

Gas Pollutant Emissions from Smoldering Incense Using FTIR

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Abstract

This study characterized the effects of smoldering incenses and combustion conditions on gaseous pollutant yields. Incense comes in three types: non-smoke (A), reduced-smoke (B) and traditional-smoke incense (C and D). Each incense type was burned in a test chamber with various combustion conditions (airflow rate and relative humidity). An extractive Fourier transform infrared (FTIR) was used to measure gas pollutants from smoldering incense in real time. Concentrations of methane, ethylene, methanol, formaldehyde and ammonia were measured using the IR spectra of smoldering incense samples. The resulting order of total emission factors of the identified gas pollutants (sum of methane, ethylene, methanol, formaldehyde and ammonia) were non-smoke < reduced-smoke < traditional smoke incenses. Total gas-pollutant emission rates and factors increased logarithmically as the airflow rate increased (2–28 L/min). Finally, the emission rates and factors of ethylene and methane decreased linearly as relative humidity increased (18–97%), while those for ammonia, methanol and formaldehyde increased. Results can be utilized to solve indoor air pollution problems caused by burning incense. Assuming that incense will continue to be burned when paying respect to ancestors, using incense made of low-volatility materials, with high carbon levels, low airflow rates and high environmental relative humidity can minimize gas-pollutant production.

Keywords: Incense; Smoldering; Emission rate and factors.

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INTRODUCTION

In Taiwan, incense is commonly burned to show respect to deities and ancestors. Epidemiological investigations have demonstrated a relationship between burning incense and lung cancer (MacLennan *et al.*, 1977), brain tumors (Preston-Martin *et al.*, 1982) and childhood leukemia (Lowergard *et al.*, 1987).

Burning incense is an incomplete combustion. Smoldering incense produces gas and particle pollutants. The gas pollutants produced include carbon monoxide, carbon dioxide, nitrogen oxygen, formaldehyde and volatile organic compounds. Moreover, the particles are typically < 1 μm and the particle surface areas absorb numerous organic compounds, such as polycyclic aromatic hydrocarbons (PAHs) (Chang *et al.*, 1997; Lin and Lee, 1998) and aldehyde (Schoental and Gibbard, 1967; Lin and Tang, 1994; Lin and Wang, 1994; Lee and Wang, 2004). Particles also contain heavy metals, acid materials, elemental compounds (ECs) (Wang *et al.*, 2006) and reactive oxygen species (ROSs) (Kao and Wang, 2002). Adverse health effects are likely associated with an extremely high number of particles, organic compounds, heavy metals, acid materials, ECs and ROSs.

Most existing studies have focused on emission characterization of gas pollutants for incense burned indoors, in temple environments, or in a large test chamber. Environmental conditions, such as temperature, humidity and air-flow rate, indoors and in temples were not controlled well or, in large test chambers, they were controlled at a fixed condition (Cheng *et al.*, 1995; Lee and Wang, 2004; Wang *et al.*, 2006). Hence, such studies could not determine the effects of environmental conditions and chemical compositions of incense on gas pollutant emissions. Additionally, past techniques for measuring gas pollutants primarily used simple real-time instruments to quantified gas pollutants. These instruments must use several sensors to analyze multiple gas pollutants simultaneously. Today, the new Fourier transform infrared (FTIR) instrument has two principal advantages: Multiple gas pollutants can be monitored in real time; and, IR spectra of samples can be analyzed and preserved for a long time. This study evaluates the effects of environmental conditions (air-flow rate and relative humidity) and various incense types (chemical compositions of materials) on gas-pollutant emissions using FTIR. The adverse health effects of the gaseous compounds identified by FTIR were studied using material safety data sheets and information from the International Agency for Research on Cancer, as well as data presented in other studies. Experimental results elucidate which environmental conditions and chemical compositions of incense produced the lowest emissions of gas pollutants to reduce the adverse health effects of smoldering incense.

METHODS

The incense was burned in a standard combustion system that has been used in previous studies and described in detail (Yang *et al.*, 2005; 2006). Briefly, the system was modified by adding a controlling device to maintain relative humidity in the test environment. The new system consisted of an incense smoke generation (smoldering) chamber in which incense was burnt, a test chamber and a device to control the relative humidity of the air-flow. A cylinder of air, after it had been made aerosol-free using HEPA units, was controlled by mass flow controllers. Then, the air entered the smoldering chamber (2 cm ID, 15 cm long). The incense was ignited and inserted into a fine stainless steel tube at the bottom of the smoldering chamber. Finally, the incense smoke was injected into a 7.8 L test chamber (10 cm ID., 100 cm long).

Three test conditions were prepared for the study. In the first, three incense types made from different materials were used: non-smoke (A, 3 mm in diameter); reduced-smoke (B, 2.2 mm in diameter); traditional smoke incense (C, 3.5 mm in diameter); and, traditional smoke incense (D, 2 mm in diameter). The reduced-smoke incense was made in Japan, and all others were made in Taiwan. Those were individually ignited and burned at a flow rate of 10 L/min. Under the second test condition, reduced-smoke incense smoldered at air-flow rates of 2, 6, 10, 15 and 28 L/min. The air-flow rates of 2, 6, 10, 15 and 28 L/min in the smoldering chamber (circle tube) were calculated to correspond to air velocities of 0.11, 0.32, 0.53, 0.80 and 1.49 m/s, as determined by the cross-section area of the circular tube. Under the third test condition, non-smoke incense (3.75 mm in diameter) was burned in 18, 62 and 97% relative humidity. The material in the non-smoke incense was the same as that for incense A. Additionally, an elemental analyzer (2400 CHN Elemental Analyzer, Perkin-Elmer, USA) was utilized to measure levels of carbon, hydrogen, and nitrogen. The heating value was measured with an oxygen bomb calorimeter (1271 Oxygen Bomb Calorimeter, Parr Instrument Company, USA). Before and after burning, the incense was weighed for calculation of net mass loss and burning rate. In this study, each test condition was performed three times.

