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PM10-Associated Heavy Metals and Health Risk Assessment in Charcoal Production Communities: A Case Study in Phitsanulok Province, Thailand

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Abstract

This research aimed to assess the concentration and health risks associated with exposure to particulate matter less than 10 µm in diameter (PM_{10}) in charcoal production communities in Phitsanulok Province. The study area was divided into 2 zones: residential and charcoal kiln areas. Samples were collected using a personal sampling pump, and the concentrations of heavy metals (Zn, Fe, Cd, Cu, and Pb) in the PM_{10} samples were determined using flame atomic absorption spectroscopy (FAAS). The results showed that the average concentration of PM_{10} in the kiln zone exceeded both Thailand's National Ambient Air Quality Standards (NAAQS) and the World Health Organization (WHO) recommendations. The mean concentrations of metals in PM_{10} were ranked as follows for the kiln zone: Fe (107.87 ng/m^3) > Zn (86.83 ng/m^3) > Pb (65.20 ng/m^3) > Cu (17.39 ng/m^3) > Cd (5.07 ng/m^3). In the residential zone, the mean concentrations of Fe, Zn, Pb, Cu, and Cd were 12.59, 16.43, 7.09, 1.86, and 1.08 ng/m^3, respectively. All heavy metals were found to be well within the permissible safe limits set by the US. EPA, except for Cd. The health risk assessment, based on the Hazard Quotient (HQ), revealed HQ values ranging from 0.83 to 11.26 in the residential area and from 1.87 to 14.41 in the kiln area, both of which are greater than 1.0, indicating potential human health risks.

Introduction

Charcoal is an important biomass-based material in the energy sector and is extensively used in developing countries. It increases local workers' incomes by creating employment, particularly in rural areas, and plays a role in the growth of the social economy (Asare et al., 2022; Njenga et al., 2013). Many types of wood, such as palm oil shells, bamboo, and mangroves, may be used to produce charcoal. The quality and productivity of charcoal vary depending on the raw materials used (Toan et al., 2023). Additionally, various equipment and processes, including furnaces, drum kilns, and conventional burning kilns, can produce charcoal. These methods include a complex carbonization process that finally converts the raw materials into charcoal (Toan et al., 2023). Charcoal production is dangerous to the environment as it utilizes ineffective carbonization

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processes (El-Batrawy, 2019; Njenga et al., 2013). Charcoal kilns discharge significant amounts of particulates and toxic gases (Latif et al., 2012; Pennise et al., 2001). The substances emitted from the charcoal kilns may negatively affect the environment and human health (Mencarelli et al., 2023; Toan et al., 2023). Charcoal smoke is a complicated mix of solid, liquid, and gaseous substances, including carbon monoxide, carbon dioxide, ammonia, nitrogen oxides, polycyclic aromatic hydrocarbons, sulfur oxides, and fine particulate matter (PM_{10} and $PM_{2.5}$) and heavy metals (Mencarelli et al., 2023). The United States Environmental Protection Agency (US. EPA, 2014) has classified heavy metals such as cadmium (Cd), chromium (Cr), copper (Cu), manganese (Mn), nickel (Ni), lead (Pb), vanadium (V) and zinc (Zn) as an environmental priority pollutant due to their typical presence in PM. There are three pathways through which humans are exposed to heavy metal-bounded PM: inhalation, ingestion, and dermal absorption. Heavy metals, including arsenic, may cause toxicity even at low levels of exposure. It has been established that various biochemical and physiological functions involve the presence of essential nutrients, including Co, Cu, Cr, Fe, Mg, Mn, Mo, Ni, Se, and Zn. However, these metals become toxic when present in excessive quantities. Other heavy metals, such as Cr, Cd, Hg, and Pb, are extremely harmful to human health (Das et al., 2023). These toxins can cause cancer, heart disease, and lung disease (Hamatui et al., 2016).

