Characterization of Polycyclic Aromatic Hydrocarbons and their Derivatives on Indoor Biomass Burning in Rural Thailand

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学位論文要旨

Characterization of Polycyclic Aromatic Hydrocarbons and their Derivatives on Indoor Biomass Burning in Rural Thailand
タイ農村の屋内バイオマス燃焼に由来する多環芳香族炭化水素とその誘導体の特性解析

Walaiporn Orakij

Abstract

Household fuel combustion for cooking is an important source of hazardous pollutants, especially in developing countries. This study focused on carcinogenic polycyclic aromatic hydrocarbons (PAHs) and their nitro-derivatives (NPAHs) generated from incomplete combustion of organic materials. Two field investigations were conducted in rural areas, Chiang Mai and Lampang, in upper northern Thailand. In rural households in Chiang Mai, $PM_{2.5}$ samples were collected both inside and outside the houses during cooking and noncooking periods. The most severe contamination was observed inside the house during cooking. Some data analysis demonstrated that the main source of indoor PAHs and NPAHs was biomass burning. During cooking periods, the total carcinogenic risks exceeded the WHO guideline values and would be classified as definite risks. In the study on personal inhalation exposure for rural residents in Lampang, inhaled $PM_{2.5}$ samples were collected using personal samplers. The most important factor concerning the exposure of rural populations to PAHs was cooking activity, especially the use of open charcoal fires. Furthermore, the personal inhalation cancer risks for all rural subjects exceeded the value recommended by the EPA, suggesting that the residents have a potentially increased cancer risk. The use of open charcoal fires showed the highest cancer risk.

Introduction

Approximately almost half the world's population, mainly in rural area of developing countries, still relies on solid fuels (e.g., wood, charcoal, crop residues, coal) for cooking and heating. These fuels have been typically burnt indoors with open-fires or inefficient stoves, resulting severe air pollution from incomplete combustion. According to WHO, 4.3 million deaths a year were attributed to the exposure to indoor air pollution. Emission from incomplete combustion of biomass burning contains hazardous pollutants such as polycyclic aromatic hydrocarbons (PAHs) and their nitro derivatives (NPAHs) which impact indoor air quality and human health. PAHs and NPAHs are of great concern because they are a human health hazard and a number of them are well-known carcinogenic and mutagenic compounds.

Lung cancer has been defined as a major health problem in Thailand. In particular, lung cancer incidence is significantly higher in upper northern Thailand than in other regions of the country and the highest incidence rate of lung cancer was recorded in Lampang province. The indoor air pollution may be a factor resulting in high incidence rate of lung cancer in northern Thailand. The use of biomass fuels with traditional open fire stoves for daily cooking in rural household is common. Although indoor air quality on residential scale caused by the use of biomass fuel for cooking has been of particular interest in recent years, there is a lack of information about indoor air pollution in this country. This study was conducted in rural areas in Chiang Mai and Lampang province in the upper northern Thailand. This study consists of two investigations; (1) Characterization of atmospheric PAHs and NPAHs from indoor biomass fueled cooking in rural Thailand (2) Evaluation of personal exposure of rural residents to fine particulate matters ($PM_{2.5}$), PAHs, and NPAHs in northern Thailand.

Study 1: PAHs and NPAHs from Indoor Biomass Fueled Cooking in Rural Thailand

The study of two rural houses in Chiang Mai was the first investigation of indoor air pollution from open-fire cooking with wood as the fuel. One house a large family of 17 people (House 1), and the other was a family of two (House 2). House 1 was built of concrete blocks and had very poor ventilation. Two traditional open stoves fueled by wood were normally used for cooking. House 2 was a wooden structure and had better ventilation than House 1 because of air exchange through the rough lattice-shaped material of the walls. Cooking in House 2 was done on an open fire.

Air sampling was conducted on two days, March 9-10 and 11-12 in 2012. Indoor (kitchen) and outdoor $PM_{2.5}$ samples were collected over 24 h on both days. All samples were collected using a personal air sampler at a flow of 1.5 L/min. A personal dust sensor was used for real time $PM_{2.5}$ monitoring at 10 sec intervals. The air samples from noncooking periods and the two meal preparation periods (dinner and breakfast) were collected separately. The $PM_{2.5}$ samples were extracted with dichloromethane (DCM) under ultrasonication, and then, the extracts were purified prior to determination of PAHs and NPAHs using HPLC with fluorescence detection (HPLC-FL). Levoglucosan (LG), a tracer of biomass burning was also determined.

