Research article

Index of Atmospheric Purity (IAP) Related to Potential Ecological Risk Indexes (RI) of Heavy Metals Accumulation in Urban Area

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Abstract

Keywords

environmental assessment;
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Heavy metal contamination in protected areas can cause sensitive ecosystems to be at risk. Bioindicators for monitoring heavy metal contamination need to be investigated. The objectives of this study were to determine heavy metal residues in soil in an urban area, and also lichens diversity. Twenty-two sampling plots of 1 km x 1 km size were selected in Nong Saeng sub-district, Pak Phli district in Nakhonayok province, Thailand. Lichens diversity was investigated, and soil samples were collected to analyze the amount of heavy metal residues in the soil. Afterwards, the potential ecological risk indexes (RI) and index of atmospheric purity (IAP) were presented. The results indicated that trace element concentrations in forest (For), urban (Urb) and agriculture (Agr) soils were not significantly different, and that land use type did not affect heavy metal contamination. However, two areas with high RI values of 358.27 and 483.76 were designated as being at considerable ecological risk. These values related to the lowest air quality index in distribution mapping of IAP. The relationship between index of atmospheric purity and potential ecological risk indexes (RI) of heavy metal accumulation in urban area implied that long-range transboundary air pollution may be a source of heavy metals contamination in some areas. The highest RI value related to Hg concentration in low land implied that it was possible that concentrations of heavy metals could have been affected by the discharge of wastewater into the low land, and especially in the study area that had acidic soil.

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1. Introduction

Khao Yai National Park is a protected area and an important large ecosystem in Thailand. However, increases in tourism activity, intensive travelling and agricultural activity have caused increased release of pollutants in urban areas near the national park. The chemicals used in farming can leave long term chemical residues in the environment. Some of them can be accumulated in living organisms and can affect their health and ecology. In addition, some contaminants can be transferred to nearby areas and into the protected areas in the highlands by atmospheric factors such as wind [1]. Long-range transportation of air pollution has been recognized as one factor affecting the ecosystem, and especially atmospheric metal contamination in a natural park in Thailand was observed [2]. Heavy metals are common air pollutants and are emitted as a result of various activities, and soils are the main sink for heavy metals released into the environment [3]. In addition, land utilization of agrochemicals and wastewater irrigation have led to heavy metal contamination in agricultural and urban areas [3, 4]. However, long-range transboundary air pollution may be the source of some heavy metal contamination in the area. The heavy metals in air pollution can be accumulated in trees, tree bark, mosses, and lichens along the pollution sites [5]. Lichens are widely used in biomonitoring. The air pollution affecting epiphytic lichen flora is determined and expressed as an air purity index known as the index of atmospheric purity (IAP), which relates to lichen diversity and includes an ecological index for each lichen species [6, 7]. Many researchers apply the IAP value for determination of urban air quality [8-10]. The aim of this study was to determine the potential ecological risk index (RI) by measuring heavy metal accumulation and the index of atmospheric purity (IAP) by the study of lichens diversity. The relation between RI and IAP was presented by mapping the spatial distribution of their values. The relationship between index of atmospheric purity (IAP) and potential ecological risk indexes (RI) of heavy metal accumulation in urban area implies that long-range transboundary air pollution may be another source of heavy metal contamination in some areas. In addition, lichen bioindicators are useful for monitoring heavy metal pollution in urban areas before the pollutants are transported to the national park and affect the spatial ecological system in the conserved area.

2. Materials and Methods

2.1 Study area

The study was conducted in the urban areas of Nong Saeng sub-district, Pak Phli district in Nakhonayok province, Thailand. The Nong Saeng sub-district is located at latitude 14°12'09.4"N and longitude 101°18'14.3"E. It is recognized as a main ecotourism area with a lot of organic farming activity. It has a population of 3307. The elevation of most town areas is in the range of 7-30 m above sea level. The area occupies about 20 square kilometers, and agriculture activities such as rice paddy fields and other forms of farming dominate. The north of sub-district is close to Khao Yai National Park and other sites are close to attractive places for tourists. This town is a travelling hub, and dozens of roads pass through Nong Saeng sub-district. Twenty-two sampling sites, which were 1x1 km sample plots, were located within or nearby roads, and farming and residential areas. The details are shown in Figure 1.