In Thailand, most of the charcoal produced is made from woody biomass and agricultural wastes. According to the country's energy consumption data from the Department of Alternative Energy Development and Efficiency, charcoal usage has grown over the previous five years. Charcoal is mainly used in the household sector, with about 4.2 million households (Kajina et al., 2018). The rural area represents about 97% of charcoal consumption (Kajina et al., 2018). Phitsanulok, located in the lower north region of Thailand, has been recognized for its Bang Krathum and Bang Rakam Districts, which have been identified as places with high charcoal production potential. The primary materials comprise different types of wood that are readily available, reasonably priced and contain a high carbon content. Tamarind trees are locally favored as a source of charcoal wood products. Charcoal is produced in this area using traditional kiln techniques, which do not have control systems set up to limit the discharge of substances during the carbonization process. The emissions originating from charcoal kilns have the potential to have adverse impacts on the environment. However, no research has been conducted in this area regarding the effects of charcoal production on human health. Thus, this study aims to analyze concentrations of heavy metals (Zn, Fe, Cd, Cu and Pb) in PM_{10} and determine the hazards to human health. In addition, a preliminary step is implemented to reduce the pollution released during the charcoal production process.

Materials and methods

1. Study area

The present study focused on charcoal production communities in the Bang Krathum District of Phitsanulok Province, which has a population of 45,918. The district lies between latitude 16°34′44″N and longitude 100°17′50″E (Fig.1). Total area of Bang Krathum District is 447 km^2 and its topography consists of flat, fertile lowlands. The main agricultural products of Bang Krathum are rice, sugar cane, fruits such as oranges, tamarinds, cassavas, as well as soybeans and mung beans. The villagers have an extra occupation of producing and selling tamarind charcoal. In order to produce charcoal, wood must be chopped, buried beneath a mound of the ground, and then ignited to produce a small amount of air. The pyrolysis combustion process might take many days or weeks. Based on the findings of our observation report, it was seen that workers involved with the production of charcoal needed to be adequately provided with the appropriate safeguards for their secure conduct of tasks at different stages of the process, particularly during the charring process. Therefore, the air quality condition assessed in this study provides evidence for using the examined contaminants for risk assessment.

2. Sample collection

A PM_{10} monitoring campaign was conducted at two villages as shown in Fig. 1. (1: 16°33′30″N 100°15′00″E and 2: 16°38′00″N 100°20′00″E), each sampling site was divided into residential and kiln zones. PM_{10} was collected using a personal air sampler pump (model 224-PCXR 8, SKC) with a size-selective cyclone and an average standard flow of 2.5 L/min. During May and June 2023, PM_{10} samples were collected using 37 mm filter quartz-fiber filters with a porosity of 2 µm over 24 h for 3 days a week. A total of 48 samples were collected from the four selected sampling locations. The concentrations of PM_{10} were measured using the NIOSH method 0600 by the procedure outlined in NIOSH (2003).