An extractive FTIR (Illuminator, MIDAC, California, USA) was applied to measure gas pollutants released from smoldering incense. The spectrum was collected with a sampling regime using 64 co-averaged scans at 1 1/cm resolution. The scanning wavenumber range was 700–4500 1/cm. Total beam path length was 3 m. Gas pollutants were identified by comparing the IR spectra of samples with standard IR spectra obtained from the US EPA and a commercial infrared analysis company. Calibration technique was used to quantify identified compounds. The standards at various concentrations versus absorbance were analyzed to establish standard calibration curves (the range of concentrations was 2.2–113 ppm. The correlation coefficients of calibration curves were all > 0.995).

The emission rates and emission factors of gas pollutants were derived from the following equation, which is based on the conservation of mass.

$$V \times \frac{dC_i}{dt} = R \times E_f - Q \times C_i \quad (1)$$

Where: Q (L/min) is the air flow rate; R (g/hr) is the incense-burning rate; C_i (mg/m³) is the concentration of gas at a given time; V (m³) is the volume of the test chamber, and; E_f (mg/g) is the emission factor of each pollutant. $dC_i/dt = 0$ when generation and removal are in dynamic equilibrium. Equation 1 is then rewritten as follows.

$$E_f = \frac{Q \times C}{R} \quad (2)$$

In Eq. 2, the emission rate is defined as $Q \times C$, which is the flow rate multiplied by the equilibrium concentration of the pollutant. Then, the emission factor can be thought of as the specific emission rate normalized to the incense burning rate. The expression for the concentrations of gaseous pollutants was converted from ppm (v/v at 25°C, 1 atm.) to mg/m³ based on the ideal gas law.

RESULTS AND DISCUSSION

Incense burning rate

The incense burning rates for non-smoke (A), reduced-smoke (B) and traditional smoke (C and D) incense were 1.75 ± 0.01 , 1.02 ± 0.01 , 1.71 ± 0.03 and 1.35 ± 0.03 g/hr, respectively (Fig. 1). Additionally, the burning rate of reduced incense increased logarithmically as air-flow rates increased (2–28 L/min). The burning rate ranged from 0.87 to 1.09 g/hr under various air flow-rate conditions. The logarithm model was Y (g/hr) = $0.79 + 0.09 \ln X$ (L/min), $R^2 = 0.95$. On the other hand, the burning rate ranging from 2.35 to 2.10 g/hr showed a linear decline as relative humidity increased (18–97%) for non-smoke incense with 3.75 mm in diameter. The linear model was Y (g/hr) = $-0.0032X$ (%) + 2.4, $R^2 = 0.94$ (Fig. 1). Flowing air with a high relative humidity contains more water molecules than that with low relative humidity. The water molecules absorbed the heat that was released from the burning tip of the incense, reducing its temperature. Therefore, the incense burning rate at a high relative humidity was lower than that at low relative humidity.

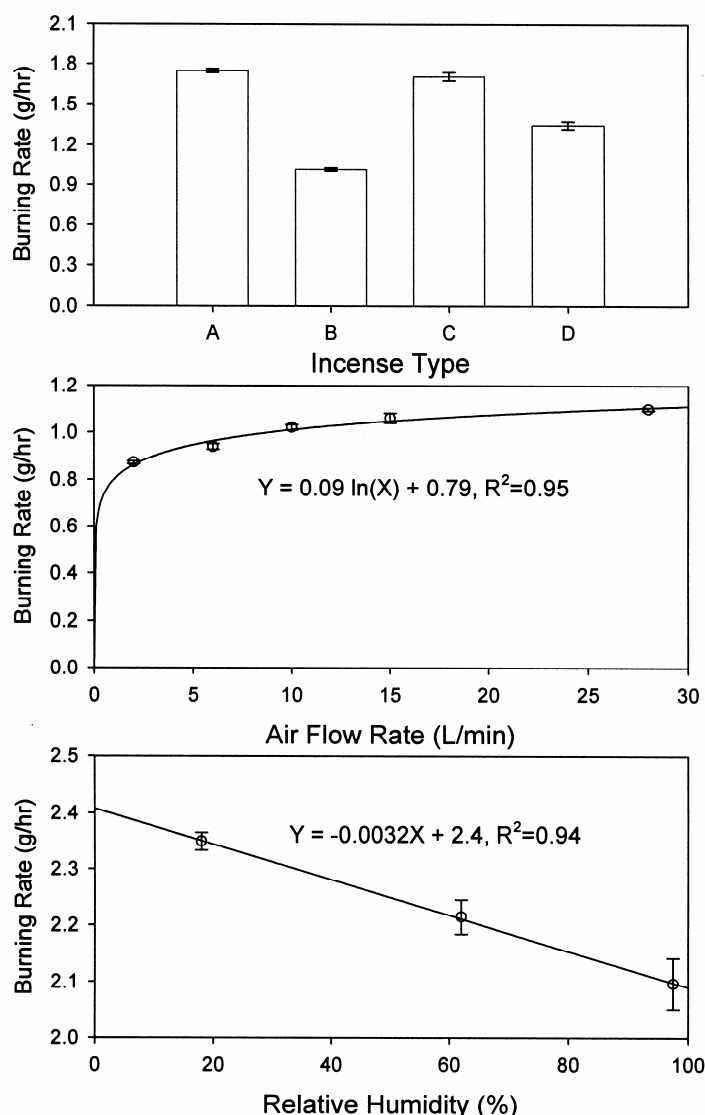


Fig. 1. Effects of incense type, air flow rate and relative humidity on incense burning rate.