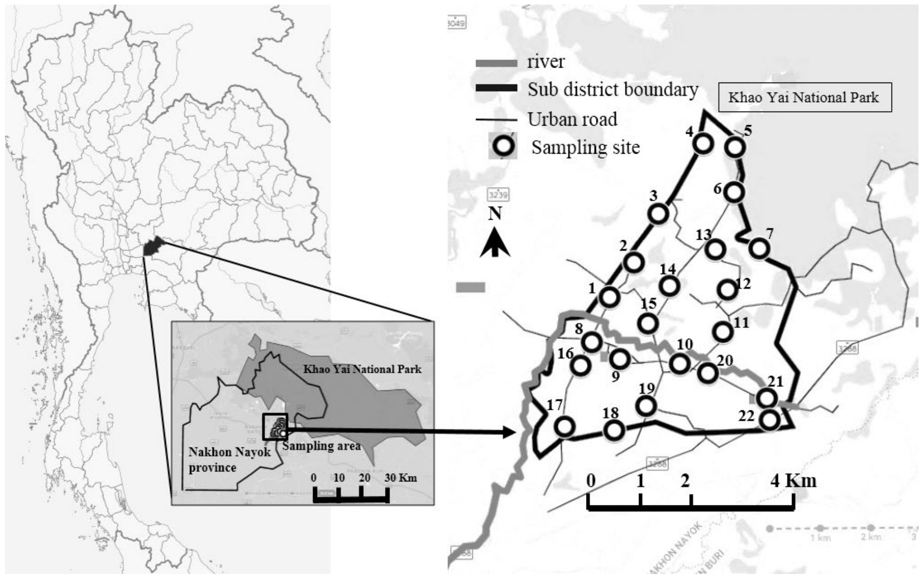

Figure 1. Study sites in Nong Saeng subdistrict, Pak Phli district, Nakhon Nayok province. The numbered white circles are sampling points.

2.2 Determination of lichen diversity and index of atmospheric purity (IAP)

The lichen diversity was determined during May-September 2018. In 1x1 km sampling site, three trees of about 20-30 cm in diameter and with the same bark surface character were selected. Three quadrats of 10x10 cm in 4 directions, north, east, west, and south faces of each tree at a height of 1.5 from the ground surface were used for lichen diversity measurement. The lichen thalli were separated from the tree bark surfaces, and then the thalli were identified under a microscope. All sampling point geographical coordinates were recorded using a GPS device (Garmin 62c). Then IAP was calculated using the following formula [11]:

$$IAP = 1/10 \sum_{i}^{n} Q \times f$$

The frequency-coverage (f) of each species is estimated by multiplying the frequency of occurrence and coverage. Frequency of occurrence is calculated as the percentage of quadrat on which a species physically presents at a given site while the coverage is the surface area of the species at that site. The ecological index (Q) of each species is defined as the average number of companion species that coexisted with each species. While n is the total number of species at that site.

2.3 Heavy metal contents analysis in soils

For soil sample preparation, 50 g of 0-15 cm layer soil from four corners around the tree were collected and then were mixed to make one composite sample. All samples were kept in clean polyethylene bags and labeled to ensure that samples were of the same size, and then brought to the laboratory for analysis. The soil samples were dried at 60°C, and then sieved through a 0.25 mm stainless steel mesh. For heavy metal analysis, the digestion method used followed that of Thummajitsakul *et al*. [12, 13]. Two grams of dried sieved soil were digested in 100 mL beaker with 15 mL of conc (68) HNO_3 at 80°C until the solution became maximally transparent. The digestion was done in three replicates. After cooling, distilled water was added and the volume was brought up to 50 mL. The samples were filtrated and then adjusted to 50 mL. The filtrated samples were analyzed by using flame oxidizing atomic absorption spectroscopy (Agilent-Model 280AA, CA 95051, USA), and the heavy metal concentration were determined by absolute calibration with solutions of known concentration of heavy metals that had been made from 1000 ppm stock solution [14].