Time-dependent changes in $PM_{2.5}$ counts inside both houses during the sampling period are shown in Fig. 1. There was a substantial variation with higher levels recorded during cooking periods due to the increase of $PM_{2.5}$ generation from biomass burning. During noncooking periods, the variation disappeared. The contamination levels in House 1 with the larger number of inhabitants were higher than those in House 2.

Fig. 2 shows the indoor and outdoor mean concentrations of total PAHs during cooking and noncooking periods. The indoor PAH levels during cooking periods in House 1 and 2 were higher than those in noncooking periods, suggesting that biomass burning was a major source of PAHs. The concentrations during each cooking period and in each house varied, probably due to differences in fuel consumption, cooking time and number of inhabitants. Second factor was the difference in

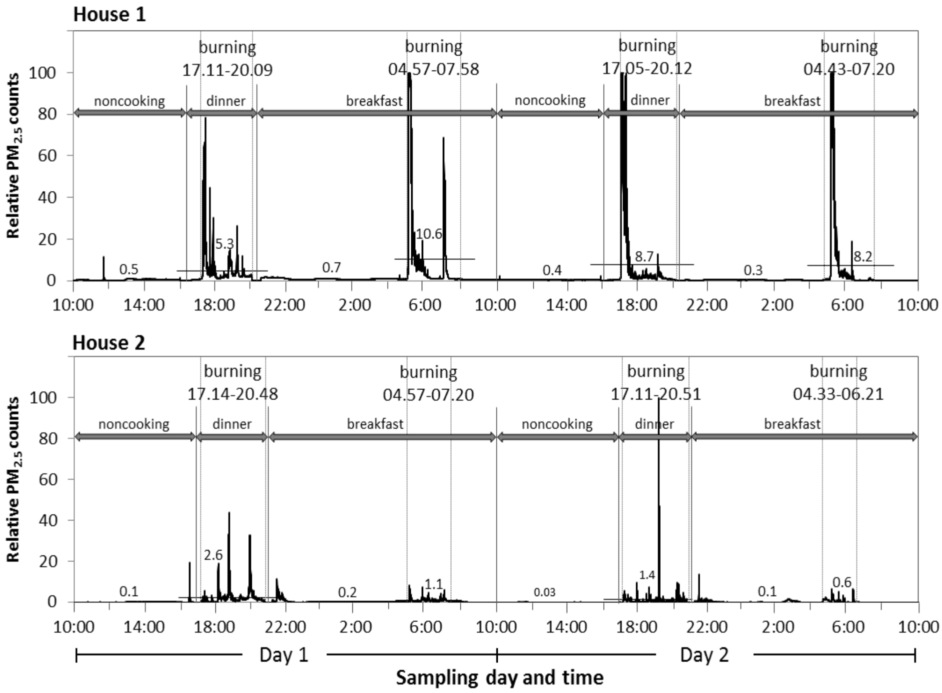

Fig. 1 Time-dependent changes in indoor $PM_{2.5}$ counts.

The highest level of $PM_{2.5}$ was taken as 100. Horizontal lines and the values in the figure show mean values of $PM_{2.5}$ counts during burning and nonburning periods.

house structure. The concentration of outdoor PAHs was also higher during cooking periods than noncooking periods. These suggest that indoor cooking increased the PAH concentrations of both indoors and outdoors. The mean concentrations of total NPAHs are shown in Fig. 2. The pattern of NPAH levels during cooking and noncooking periods was similar to that of PAH levels.

LG has been suggested as a marker of biomass burning. The LG levels in this study are shown in Fig. 2. The indoor air in House 1 on the first study day showed the highest LG level, as well as the highest levels of PAH and NPAH. PAH and NPAH concentrations showed significant correlations ($p < 0.01$) with LG, especially, in House 1 during cooking, suggesting that traditional open wood stove combustion for cooking was a major source of PAHs and NPAHs. The concentrations in House 2 during cooking periods were more weakly correlated, due to differences in fuel consumption and house structure.