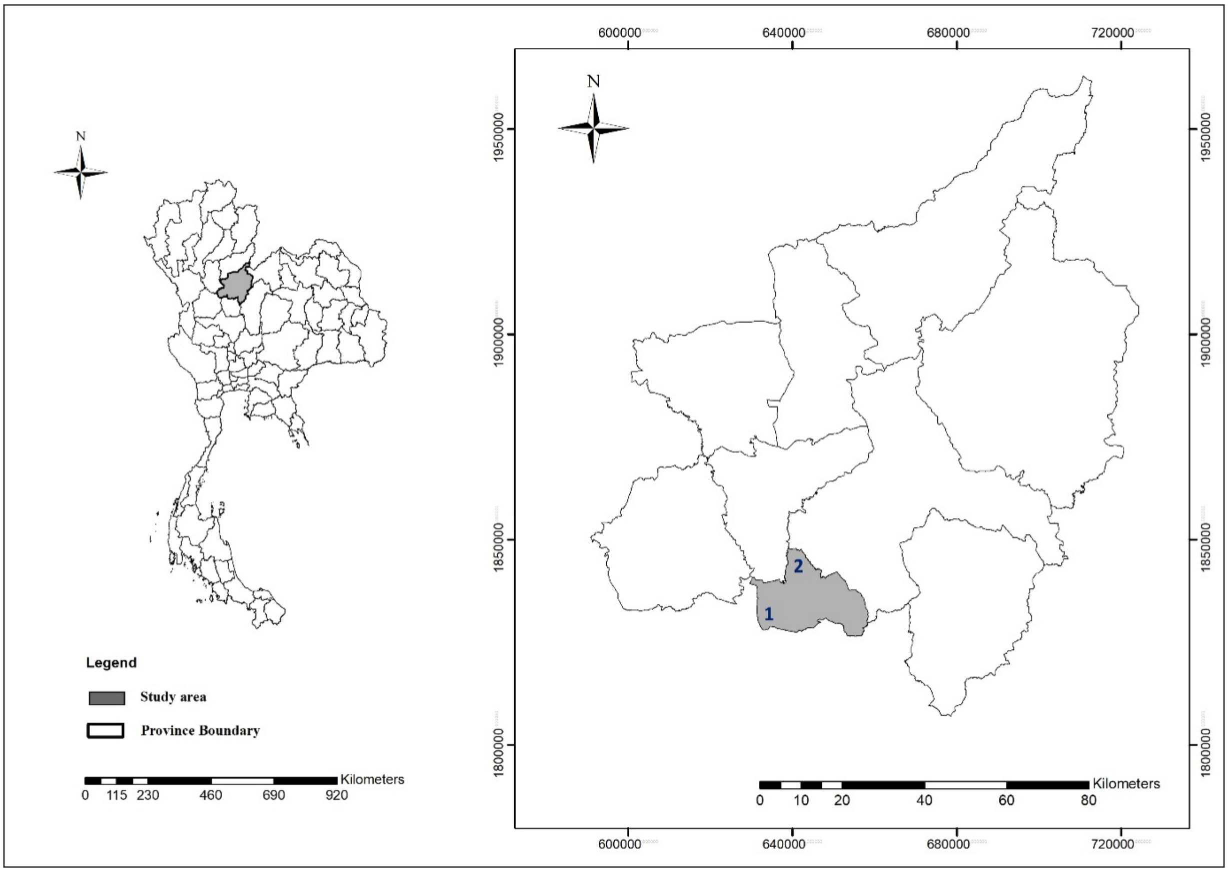

Fig. 1 Study area

The concentration of PM_{10} was calculated with the use of the gravimetric reference method, using a microbalance (Sartorius MC5, Germany) with a reading accuracy of 0.01 mg. The concentration of PM_{10} was determined by calculating the mass of the collected particulate matter and dividing the difference in filter weight before and after sampling by the observed air volume, accordance with the equation (1):

$$PM_{10}\ (\mu g/m^3) = (Wf - Wi) \times 10^6 / V \tag{1}$$

Where PM_{10} = PM_{10} mass concentration, 10^6 = conversion factor for g to µg, Wf = final weight of exposed filter, Wi = initial weight of unexposed filter and V = volume of air sampled.

Before weighing, the filters were conditioned in desiccators for 24 h, both before and after the sampling process. The data were collected inside a controlled setting, with a temperature of 23±5°C and a relative humidity of 45±5%. Following sampling, the filters were wrapped adequately in aluminum foil and kept in a refrigerator (4°C) until analysis.

3. Heavy metal analysis

Each weighted particle filter paper sample was cut into smaller pieces. The sample was extracted using a combination of nitric acid (HNO_3) and hydrogen peroxide (H_2O_2). Sample extraction was carried out using a microwave-assisted digestion apparatus. The heavy metal elements of the PM_{10} samples were determined using atomic absorption spectroscopy (AAS) (AA-6200, Shimadzu, Japan). All PM samples were analyzed for the following five elements: Zn, Fe, Cd, Cu and Pb. The procedure methods employed in the study are described by Srithawirat et al. (2016) and Srithawirat & Latif (2015).

The limit of detection (LOD) for each metal was determined by measuring the signal-to-noise ratio. The LOD values for Zn, Pb, and Fe were 0.1 ng/m^3, Cd was

0.03 ng/m^3, and Cu was 0.07 ng/m^3. Metal recoveries ranging from 98% to 115% were obtained using acid digestion. The validity of the blank filter sample was confirmed by subjecting it to unexposed filter sheets that had been treated with field samples. Heavy metal background contamination was quantified by removing the concentrations of field blank values. In this study, measurements were adjusted using background contamination.