2.4 Potential of ecological risk

Each heavy metals concentration in the soil was used for evaluation of potential of ecological risk. It was calculated as:

$$\boldsymbol{Pi = Ci\ /\ Si}$$

where P_i is the pollution index; C_i is the measured concentration of heavy metal in the soil (mg/kg); and S_i is the evaluation standard for that heavy metal (the standard values of trace elements in the Thailand soils are: As, 30; Cd, 0.15; Co, 20; Cr, 80; Cu, 45; Hg, 0.10; Ni, 45; Pb, 55 and Zn, 70 mg/kg) [15]. The potential ecological risk (*Er*) assesses the toxicity of some trace elements in sediments [16, 17]. To calculate the *Er* for each metal, the following equation was used:

$$\boldsymbol{Er = Tr \times Pi}$$

where the toxicity coefficient of each metal (*Tr*) standard values are: Hg = 40, Cd = 30, As = 10, Co = 5, Cu = 5, Ni = 5, Pb = 5, Cr = 2 and Zn = 1 [18]. The potential of ecological risk index (*RI*) was calculated using the following equation:

$$\boldsymbol{RI = \sum_{i-1}^{n} Er}$$

2.5 Analysis of spatial distribution of heavy metals

The RI and IAP were treated statistically using the SPSS package. The Bivariate-Correlation was used for determination of heavy metals relationships. The IAP and RI for every site were plotted with the Kriging interpolation using QGIS v.2.8 (Golden Software Inc., Colorado, USA) to show spatial distribution mapping of IAP and RI.

3. Results and Discussion

3.1 Soil heavy metal content analysis and ecological risk

The heavy metal content in soil was determined to investigate pollutant correlation and to estimate sources of heavy metal contaminations in this area. Six heavy metals were detected and quantified, and the results were: Cr>Zn>Cu>As>Ni>Hg with means of 55.47±6.86, 27.15±3.67, 1.65±0.75, 0.24±0.014, 0.63±0.039, and 0.16 ±0.14 mg/kg, respectively, while Cd, Mn and Pb were below the detection limit (Table 1). *The maximal permissible* addition (MPA) of heavy metals and metalloids (mg/kg) are As = 1-40 mg/kg; Cr = 1-1000 mg/kg; *Cu* = 2-100 mg/kg; Ni = 5-500 mg/kg; Pb = 2-200 mg/kg; Zn = 10-*300* mg/kg [14]. Heavy metals in the sampled soil did not exceed the *maximal permissible* limits. Previous research in the neighborhood reported that soil samples contained 0.14-0.5 mg/kg of Zn, and 0.5-1.01 mg/kg of Cu [14]. The heavy metal content in the soil was close to that in other reports from Phra Nakhon Si Ayutthaya, Thailand [19]. Since most of the study area in our work was agricultural land, it is possible that the contamination came from N fertilizers and some kinds of pesticides and germicides [19]. The concentrations of Cd, Cu, and Zn may have been associated with fertilizer inputs. It was found that the land use patterns had significantly different effects on the accumulation of the heavy metals Cr, Cu, Cd, and Zn [20]. However, the heavy metals Cd, Cu, Pb, and Zn may have originated from vehicles [21], and Cr, in particular, may be associated with the wearing of vehicle brakes [22].

The potential ecological risk index (RI) relates to the potential ecological harm from multiple elements [9]. The potential ecological risk factor was in the range of 0-14.75, 0-8.42, 0-1.06, 0.36-2.61, 0.54- 4.62, and 0-470.26 for Cr, Cu, Ni, Zn, As, and Hg, respectively (Table 2). The categories established for the RI index for classifying risk are: low ($RI < 150$), moderate ($150 \leq RI < 300$), considerable ($300 \leq RI < 600$) and very high ecological risk ($RI > 600$) [23]. Sixteen study areas were categorized as low risk as they fell in the range of 3.79-136.63. Four other areas including sampling sites S2, S10, S16 and S21 were moderate in risk with the range between 212.22 and 253.72, and two areas including sampling sites S18 and S19 were categorized as being considerable risk with the range of 358.27 and 483.76, respectively (Table 2). It was observed that only a small proportion (3.08%) of the soil samples were considered to be in the risk category for potential contamination. The Er of Hg relates to the high value of RI in an urban and agricultural area. The high value of RI relates to Er of Hg, where the toxicity coefficient (Tr) of Hg is 40. This indicates the hazardousness of Hg [23].