The total excess lifetime risk of lung cancer from exposure to PAHs and NPAHs was calculated. The highest risk arose during indoor cooking in House 1 on the first study day. Furthermore, the carcinogenic risk associated with indoor biomass burning in this study exceeded the WHO recommended value, which suggests adverse health effects. The results revealed serious health risks from the inhalation of PAHs and NPAHs during cooking with the potential to increase the incidence of cancer in this area.

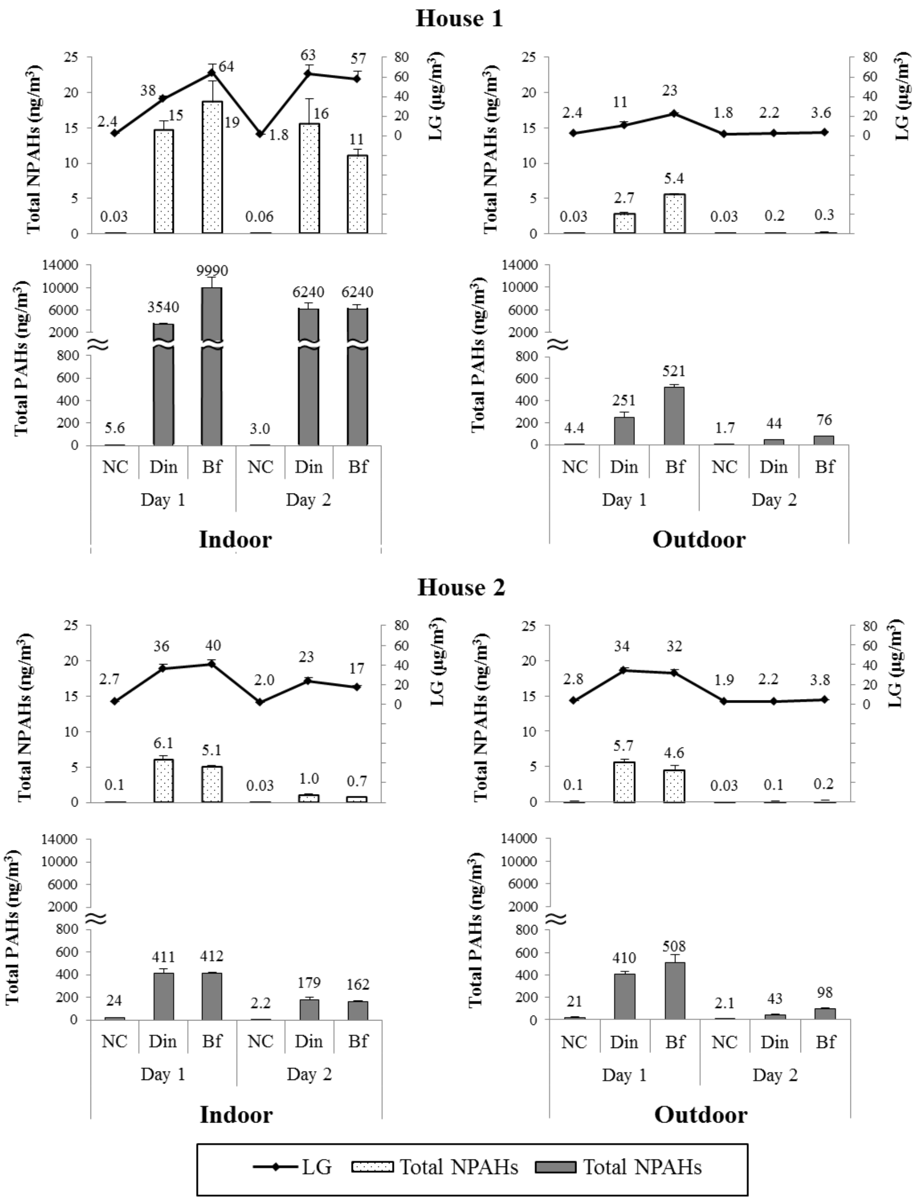

Fig. 2 Indoor and outdoor concentrations (mean ± SD) of total PAHs (ng/m^3), NPAHs (ng/m^3), and LG (μg/m^3) during cooking (dinner and breakfast) and noncooking periods.
NC: noncooking, Din: dinner, Bf: breakfast

Study 2: Personal Exposure of Rural Residents to PAHs and NPAHs in Northern Thailand

The second study was a new challenge to find important factors that may be related to the highest incidence of lung cancer in Thailand. A personal inhalation exposure and cancer risk assessment of rural residents in Lampang, Thailand, was conducted for the first time. Personal exposure to particulate matter ($PM_{2.5}$ and PM_{10}), PAHs, and NPAHs of rural residents living a typical lifestyle was investigated in addition to stationary air sampling in an urban area.