Identification of gas pollutants

The IR spectra of samples compared with the standard picture from the US EPA and the infrared analysis company identified carbon dioxide, carbon monoxide, methane, methanol, ethylene, ammonia, formaldehyde and water in incense smoke. Ammonia, ethylene and methanol were identified in the 800–1250 1/cm spectral range, while methane and formaldehyde were identified in the 2600–3200 1/cm spectral range (Fig. 2). This study only discusses adverse health effects, emission rates and factors for methane, methanol, ethylene, ammonia and formaldehyde because carbon dioxide and monoxide have been investigated elsewhere. Hence, the adverse health effects for the compounds identified are based on the material safety data sheets (MSDS) (<http://www.iosh.gov.tw/msds.htm>), International Agency for Research on Cancer (IARC) data, and that in previous studies.

Information from IARC and MSDS has shown the adverse health effects of formaldehyde, which is probably a human carcinogen (2A). Additionally, concentrations of ammonia > 1 ppm irritate the eyes, throat and nose. High concentrations of ammonia resulted in serious burns and permanent damage to skin, lungs and eyes (Amshel *et al.*, 2000). Furthermore, low dose exposure to methanol affects the nervous system; initial symptoms are dizziness, headache, nausea, lack of coordination and confusion. The ceiling permissible exposure limit for formaldehyde set by the Occupational Safety and Health Administration (OSHA) is 1 ppm. The suggested 1h concentration of formaldehyde in indoor air in Taiwan is 0.1 ppm. The time-weighted average permissible ammonia and methanol exposure limits were 50 and 200 ppm, respectively. In this work, all tested formaldehyde concentrations exceeded the standard for smoldering incense, but those of ammonia and methanol were lower than the standards. On the other hand, inhaled high concentrations of ethylene can cause a lack of oxygen and anesthesia. Finally, methane is a very dangerous gas that is flammable and explosive, but not toxic.

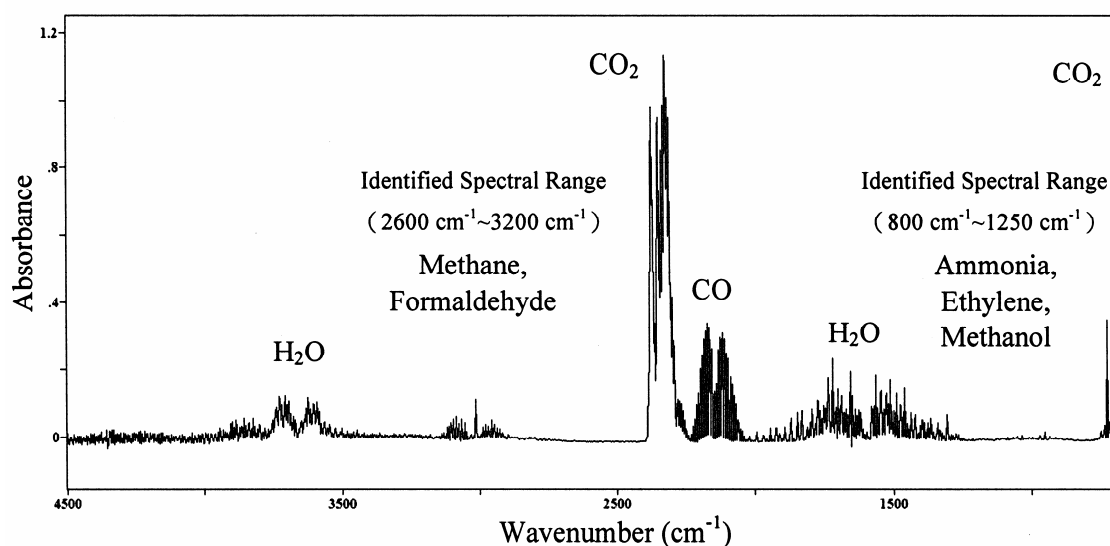


Fig. 2. Example of FTIR spectra of sample from smoldering incense.

Effect of incense chemical compositions on gas emissions

Changes to concentrations of gas pollutants versus time were recorded in real time in the sampling chamber. Measurement data demonstrated that individual gas pollutants in the test chamber reached equilibrium at about 3 minutes (Fig. 3). Therefore, emission rates and factors for methane, methanol, ethylene, ammonia and formaldehyde were calculated according to concentrations at equilibrium.

The fraction of individual pollutants under total-identified chemical amount (sum of methane, methanol, ethylene, ammonia and formaldehyde) was not consistent for each incense type. The methane fraction under total chemical amount was the maximum for the incense tested (Fig. 4).