3.2 Diversity of lichen in the urban areas

The lichen diversity is shown in Figure 2. There were two fruticoses, forty folioses, and nine crustoses composed of 443 lichens belonging to 34 genera and 14 families. The commonest families were Parmeliaceae and Graphidaceae, and *Lepraria* sp. had the highest population. The lichen distribution with land use type is shown in Figure 3. Twenty-three species were presented in urban areas, 11 species were in agricultural areas and 13 species were in forest areas (Figure 3A). From the lichen diversity analysis, total lichen was in the range of 8-50, and the Shannon-Weiner index (H'-index) was in the range of 0.0-0.78. Frequency was in the range of 0.75-2.75, and richness was in the range of 1-9 (Figure 3B). However, the comparison of lichen diversity index with Prost Hoc test (Duncan) in one-way ANOVA showed that there were no significant differences between urban, agriculture and forest areas (Figure 3B). Some lichen can live in polluted areas because they tolerate high concentrations of metals due to cellular mechanism [24]. Many reports indicated that the lichen *Dirinaria picta* showed correlated-metabolic profiles when exposed to the heavy metals Fe, Zn, Cu, Cd, Pb and Co [25]. Three lichens, *Usnea amblyoclada, Ramalina celastri* and *Tillandsia capillaris*, were tested for their bioaccumulation of transition metals in an urban area [26, 27].

Table 1. Trace element concentrations in sampling surface soil

Sites	Cr	Cu	Hg	Ni	Zn	As	Soil pH	Altitude (m)	Latitude	Longitude	Land Used
maximum permeable (mg/kg)	300	100	23	1600	300						
S1	30.39	10.08	BDL	BDL	24.23	0.10	6.00	9	14.21156	101.29414	Agr
S2	BDL	12.80	0.12	BDL	48.94	0.56	5.90	14	14.21788	101.2986	Agr
S3	BDL	1.91	BDL	BDL	48.56	0.48	4.60	16	14.22639	101.30304	Agr
S4	43.56	BDL	0.02	BDL	20.72	0.44	4.40	21	14.23885	101.31134	Agr
S5	28.66	BDL	BDL	BDL	19.94	0.53	4.50	37	14.23831	101.31714	For
S6	68.77	BDL	BDL	BDL	24.35	0.49	4.20	30	14.23033	101.31699	For
S7	1.96	BDL	BDL	BDL	12.64	0.60	4.10	37	14.22034	101.32154	For
S8	38.17	BDL	BDL	BDL	19.94	0.72	3.80	7	14.20364	101.29093	Urb
S9	40.32	6.18	0.03	BDL	15.96	0.70	5.00	4	14.19741	101.29269	Urb
S10	59.58	0.64	0.12	BDL	79.94	0.69	5.30	14	14.19979	101.307	Agr
S11	38.09	BDL	0.02	BDL	23.91	0.84	5.40	7	14.20549	101.31482	Agr
S12	41.70	BDL	BDL	BDL	12.24	0.88	5.10	17	14.2129	101.31568	Agr
S13	72.85	BDL	0.06	BDL	11.84	0.74	4.60	18	14.22011	101.31345	Urb
S14	76.91	BDL	0.02	BDL	18.64	0.51	4.10	13	14.21368	101.30519	Agr
S15	72.59	2.67	BDL	BDL	38.87	0.72	5.20	6	14.20705	101.30123	Agr
S16	80.92	1.97	0.11	3.13	54.12	0.50	5.30	11	14.1995	101.28884	Urb
S17	62.19	BDL	0.05	BDL	17.27	0.78	5.10	13	14.18864	101.28595	Agr
S18	75.06	BDL	0.17	BDL	35.79	0.69	4.90	12	14.18802	101.29500	Agr
S19	87.19	BDL	0.24	0.23	20.90	0.45	4.00	10	14.19226	101.30081	Urb
S20	123.94	BDL	0.06	BDL	19.50	0.85	4.10	14	14.19815	101.31208	Agr
S21	92.84	BDL	0.10	BDL	11.15	0.79	4.80	11	14.19359	101.32301	Agr
S22	84.63	BDL	0.01	0.06	17.81	0.71	4.00	12	14.18991	101.32345	Urb
Min	BDL	BDL	BDL	BDL	11.15	0.10	3.80	4			
Max	123.94	12.80	0.24	3.13	79.94	0.88	6.00	37			
Mean	55.47	1.65	.051	0.16	27.15	0.63	4.75	15.14			