Personal sampling for the residents was conducted for three days between 9th and 12th of March in 2013. Fifteen rural residents were willing to participate in our study. They were considered with respect to smoking habits and then divided into three groups: smoker (n = 3), passive smoker (n = 4), and nonsmoker (n = 8). All subjects were asked to carry personal air samplers throughout the day. Moreover, they were required to be interviewed using a questionnaire survey in order to record their activities each day. In addition to personal monitoring, stationary air samples were collected in a central area of the city on 8th March. All samples were collected using a personal air sampler with an air flow of 1.5 L/min. The $PM_{2.5}$ samples were extracted with DCM, and then the determination of PAHs and NPAHs were performed by HPLC-FL.

The levels of $PM_{2.5}$, ΣPAHs, and ΣNPAHs in the personal exposure samples and urban ambient air are shown in Fig. 3. The levels of $PM_{2.5}$ in personal inhalation exposure ranged from 44.4 to 316 μg/m^3. Almost all personal $PM_{2.5}$ levels were higher than in the sample collected at the urban site (46.2 μg/m^3). The lowest level of personal $PM_{2.5}$ on the 1st day (44.4 μg/m^3) was higher than the maximum levels recommended by the WHO (25 μg/m^3) and the EPA (35 μg/m^3). This indicates that rural residents are exposed to $PM_{2.5}$, which can cause adverse health effects. The ΣPAHs and ΣNPAHs in the personal inhalation exposure samples ranged from 4.2−224 ng/m^3 and 120−1,449 pg/m^3, respectively, which were higher than that in urban ambient air.

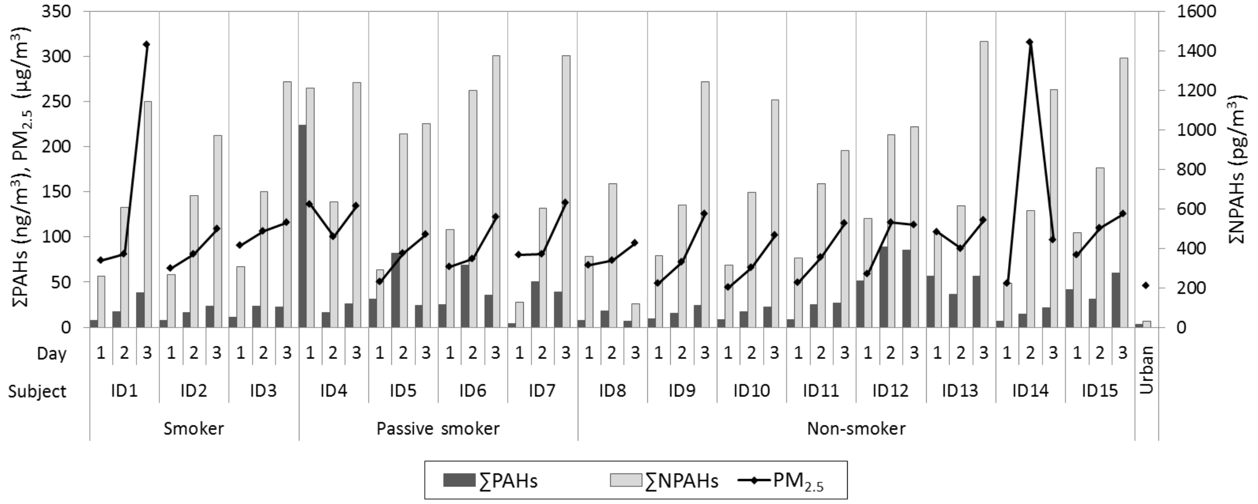

Fig. 3 Concentrations of $PM_{2.5}$, ΣPAHs, and ΣNPAHs in personal samples collected by fifteen rural residents for three days and urban ambient air.