4. Health risk assessment

This study assessed the health risk assessment for PM_{10}-bound heavy metals in the inhalation exposure route. The risk assessment analysis, as outlined by the US. EPA (2009) method, identifies three routes of exposure: dermal contact, inhalation, and ingestion. The hazard quotient (HQ) was used to determine non-carcinogenic risk. The target non-carcinogenic risk value was established at 1 (US. EPA, 2009), indicating that risk values less than 1 did not harm human health. The non-carcinogenic risk associated with PM_{10} was determined by calculating the reference dose (RfD) accessible in toxicological databases (de Oliveira et al., 2012; US. EPA, 2021). This analysis focused on three specific subpopulations: adults (>7 years), children (1–7 years), and infants (0-1 year). Based on the available reference values, the average daily dose (ADD) values were calculated using Equations (2) (US. EPA, 2009) to determine the daily intake of pollutants via the inhalation exposure route.

$$ADD = (C \times IR \times ET \times EF \times ED)/(BW \times AT) \quad (2)$$

The variables denoted as ADD, average daily dose (mg/kg.day); C, contaminant concentration in air (measured values were converted to mg/m^3) EF, exposure frequency (days/year); ED, exposure duration (years); IR, inhalation rate (m^3/h); ET, exposure time (h/day); BW, body weight; AT, time on average: ED, measured in years; 365 days/year; 24 h/day. Table 1 lists the exposure parameters utilized in the scenarios that were examined.

Table 1 Exposure parameters used in the calculations of the risk assessment

Exposure parameters	Adult	Child	Infant	References
IR, inhalation rate per person (m^3/h)	0.83	0.31	0.19	(US. EPA, 2009; 2011)
ET, exposure time per person (h/day)	24	24	24	(US. EPA, 2011)
ED, exposure duration (years)	24	6	1	(US. EPA, 2001)
EF, exposure frequency (days/year)	365	365	365	This study
BW, body weight (kg)	70	16	10	(US. EPA, 2011)
AT, averaging time (h)	210,240	52,560	8760	(US. EPA, 2011)

While all the contaminants that were examined have been classified as hazardous, only the non-carcinogenic risk was assessed using the hazard quotient (HQ) values, as described by Equations (3) (US. EPA, 2009), based on currently available toxicological data.

$$HQ = ADD/RfD \quad (3)$$

where ADD is the average daily dosage (mg/kg.day), RfD is the reference dose (mg/kg.day), and HQ is the hazard quotient (unitless). The computations were done using the RfD values. As mg/kg.day, PM_{10} equals 1.1×10^{-2} (Garbero et al., 2012; Gruszecka-Kosowska et al., 2021).

Results and discussion

1. Concentration of PM_{10} and its heavy metal composition

Concentration PM_{10} was determined using Thailand's National Ambient Air Quality Standards (NAAQ) and recommended by the World Health Organization (WHO). The standards revealed the 24-h quality guideline recommended for PM_{10}, set at 50 µg/m^3 by WHO and 120 µg/m^3 by NAAQ. The mean concentration of PM_{10} in the kiln zone exceeded the standards, whereas the PM_{10} in the residential zone did not exceed the permissible contents. Average, maximum, and minimum concentrations of PM_{10} in both areas are shown in Table 2. The concentrations of PM_{10} in the kiln and residential zones ranged from 103.58–1742.81 µg/m^3 and 9.04–110.37 µg/m^3, respectively. These charcoal production communities mainly produced tamarind wood charcoal. The mass concentration of PM_{10} varied depending on burning different types of biomass, ranging from 689 µg/m^3 for the honeycomb briquette to 10694 µg/m^3 for the raw powdered coal (Shao et al., 2016). El-Batrawy (2019) reported that the concentration of PM_{10} varied from 1127 µg/m^3 to 3716 µg/m^3 (El-Batrawy, 2019) from the area of charcoal-making kilns in New Damietta City, Damietta, Egypt, where earth mound kilns are used for charcoal production.