*Values of heavy metal content in the present study are given as means, samples were analyzed in triplicates. BDL: below detectable limit including Zn (0.01 ppm), Ni (0.10 ppm), Pb (0.10 ppm), Cu (0.03 ppm) and Mn (0.02 ppm) following optimum working range for Agilent manufacturing recommendation. Land use types were forest (For), urban (Urb) and agriculture (Agr).

Table 2. Individual (*Er*) and potential (*RI*) ecological risk index of the surface soil

Sites	Potential Ecological Risk Factor (Er)						RI	Pollution Degree
	Er-Cr	Er-Cu	Er-Hg	Er-Ni	Er-Zn	Er-As		
S1	3.62	6.63	0.00	0.00	0.79	0.54	11.58	Low
S2	0.00	8.42	240.77	0.00	1.60	2.93	253.72	Moderate
S3	0.00	1.26	0.00	0.00	1.59	2.50	5.34	Low
S4	5.19	0.00	49.78	0.00	0.68	2.30	57.94	Low
S5	3.41	0.00	0.00	0.00	0.65	2.80	6.86	Low
S6	8.19	0.00	0.00	0.00	0.80	2.58	11.56	Low
S7	0.23	0.00	0.00	0.00	0.41	3.15	3.79	Low
S8	4.55	0.00	0.00	0.00	0.65	3.81	9.00	Low
S9	4.80	4.07	68.44	0.00	0.52	3.68	81.50	Low
S10	7.09	0.42	235.92	0.00	2.61	3.61	249.65	Moderate
S11	4.54	0.00	30.54	0.00	0.78	4.44	40.29	Low
S12	4.97	0.00	0.00	0.00	0.40	4.62	9.98	Low
S13	8.67	0.00	123.66	0.00	0.39	3.91	136.63	Low
S14	9.16	0.00	39.54	0.00	0.61	2.66	51.97	Low
S15	8.64	1.76	0.00	0.00	1.27	3.78	15.45	Low
S16	9.63	1.30	223.58	1.06	1.77	2.62	239.96	Moderate
S17	7.40	0.00	106.62	0.00	0.56	4.08	118.67	Low
S18	8.94	0.00	344.52	0.00	1.17	3.64	358.27	Risk
S19	10.38	0.00	470.26	0.08	0.68	2.36	483.76	Risk
S20	14.75	0.00	111.80	0.00	0.64	4.48	131.67	Low
S21	11.05	0.00	196.64	0.00	0.36	4.16	212.22	Moderate
S22	10.08	0.00	20.26	0.02	0.58	3.76	34.70	Low
Min	0	0	0	0	0.36	0.54	3.79	
Max	14.75	8.42	470.26	1.06	2.61	4.62	483.76	
Mean	6.60	1.08	102.83	0.05	0.88	3.29	114.76	

The categories established for the RI index for classifying risk are: low ecological risk of potential contamination ($RI < 150$), moderate ecological risk $150 \leq RI < 300$), considerable ecological risk ($300 \leq RI < 600$) and very high ecological risk ($RI > 600$).