Experimental results also showed formaldehyde concentrations for non-smoke (A), reduced-smoke (B) and traditional incense (C and D) were 11.31 ± 0.18 (A), 12.33 ± 0.67 (B), 41.88 ± 0.49 (C) and 34.21 ± 1.57 (D) ppm, respectively. Emission rates were 8.33 ± 0.13 (A), 9.08 ± 0.49 (B), 30.85 ± 0.36 (C) and 25.20 ± 1.15 (D) mg/hr, respectively. Emission factors were 4.76 ± 0.10 (A), 8.90 ± 0.41 (B), 18.07 ± 0.42 (C) and 18.72 ± 0.75 (D) mg/g, respectively. The lowest formaldehyde emission rates and factor were for non-smoke incense.

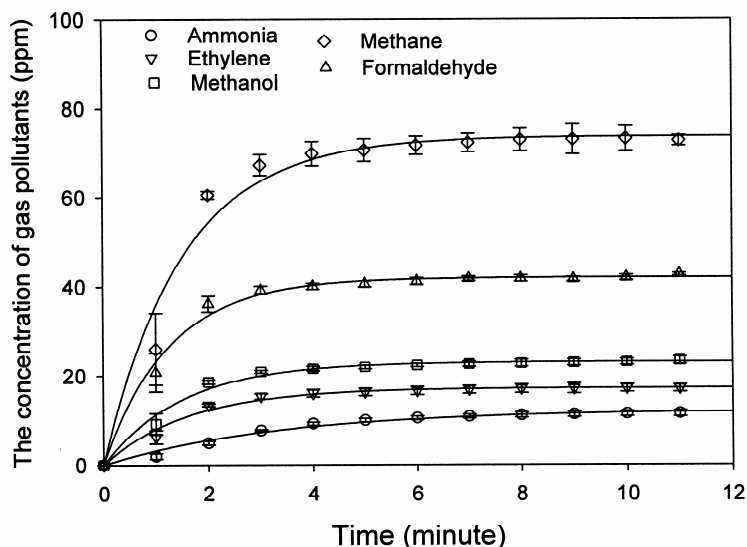


Fig. 3. Example of the variability of ammonia, ethylene, methanol, methane and formaldehyde concentrations versus time in the sampling chamber.

Methane concentrations for non-smoke (A), reduced-smoke (B) and traditional incense (C and D) were 62.23 ± 1.06 (A), 36.91 ± 0.89 (B), 72.70 ± 2.26 (C) and 64.70 ± 4.12 (D) ppm, respectively. Emission rates were 24.45 ± 0.65 (A), 14.50 ± 0.35 (B), 28.56 ± 0.89 (C) and 25.42 ± 1.62 (D) mg/hr, respectively. Emission factors were 13.97 ± 0.33 (A), 14.22 ± 0.47 (B), 16.73 ± 0.55 (C) and 18.87 ± 0.80 (D) mg/g, respectively. The lowest emission rates and factor of methane were for reduced and non-smoke incense.

The concentrations of methanol for non-smoke (A), reduced-smoke (B) and traditional incense (C and D) were 1.12 ± 0.07 (A), 5.25 ± 0.24 (B), 22.96 ± 0.67 (C) and 13.53 ± 1.00 (D) ppm, respectively. Emission rates were 0.88 ± 0.05 (A), 4.13 ± 0.19 (B), 18.04 ± 0.53 (C) and 10.63 ± 0.78 (D) mg/hr, respectively. Emission factors of methanol were 0.50 ± 0.03 (A), 4.04 ± 0.18 (B), 10.57 ± 0.51 (C) and 7.90 ± 0.70 (D) mg/g, respectively. Non-smoke incense had the lowest emission rate and factor of methanol.

The concentrations of ethylene for non-smoke (A), reduced-smoke (B) and traditional incense (C and D) were 4.61 ± 0.11 (A), 7.83 ± 0.08 (B), 16.98 ± 0.11 (C) and 22.05 ± 2.76 (D) ppm, respectively. Emission rates were 3.17 ± 0.08 (A), 5.38 ± 0.05 (B), 11.67 ± 0.62 (C) and $15.16 \pm$

1.90 (D) mg/hr, respectively. Emission factors were 1.81 ± 0.04 (A), 5.27 ± 0.11 (B), 6.83 ± 0.29 (C) and 11.25 ± 1.17 (D) mg/g, respectively. Non-smoke incense had the lowest emission rate and factor for ethylene.