The observed PM_{10} tendency was similar to that of ambient PM_{10} levels obtained from the monitoring station of the Pollution Control Department (PCD). Our urban PM_{10} data was consistent with the PCD data, suggesting that our PM_{10} data can bear comparison with the PCD data. Mean levels of PM_{10} during each day of personal exposure were markedly higher than the PCD data and our data at the urban site (Fig. 4). These results indicate that the rural residents are constantly exposed to PM that is derived from microenvironments through their individual activities in addition to ambient air pollution. PAHs showed no change in daily concentrations, whereas NPAH concentrations increased daily similarly to $PM_{2.5}$ or PM_{10} levels. The relationships among PAHs, NPAH, and PM concentrations suggested that different sources contributed to PAHs and NPAHs and may indicate that PAHs were strongly affected by individual exposure from microenvironments such as indoor air, whereas NPAHs were affected by daily variations in the atmospheric environment in the residential area.

Smoking behavior is an important factor with respect to personal inhalation exposure. Fig. 4, the $PM_{2.5}$ concentrations around smokers and passive smokers were not significantly higher than those for nonsmokers. However, the smoking behavior of the subjects did not contribute significantly to their exposure levels of $PM_{2.5}$. The median values of ΣPAHs and ΣNPAHs for smokers were not significantly higher than those for nonsmokers. Their levels for passive smokers were also not significantly higher than those for nonsmokers, suggesting that passive smokers were exposed to