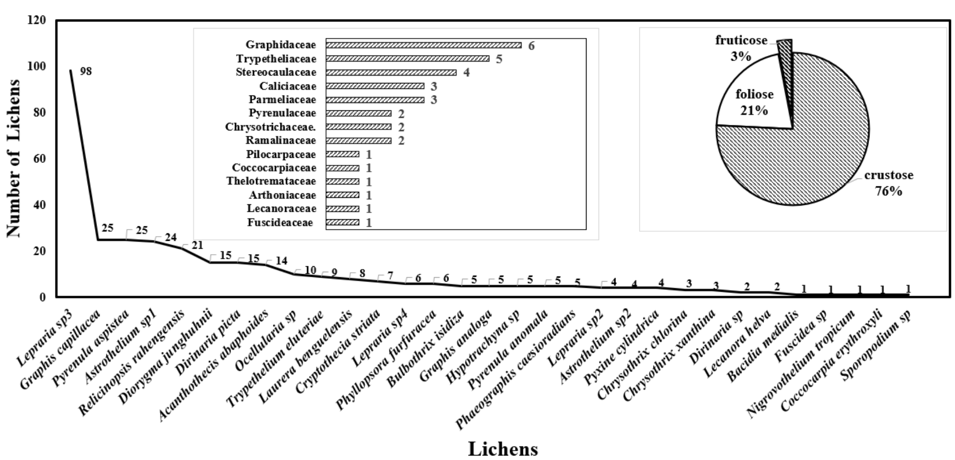

Figure 2. The 14 families were shown in bar graph. The lichen group was shown in circular graph while the genera and the number of lichens were shown in line graph.