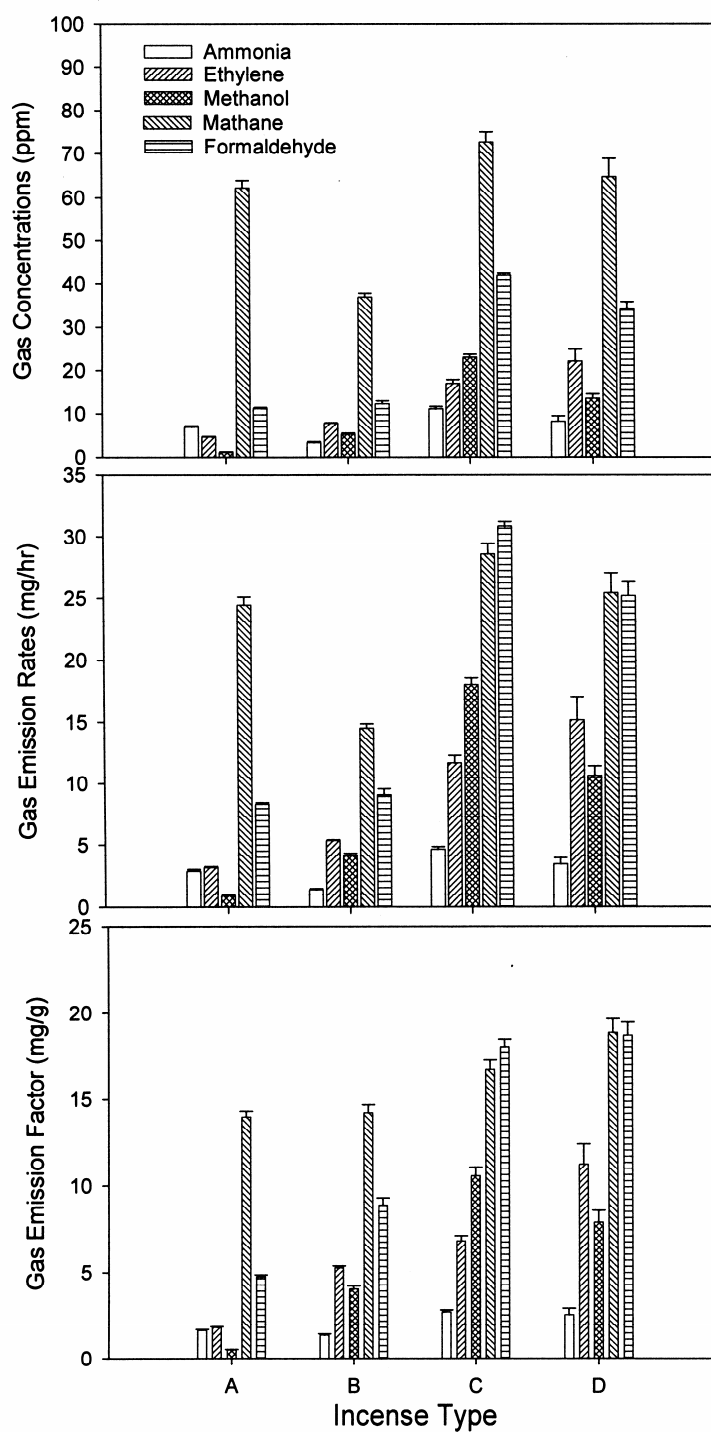


Fig. 4. Effect of incense type on the concentration, emission rate and factor of the ammonia, ethylene, methanol, methane and formaldehyde.

Ammonia concentrations for non-smoke (A), reduced-smoke (B) and traditional incense (C and D) were 6.86 ± 0.31 (A), 3.3 ± 0.22 (B), 11.1 ± 0.56 (C) and 8.19 ± 1.25 (D) ppm, respectively. Emission rates were 2.86 ± 0.13 (A), 1.38 ± 0.09 (B), 4.63 ± 0.23 (C) and 3.42 ± 0.52 (D) mg/hr, respectively. Emission factors were 1.64 ± 0.06 (A), 1.35 ± 0.09 (B), 2.71 ± 0.11 , (C) and 2.54 ± 0.38 (D) mg/g, respectively. Reduced-smoke incense had the lowest emission rate and factor of ammonia.

Finally, total concentrations of ammonia, ethylene, methane, methanol and formaldehyde for non-smoke (A), reduced-smoke (B) and traditional incense (C and D) were 86.13 ± 2.33 (A), 65.62 ± 2.08 (B), 165.6 ± 4.9 (C) and 142.69 ± 10.69 (D) ppm, respectively. Total emission rates were 39.69 ± 1.05 (A), 34.47 ± 0.17 (B), 93.76 ± 2.64 (C) and 79.83 ± 5.97 (D) mg/hr, respectively. Total emission factors were 22.69 ± 0.56 (A), 33.78 ± 1.25 (B), 54.91 ± 1.87 (C) and 59.27 ± 3.80 (D) mg/g, respectively. Non-smoke incense had the lowest amounts of total emission factors. Lin *et al.* (1994) reported that the ranges of formaldehyde concentrations from burning incense were 7.18-13.73 ppm. These values were consistent with the emission of formaldehyde from smoldering non-smoke and reduced-smoke incense; but, the traditional incenses yielded higher values than those determined by Lin *et al.* Lee *et al.* (2004) found methane concentrations of 1.11-4.57 ppm from burning 10 types of incense. These values differed in this investigation. Values on other gas pollutants (methanol, ethylene and ammonia) have not been reported in previous studies.

Conclusively, this data compared with elemental and molecular chemical phases of incense materials (non-smoke incense: carbon, 62%; volatility, 34%), (reduced-incense: carbon, 53%; volatility, 28%), (traditional incense C: carbon, 48%; volatility, 54%) and (traditional incense D: carbon, 44%; volatility, 76%) determined that smoldering incense with high carbon content and low volatility incense minimized the total gas emission rate and factor.