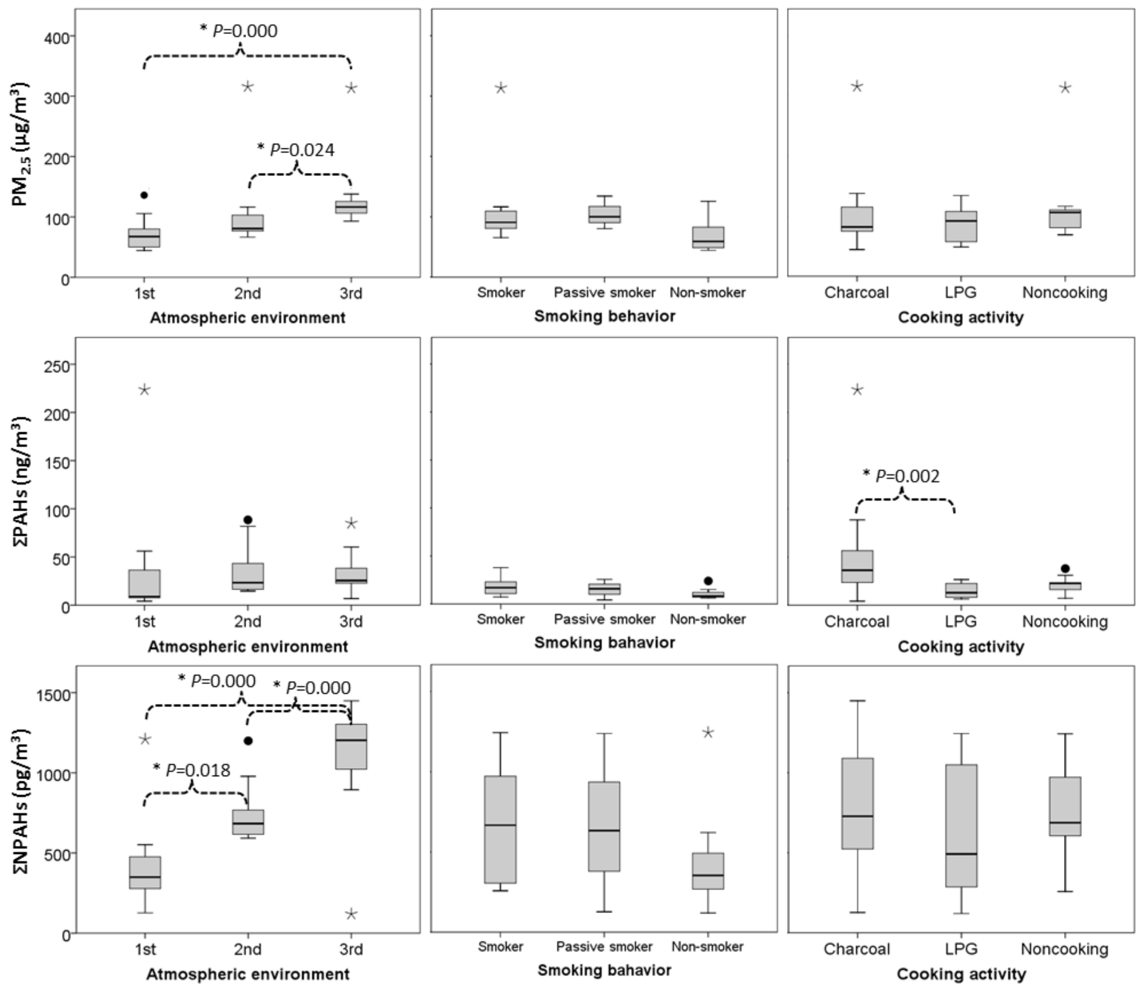

Fig. 4 Comparison of concentrations of $PM_{2.5}$, ΣPAHs, and ΣNPAHs among rural resident groups divided by factors related to their exposure: atmospheric environment, smoking, and cooking.

The box boundaries indicate the 25th and 75th percentiles.

them at almost the same levels as smokers. Since the cigarette consumption of the subjects or their family members was limited to 4−7 cigarettes or cigars, the effects of smoking behavior may be relatively small.

In terms of cooking as a factor, the use of an open fire inside an inadequately ventilated room considerably raises exposure to PM. The median values of ΣPAHs and ΣNPAHs for charcoal open fire users were higher than those for LPG stove users with significance for the PAH levels (Fig. 4), suggesting that charcoal as a combustion source significantly contributed to PAH exposure. The use of LPG as a cleaner fuel can reduce exposure to air pollutants at home. The ΣPAHs and ΣNPAHs for noncooking subjects were higher than those for subjects who cooked with LPG, presumably due to the existence of many smokers among the noncooking subjects.

Furthermore, personal inhalation cancer risks of all rural residents during the study period exceeded USEPA guideline value, suggesting that the exposure levels may have potentially increased cancer incidence of the residents. The use of charcoal open fire for cooking had the most severe cancer risk level, indicating that cooking with charcoal was a major factor of the exposure which may be related to lung cancer incidence in this study area. The use of clean energy such as LPG as cooking fuel can reduce the exposure of the residents to PAHs and NPAHs and consequently may decrease their cancer risk.

Conclusion

The studies on contribution of indoor air pollution from biomass burning to its health effects in rural residents provide valuable information about air quality management and evaluating the health risk caused by exposure to PAHs and NPAHs. The obtained results should lead to improvement of air quality and population health in developing countries. Possible ways to reduce air pollution in the area are to control open biomass combustion during dry season and to encourage rural residents to use clean fuel such as LPG or electricity for their daily cooking.

審査結果の要旨

〔審査経過〕平成 29 年 2 月 1 日に、学位申請論文に対する各審査委員による諮問を実施した。次いで平成 29 年 2 月 10 日に、審査方針に従って口頭発表（最終試験）を行い、終了後に開催した論文審査委員会において協議の結果、以下のように判定した。

〔審査結果〕発展途上国の農村では、未だに燃料として薪や作物残渣を使用した室内調理を行っており、バイオマス燃焼により発生する微小粒子状物質($PM_{2.5}$)は、発がん性の多環芳香族炭化水素(PAH)類のような有機汚染物質を含み、居住者の健康に影響を及ぼす可能性がある。タイ北部では、特に高い肺がん罹患率が報告されており、本研究ではタイ北部農村におけるバイオマス燃焼に起因する $PM_{2.5}$ や PAH 類による室内環境汚染、及び住民の個人曝露の特性を評価することを目的とした。木材を燃料としてストーブ調理を行っている家屋で、調理時に $PM_{2.5}$ や PAH 類の室内濃度が著しく増加し、バイオマス燃焼が発生源として大きく寄与することが明確になった。住民の個人曝露調査では、地域の大気汚染に加えて個人の生活様式が個人曝露に大きく影響し、特に調理の影響が観察された。本研究により、室内のバイオマス燃焼調理が地域住民の肺がんリスクを上昇させる可能性が示唆された。以上の成果より、審査委員会は学位申請論文が博士（学術）に値すると判定した。