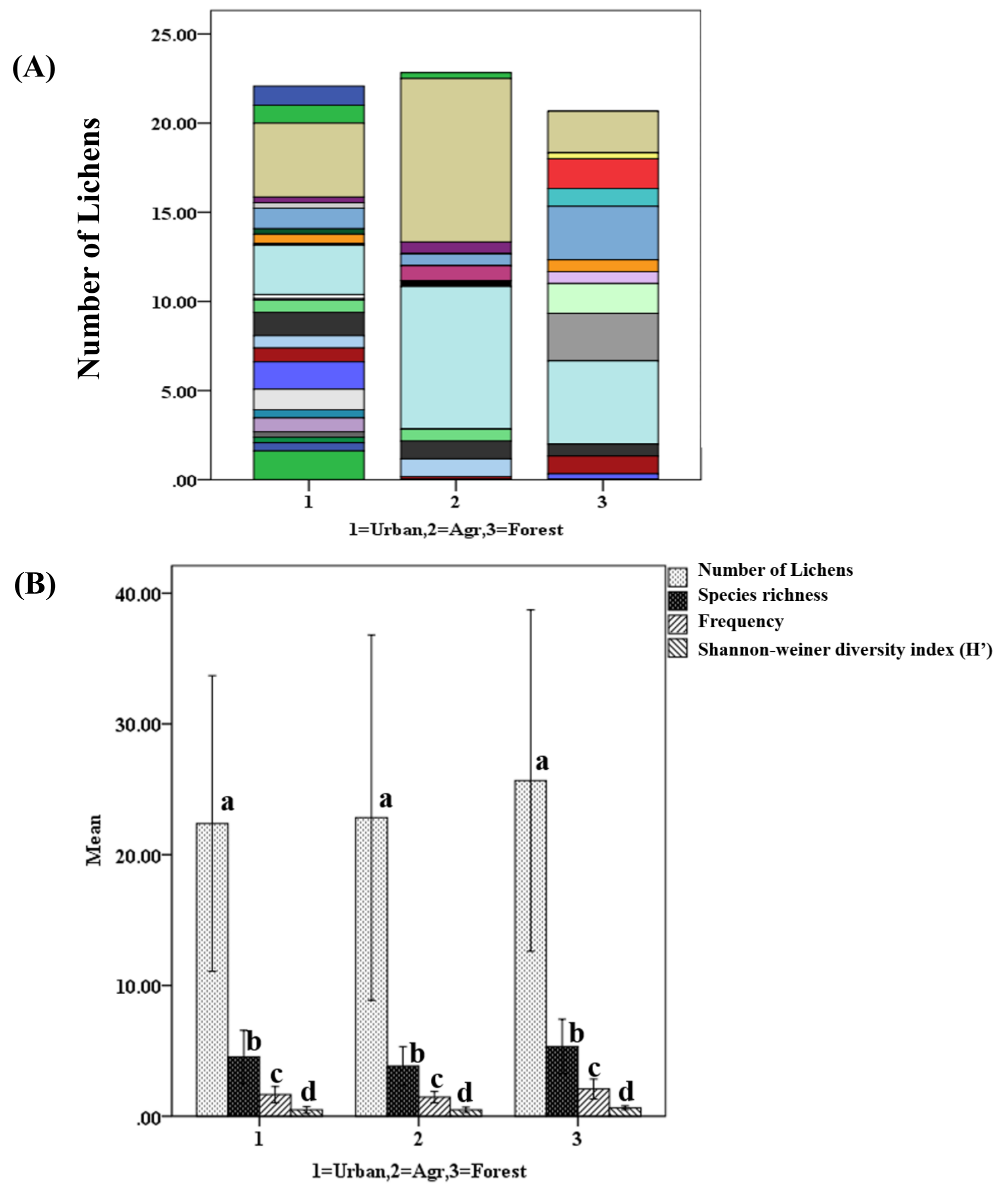

Figure 3. Lichen diversity in studied area. (A) the lichen species distribution in urban, agriculture and forest areas, (B) the diversity index was compared with Post Hoc Test (Duncan) in one way ANOVA. Means with the same letter of the alphabet are not significantly difference, at p value <0.05.

In addition, lichens such as the epigeic lichens *Cladonia rei*, can be used to indicate heavy metal pollution in soils. The loss of cell membrane integrity in *Cladonia rei* thalli elevated levels of heavy metals in the soil [28]. Some lichen species such as *Xanthoria candelaria*, *Lecanora muralis*, and *Xanthoria elegans* presented in urban areas could be used to monitor contamination of Pb, Cr, Cu, Cd, and Ni in roadside soils [28]. Based on lichen diversity studies that indicated a high index score (20-30) for areas located in the eastern part of Nong Saeng sub-district, the areas were assigned an index of C level (moderate level of pollution), although this area was nearby Kho Yai National Park. The mapping of IAP distribution showed that low IAP sites were agriculture areas. A lot of human activities affect lichen diversity, and their diversity decreased with higher human population numbers [29].

3.3 GIS mapping of spatial distribution of index of atmospheric purity (IAP) and potential ecological risk index (RI)

The distribution of heavy metal in Nong Saeng sub-district, Pak Phli district, Nakhon Nayok province was shown using distribution mapping. The geographic map of this area is shown in Figure 4A. Most of the areas are agriculture ones while the north of area is close to the national park. The atmospheric environment assessment expressed as index of atmospheric purity (IAP), which was based on lichen diversity, showed a high index score (20-30) in the eastern area of Nong Saeng sub-district, and this area was nearby Kho Yai National Park. Other areas had lower IAP (5-10) which indicated low lichen diversity in those areas. The mapping of index of atmospheric purities (IAP) distribution was shown in Figure 4B. High value of IAP was shown with dark green color which indicated that good air purity was in the north of the study area. Heavy metal content was expressed as the RI. These values in percentages were input into the attribute table in GIS data. GIS mapping of the spatial distribution of the potential ecological risk index (Figure 4C) suggested that the highest levels of potential ecological risks from heavy metals were in the south of this area. The south is mainly lowland, and most areas are connected to other urban areas. It is possible that these concentrations of heavy metals could be caused by the discharge of wastewater into low land, and especially in places of acidic soil in study area (pH 3.80-6.00). Heavy metal leaching and mobility increase when soil pH decreases, especially for manganese (Mn), cadmium (Cd), and lead (Pb) [30].

However, the high RI values were found in agriculture area (S18) and urban area (S19), while two areas (S10 and S16) contained moderate RI values corresponding to the low IAP value on IAP map.

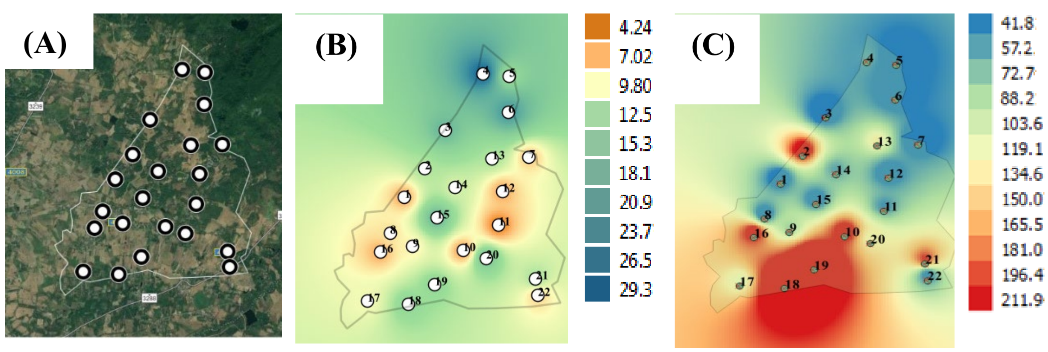

Figure 4. Spatial distribution of pollutants at site (A) geographic map of study area, (B) index of atmospheric purities (IAP) and (C) risk indexes (RI) of heavy metals in the surface soils