Effect of airflow rates on gas emissions of smoldering incense

The concentrations of ammonia, ethylene, methanol, methane and formaldehyde decayed exponentially as airflow rates increased (Fig. 5). The models of exponential decay were as follows: Y (ppm) = $5.73e^{-0.04X}$ (L/min) ($R^2 = 0.62$, ammonia); Y (ppm) = $17.5e^{-0.07X}$ (L/min) ($R^2 = 0.97$, ethylene); Y (ppm) = $11.2e^{-0.06X}$ (L/min) ($R^2 = 0.94$, methanol); Y (ppm) = $113.4e^{-0.1X}$ (L/min) ($R^2 = 0.89$, methane); and, Y (ppm) = $28.1e^{-0.06X}$ (L/min) ($R^2 = 0.84$, formaldehyde). However, the emission rates and factors of gas pollutants showed logarithm equations as the airflow rate increased. The logarithmic models of increased gas emission rates versus airflow rate were as follows: Y (mg/hr) = $0.07 + 0.64 \ln X$ (L/min), $R^2 = 0.89$, ammonia; Y (mg/hr) = $1.42 + 1.68 \ln X$ (L/min), $R^2 = 0.99$, ethylene; Y (mg/hr) = $1.03 + 1.40 \ln X$ (L/min), $R^2 = 0.94$, methanol; Y (mg/hr) = $6.4 + 3.4 \ln X$ (L/min), $R^2 = 0.97$, methane; and, Y (mg/hr) = $-0.62 + 4.99 \ln X$ (L/min), $R^2 = 0.91$, formaldehyde. Moreover, the logarithm increased models of gas emission factors were as follows: Y (mg/g) =

$0.32 + 0.51 \ln X$ (L/min), $R^2 = 0.79$, ammonia; Y (mg/g) = $2.36 + 1.22 \ln X$ (L/min), $R^2 = 0.94$, ethylene; Y (mg/g) = $1.77 + 1.03 \ln X$ (L/min), $R^2 = 0.81$, methanol; Y (mg/g) = $9.05 + 2.11 \ln X$ (L/min), $R^2 = 0.91$, methane; and, Y (mg/g) = $1.44 + 3.97 \ln X$ (L/min), $R^2 = 0.89$, formaldehyde. In conclusion, these data from smoldering incense with various airflow rates indicate that environmental combustion was controlled at low airflow rates and produced the lowest emission rates and factors for gas pollutants.

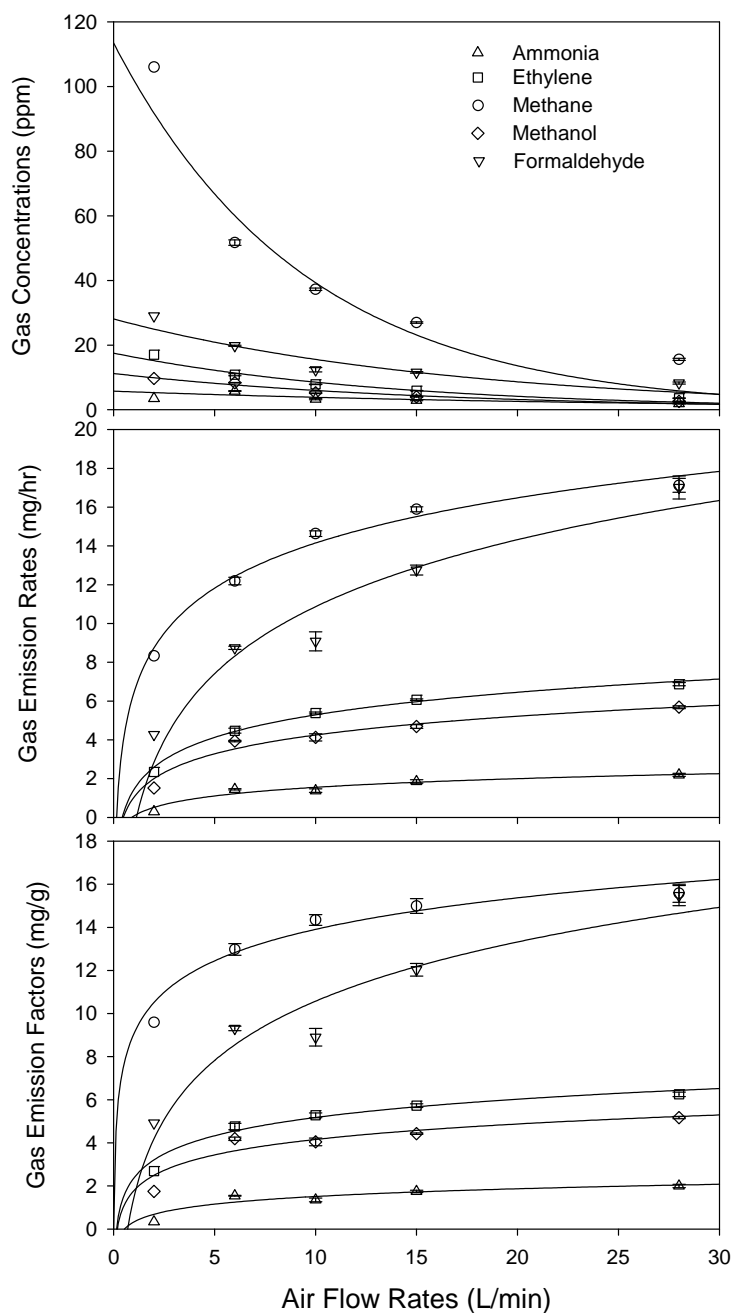


Fig. 5. Effect of air flow rate on the concentration, emission rate and factor of the ammonia, ethylene, methanol, methane and formaldehyde from reduced-smoke incense.