The statistical summary of PM10-bound heavy metal levels is presented in Table 2. In the kiln zone, the mean concentrations of metals in PM_{10} were as follows: Fe (107.87 ng/m^3), Zn (86.83 ng/m^3), Pb (65.20 ng/m^3), Cu (17.39 ng/m^3), and Cd (5.07 ng/m^3). In the residential zone, the mean concentrations of Fe, Zn, Pb, Cu, and Cd were 12.59, 16.43, 7.09, 1.86, and 1.08 ng/m^3, respectively. According to the US. EPA, the permissible limits for metal concentrations in air are 10000 µg/m^3 for Fe, 100 µg/m^3 for Cu, 500 ng/m^3 for Pb, and 0.2 ng/m^3 for Cd (Morakinyo et al., 2021). The Occupational Safety and Health Administration (OSHA) sets the permissible limit for Zn, specifically zinc oxide (dusts and fumes), at 5000 µg/m^3 in workplace air during an 8 h workday and a 40 h workweek (Plum et al., 2010). All heavy metals measured were well within the permissible safety limits, except for Cd. The average concentrations of the heavy metals, ranked from highest to lowest, were Fe > Zn > Pb > Cu > Cd in the kiln zone and Zn > Fe > Pb > Cu > Cd in the residential zone. These results indicate that air pollution in the investigated kiln area is high, with PM_{10} and its heavy metal composition being the primary pollutants contributing to elevated pollution levels. Notably, the study revealed that the mean concentration of PM_{10} in the kiln area was approximately 14 times higher than in the residential zone. The levels of Pb, Cu, and Cd in the kiln zone were about 8–9 times higher than those in the residential zone, while Fe and Zn levels were five times higher. These findings differ from those of a previous study by El-Batrawy (2019), which reported that the general order of heavy metal concentration in PM_{10} from charcoal kilns was Pb > Ni > Co > Cd > Zn > Fe > Cu. The presence of Pb, Ni, Cu, Cd, and Zn in the atmosphere is often linked to burning processes (Srithawirat et al., 2016; Yadav & Satsangi, 2013). The presence of Fe may be attributed to its natural occurrence in the soil (Yongjie et al., 2009). When Zn and Fe are combined, pollution may be traced to natural sources, burning of fuel, or incineration (Manalis et al., 2005).

2. Health risk assessment

The research identifies PM_{10} as the toxicological parameter value. As a result, the hazard quotient (HQ) values were computed for this hazardous substance. The study findings indicate a decreasing trend in possible ADD values across several age groups, including newborns, children, and adults, in both kiln and residential zones (Table 3). Table 4 presents the calculated HQ values for each subgroup. In the kiln zone, the mean HQ for PM_{10} were 0.67, 1.12 and 1.08 for adults, child and infant, respectively. The overall non-carcinogenic risk, determined by the HQ values for PM_{10}, exceeded the intended risk value of 1. Infants and children had the highest non-carcinogenic risk values above 1, suggesting that all subpopulations were exposed to the inhalation route. Furthermore, the results are fairly similar to those reported by Thongchom et al. (2021), according to their observations regarding the risk of PM_{10} and effects of sick building syndrome in office workers in Thailand, the mean HQ for indoor PM_{10} was 4.7±3.2 among all participants, which was more than one, thereby indicating an unacceptable risk for human health. Moreover, the binary logistic regression analysis showed that working period (more than 8 h daily) significantly increased the risk (Thongchom et al., 2021).

Furthermore, as humans breathe every day of their lives, the exposure factor in the inhalation exposure route reaches its maximum (equal to 1) (El-Batrawy, 2019). The HQ value of adults in the kiln zone was less than 1, although the workers who produce charcoal are often coated in charcoal residue and exposed to smoke. The actual exposure time is defined as the burning duration. The workers were exposed to the fumes emitted by combusting kilns for an average of 8 h daily for 2 to 4 weeks. Traditionally, a charcoal production plant in Thailand consists of several simultaneously operating kilns. Workers may carry out a variety of tasks. However, the main tasks are loading wood into the kiln,

Table 2 Mean, maximum, and minimum concentrations of PM_{10} (µg/m^3) and heavy metals (ng/m^3) for each zone

	Kiln zone			**Residential zone**			**Permissible safe limits**
	Mean ± stdev	Max	Min	Mean ± stdev	Max	Min	
PM_{10}	627.58±256.98	1742.81	103.58	44.78±25.30	110.37	9.04	120 (US. EPA)[a]
Cd	5.07±2.09	7.95	1.95	1.08±0.80	2.22	0.16	0.2 (US. EPA)[a]
Fe	107.87±96.50	227.88	10.26	12.59±11.26	27.18	0.75	10 000 000 (US. EPA)
Cu	17.39±2.38	21.75	BLD*	1.86±0.25	2.21	BLD*	100 000 (US. EPA)
Zn	86.83±69.81	270.35	13.47	16.43±10.06	47.77	3.12	5000 000 (OSHA)
Pb	65.20±63.15	172.22	BLD*	7.09±5.11	16.17	BLD*	500 (US. EPA)

