s and 0.31 - 0.40 cm^2/s , respectively in forest. While the V_d values of NO_3^- and NH_4^+ in forest were higher than (0.45 and 0.36 cm^2/s , respectively) in grass area (0.13 cm^2/s). The seasonal variation of N deposition in gas phase and particulate phase were conducted during 2008 - 2012. The NH_3 and NH_4^+ were estimated the main deposition for gas and particulate phase, respectively.

The good correlations between N_t deposition including N_g and N_p and PM_{10} and hotspot number were found in whole periods (2008 - 2012). Meteorological parameters (amount and frequency of precipitation) and number of hotspots were found to be the most influential factors affecting N deposition.

High N deposition during 2008 - 2012 originated from west direction of Chiang Mai Province in every year, which was confirmed by wind rose deposition and backward trajectory. In the dry season was large scale of biomass burning in this region. This was supported by correlation and PCA between N species deposition and the number of hotspots. In addition biomass burning was accepted as an important source of nitrogen for this area in term both of local source and long range transport.

Acknowledgements

Financial supports from the Network Center for the Acid Deposition Monitoring Network in East Asia (EANET), Asia Center for Air Pollution Research (ACAP) are gratefully acknowledged. We would like to thank Mr. Shirai, Ms. Aoyagi and Ms. Nakamura (Data Management Department, ACAP) for the informative discussion on the data of research project. Moreover, we also thank for Pollution Control Department (PCD), Thailand, Fire Information for Resource Management System (FIRMS) and Thai Meteorological Department (TMD) for providing observation data and the NOAA Air Resources Laboratory (ARL) for the HYSPLIT dispersion model.

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