Effect of environmental relative humidity on gas emissions

Non-smoke incense (3.75 mm in diameter) was utilized as test incense for elucidating the effect of relative humidity on gas emissions. The same test data for reduced-smoke incense will be published elsewhere. The concentrations and emission rates for ethylene and methane demonstrated a linear reduced regression with high correlation as relative humidity increased

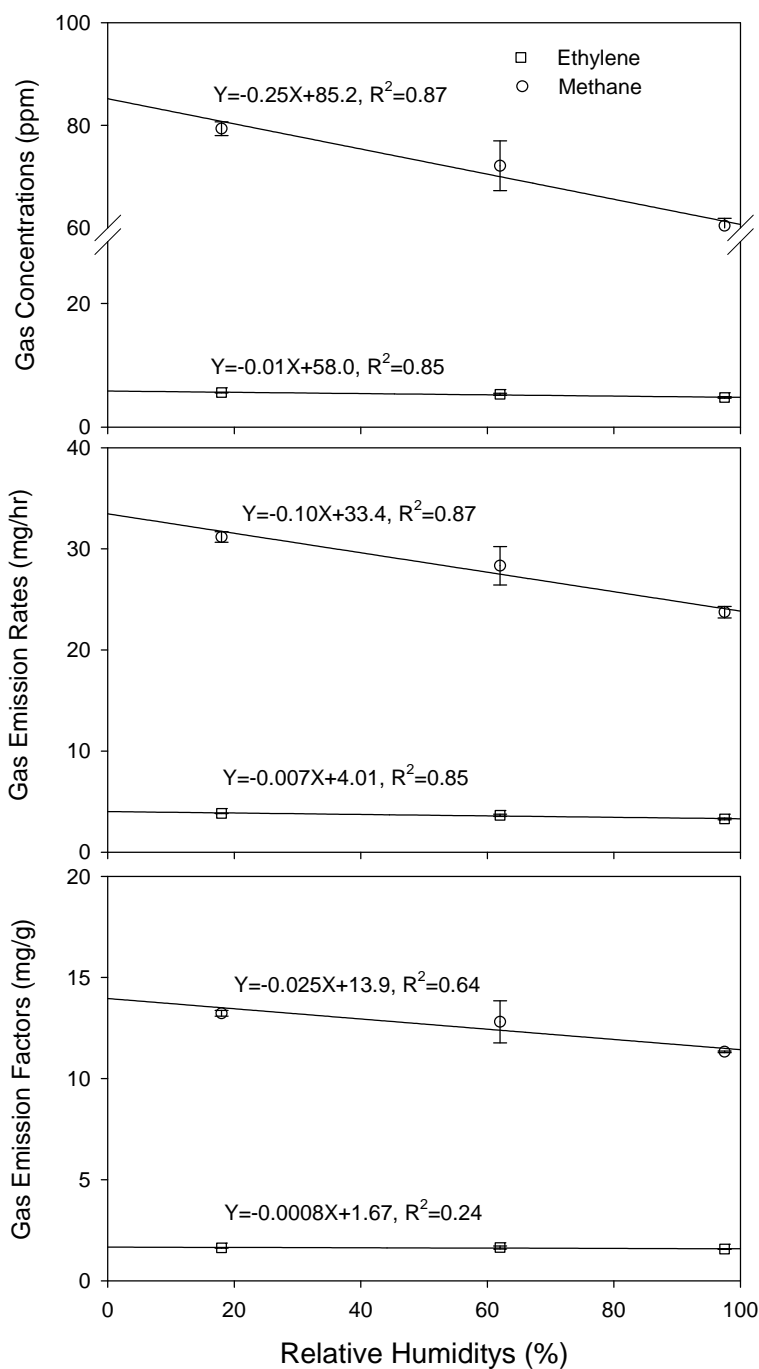


Fig. 6. Effect of relative humidity on the concentration, emission rate and factor of the methane and ethylene from smoldering non-smoke incense.