3.4 Analysis of linear correlation

The correlation of lichen number - heavy metal content was analyzed by the Pearson correlation. The linear correlations of number of lichens, the index of atmospheric purity (IAP), and potential ecological risk index (RI) are shown in Table 3. IAP was highly significantly positively correlated to population number of *Pyrenula aspistea, Astrothelium* sp., *Phaeographis caesioradians*, *Nigrovothelium tropicum*, *Chrysothrix xanthina*, and *Lepraria* sp., with correlation coefficients higher than 0.6 ($p < 0.01$). The RI was significantly positively correlated with amount of *Lepraria* sp. by Pearson correlation (0.621, $p < 0.01$), and with Spearman's rho and Kendall correlation coefficients of 0.474 and 0.395 ($p < 0.05$), while *Dirinaria* sp. was significantly negatively related to RI value. Lichens have been used as bioindicators in various atmospheric pollution assessments in several countries and metal accumulation in lichens has been used as a tool for assessing atmospheric contamination in a natural park [31]. At the macro-lichen level, the low land sites (both agricultural and urban areas) were habitats for *Dirinaria picta* and *Pyxine cocoes*. *Dirinaria picta* and *Pyxine cocoesare* were shown to be tolerant of pollution in urban sites [32]. The lichen, *Xanthoria candelaria*, *Lecanora muralis* and *Xanthoria elegans* were previously used to monitor pollution of traffic origin, and *X. candelaria*, in particular, is a very good indicator of pollution of traffic origin [33]. *Dirinaria* sp. was used as a bioindicator for airborne heavy metals at selected industrial areas in Malaysia. These results suggest the ability of lichen as an effective tool for assessing atmospheric contamination in natural areas.

Table 3. Pearson correlation analysis among trace elements, the index of atmospheric purity (IAP), number of lichens and potential ecological risk index (RI)

Lichens	**Correlations**					
	Pearson		**Spearman's rho**		**Kendall's tau_b**	
	IAP	RI	IAP	RI	IAP	RI
Lepraria sp.	0.035	0.621**	0.125	0.474*	0.104	0.395*
Chrysothrix xanthina	0.603**	-0.017	0.285	0.215	0.236	0.175
Astrothelium sp.	0.629**	-0.096	0.361	0.017	0.302	0.014
Dirinaria sp.	0.232	-0.230	0.278	-0.449*	0.236	-0.380*
Phaeographis caesioradians	0.629**	-0.096	0.361	0.017	0.302	0.014
Nigrovothelium tropicum	0.629**	-0.096	0.361	0.017	0.302	0.014
Pyrenula aspistea	0.649**	-0.105	0.499*	-0.118	0.421*	-0.092

** Correlation is significant at the 0.01 level.
* Correlation is significant at the 0.05 level.

4. Conclusions

The heavy metal content in soil was determined. Six heavy metals, Cr, Zn, Cu, As, Ni and Hg, were detected. The potential ecological risk index determination showed that 16 study areas were categorized as being of low risk of contamination, four areas were of moderate risk and two areas were of considerable risk. The lichen diversity analysis showed 23 species were in urban areas, 11 species were in agriculture areas and 13 species were in forest areas, and the IAP indexes were determined. GIS mapping of the spatial distribution of RI showed the highest levels of potential ecological risk in the south of the area. However, a population of some lichens can indicate contamination of heavy metals in soil. The relationship between index of atmospheric purity (IAP) and potential ecological risk indexes (RI) of heavy metal accumulation in the urban areas implied that long-range transboundary air pollution may be one of sources among many other sources of heavy metal contamination in some areas. The high values of RI in low land implied that it was possible that the concentrations of heavy metals could be affected by the discharge of wastewater into the low land regions, and especially into spots of acidic soil in the study area.

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