Remark: *BLD: Below Limit of Detection; [a] exceed the limit.

carrying wood from the wood stack to place it beside the kiln, firing the kiln to begin the pyrolysis process, and collecting the cooled charcoal from the kiln and placing it into sacks or baskets (Pramchoo et al., 2017). The tasks performed by one worker may vary from day to day, and on a particular day, several duties may be conducted. Consequently, the severity of smoke and particulate exposure may differ. Extended work shifts and increased job demands can mitigate the adverse effects of air pollutants on the respiratory system. The observation revealed a lack of personnel use of respiratory protection equipment throughout the combustion procedure. According to previous studies, workers in charcoal kilns have been linked to respiratory diseases and lung injury (Juntarawijit & Juntarawijit, 2020; Kato et al., 2005; Tzanakis et al., 2001). Regular interactions may result in persistent health issues. However, the risk assessments derived from the present study indicate that inadequate air quality presented a significant hazard to the kiln area. Therefore, heating installations and the combustion of high-quality fuel can substantially enhance air quality and reduce health hazards.

Table 3 Calculated average daily dose (ADD) (mg/kg.day) for the inhalation pathway, based on exposure scenarios investigated in the study

	Kiln zone			Resident zone		
	Mean ± stdev	Max	Min	Mean ± stdev	Max	Min
Adult	7.4E-03±7.2E-03	2.07E-02	1.23E-03	5.3E-04±3.0E-04	1.31E-03	1.07E-04
Child	1.2E-02±1.1E-02	3.38E-02	2.01E-03	8.6E-04±4.9E-04	2.14E-03	1.75E-04
Infant	1.1E-02±1.1E-02	3.31E-02	1.97E-03	8.5E-04±4.8E-04	2.10E-03	1.72E-04

Table 4 Calculated hazard quotient (HQ) values in inhalation pathway, based on exposure scenarios investigated in the study

	Kiln zone			Resident zone		
	Mean ± stdev	Max	Min	Mean ± stdev	Max	Min
Adult	0.67±0.15	1.87	0.11	0.04±0.02	0.11	0.009
Child	1.12±1.07*	3.06	0.18	0.0±0.04	0.19	0.01
Infant	1.08±0.17*	3.01	0.17	0.07±0.04	0.19	0.01

Remark: *HQ is greater than 1.

Conclusion

The present study investigated the in-situ monitoring of PM_{10} and its heavy metal composition, namely Fe, Zn, Pb, Cu, and Cd, in traditional kilns located in Phitsanulok, Thailand. The analysis of air quality monitoring data revealed that the emissions originating from charcoal kilns were above the established threshold. The production of charcoal will likely contribute to the elevated levels of PM_{10} and heavy metals due to its origins in wood pyrolysis. The heavy metal examination revealed that the residential zone had the highest concentration of Zn, followed by Fe, Pb, Cu, and Cd, respectively. The kiln zone had the highest concentration of Fe, followed by Zn, Pb, Cu, and Cd, in the atmosphere. All these pollutants are, to varying extents, linked to the combustion processes. The charcoal workers are employed near charcoal kilns and are exposed to elevated levels of pollutants, which present a significant health hazard. Under the kiln exposure scenario, the maximum risk values for adults, children, and infants were much higher than the desired risk values. The findings indicated that the levels of heavy metals in the study area may be a threat to human health. It also gave additional insights into the pollution issues in the study area and served as a reminder of the necessity for more stringent strategies to regulate emissions. These results should be valuable for policymakers and stakeholders in developing methods to reduce trace metal concentrations in PM. Thailand should establish new air quality guidelines for heavy metals. In Thailand, there are currently no regulations regarding heavy metals other than lead. The government should also enforce the installation of modern technology that creates cleaner emissions to monitor combustion processes. This study suggests that future research should concentrate on understanding the impact of other PM components from various sources on health outcomes in different parts of Thailand.

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