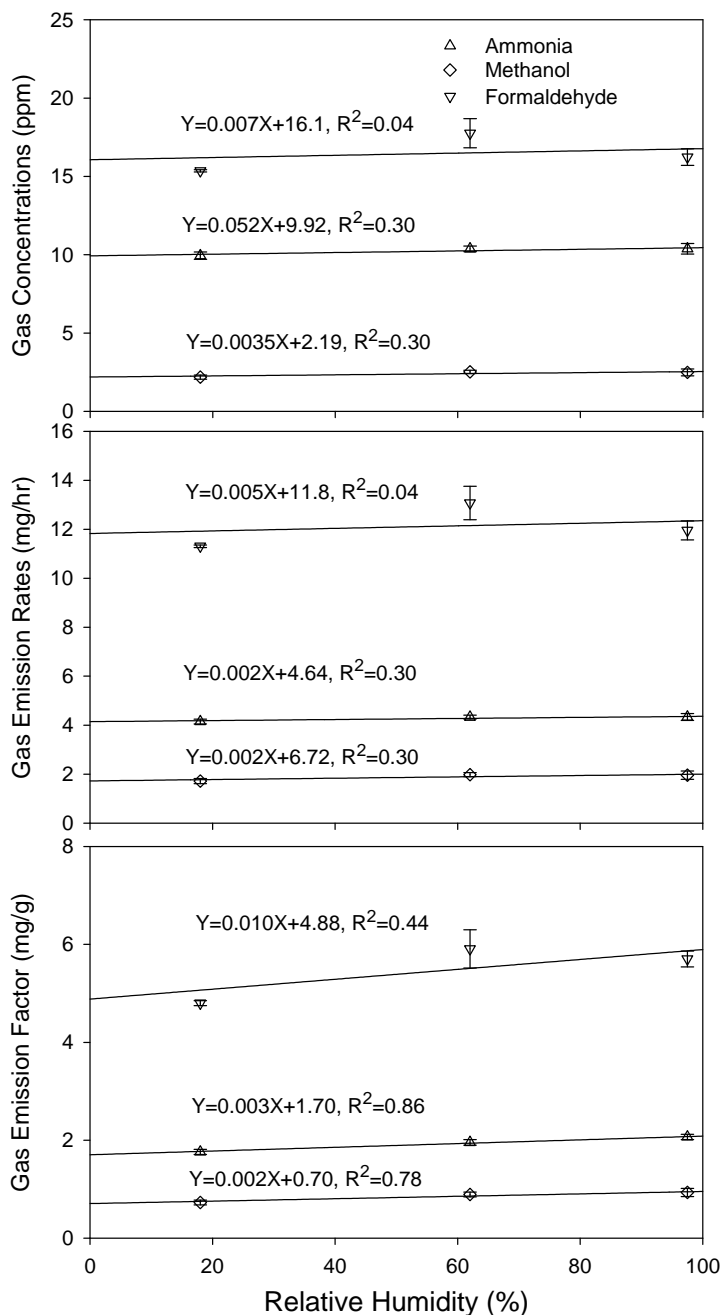


Fig. 7. Effect of relative humidity on the concentration, emission rate and factor of the ammonia, methanol and formaldehyde from smoldering non-smoke incense.

(Fig.6). The concentration models for ethylene and methane were Y (ppm) = $-0.01X$ (%) + 58.0, $R^2 = 0.85$ and Y (ppm) = $-0.25X$ (%) + 85.2, $R^2 = 0.87$, respectively. Emission rate models were Y (mg/hr) = $-0.007X$ (%) + 4.01, $R^2 = 0.85$ and Y (mg/hr) = $-0.10X$ (%) + 33.4, $R^2 = 0.87$, respectively. Emission factor models were Y (mg/g) = $-0.0008X$ (%) + 1.67, $R^2 = 0.24$ and Y (mg/g) = $-0.025X$ (%) + 13.9, $R^2 = 0.64$. Conversely, concentrations, emission rates and factors for ammonia, methanol and formaldehyde were linear and increased as relative humidity

increased (Fig. 7). Notably, the value of the slope of ammonia, methanol and formaldehyde were very small, meaning that changing relative humidity did not impact ammonia, methanol and formaldehyde emissions. The concentration models for ammonia, methanol and formaldehyde were $Y \text{ (ppm)} = 0.052X \text{ (\%)} + 9.92$, $R^2 = 0.30$; $Y \text{ (ppm)} = 0.0035X \text{ (\%)} + 2.19$, $R^2 = 0.30$ and $Y \text{ (ppm)} = 0.007X \text{ (\%)} + 16.4$, $R^2 = 0.04$, respectively. Emission rate models were $Y \text{ (mg/hr)} = 0.002X \text{ (\%)} + 4.64$, $R^2 = 0.30$, $Y \text{ (mg/hr)} = 0.002X \text{ (\%)} + 4.72$, $R^2 = 0.30$ and $Y \text{ (mg/hr)} = 0.005X \text{ (\%)} + 11.8$, $R^2 = 0.04$, respectively. Emission factor models were $Y \text{ (mg/g)} = 0.003X \text{ (\%)} + 1.70$, $R^2 = 0.86$; $Y \text{ (mg/g)} = 0.002X \text{ (\%)} + 0.70$, $R^2 = 0.78$ and $Y \text{ (mg/g)} = 0.010X \text{ (\%)} + 4.88$, $R^2 = 0.44$, respectively. This work demonstrated that the emission rates and factors of ethylene and methane were lowest at high relative humidity, according to the characterizations of gas emissions at various relative humidity levels.

CONCLUSIONS

This work demonstrated that smoldering incense produced toxic gas pollutants, such as formaldehyde, ammonia and methanol. Formaldehyde concentrations of four forms of incense significantly exceeded the ceiling limit on exposure (1 ppm, OSHA) and the 1h indoor air-quality standard of 0.1 ppm, suggested by Taiwan EPA. Although the ammonia and methanol concentrations were lower than the allowed time-weighted average permissible limit (OSHA) during the smoldering of a stick of incense, those of a smoldering bundle of many incense sticks might exceed the standard in low-ventilation microenvironments. Therefore, smoldering incense indoors adversely affects human health.

Smoldering incense produced combustible gases (methane and ethylene) that threaten environmental safety and hygiene. The concentrations associated with the lower explosion limits (LEL) for methane and ethylene are 5.1 and 3.1%, respectively. The auto-ignition temperatures of methane and ethylene are 537 and 520°C, respectively. When the concentrations of both methane and ethylene exceed their respective LELs and the temperatures reach their auto-ignition temperature at the same time, flaming combustion, or an explosion, will occur. When a stick of incense was burned in this combustion system, the methane and ethylene concentrations did not reach the LEL and the auto-ignition temperature, respectively, and flame combustion did not occur. However, the flame does occur during the smoldering of a bundle of many incense sticks in low-ventilation microenvironments, since the LEL and the auto-ignition temperature were reached simultaneously.

Hence, people must avoid burning incense at home or at temples. If people continue burning incense to pay respect to ancestors or gods, we suggest using smoldering incense with low-volatility materials, high carbon levels, low airflow rates and high environmental relative

humidity to minimize production of gas pollutants. These experimental findings have important health implications in helping to reduce the health risks from exposure to smoldering incense.

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