

Article

Chemical Characterization and Health Risk Assessment of Particulate Matter from Household Activities in Bamako, Mali, Western Sub-Saharan Africa

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Abstract: Household particulate matter (PM) is a major health concern, especially in developing regions, where biomass fuels are used quantitatively in households. Additionally, the combustion of incense (ICS) and insecticide (IST) is common. This study characterized the PM chemical composition to evaluate its health effects, as such information is lacking in developing regions, including Bamako, Mali. The composition of PM emitted from typical household activities, including cooking and combustion of ICS and IST, was characterized. These contained ions, organic carbon (OC), elemental carbon (EC), and metals. The results revealed that the chemical composition varied with emission source and combustion conditions. The dominant ions were Ca²⁺ (charcoal cooking), K⁺ (wood cooking) and F⁻ (in ICS and IST). The OC/EC ratio for IST, ICS, wood, and charcoal cooking was 59, 30, 8, and 7, respectively. Moreover, US EPA (United States Environmental Protection Agency) health risk assessment models indicated a higher hazard index (≤ 6.04) than the recommended limit (1) for nearly all emission sources. Total-CR was higher than the lower boundary limit (10^{-6}) in adults and children. Wood cooking activity and ICS combustion exceeded the unacceptable limit (10^{-4}) in children. Our findings highlight the need to develop effective air pollution mitigation strategies for health safety.

Keywords: PM; household activities; chemical composition; exposure risk assessment

1. Introduction

Researchers have started to focus on air pollution because many diseases have been linked to exposure to air pollutants worldwide. Indoor air pollution is of crucial importance to health. Humans tend to spend 90% or more of their time in indoor microenvironments [1]. Common daily indoor activities such as cooking, heating, and household cleaning result in substantial air pollutant emissions. Several studies have highlighted the effects of air pollution on health, particularly particulate matter (PM). Chronic exposure to PM in household microenvironments increases the risk of diseases, including chronic obstructive pulmonary diseases, heart diseases, stroke, lung cancers, cataracts, and childhood pneumonia [2,3].

Household PM pollution is a leading risk factor of mortality and morbidity, particularly in developing countries. In 2021, more than 2.31 million deaths were attributed to indoor air pollution globally [4]. The situation is particularly alarming in Africa, where more than 800 million people are exposed to indoor air pollution from food preparation. In

low-and middle-income countries, people rely mostly on biomass (wood and charcoal) for cooking and heating [5]. Indoor air pollution from the use of such energy sources in inefficient stoves is a major health risk. Combustion of incense (ICS) is another source of indoor air pollution. Although the use of this product is an ancient practice, it remains common in Africa and Asia. Inhabitants in these regions use ICS for a variety of reasons, including religious practices, culture, tradition (perfuming clothing, providing a pleasant scent around houses), and meditation. Malaria is a serious issue in numerous developing cities. This infectious disease is communicable through the bite of the female *Anopheles* mosquito. Hence, preventing mosquito bites can reduce malaria transmission. People in these regions rely heavily on the use of insecticides (mosquito coils) to repel mosquitoes. However, the combustion of insecticides (IST) and ICS in households largely contributes to indoor PM emissions and accumulation, particularly under inadequate room aeration conditions. These products are associated with diverse health effects [6–8]. ICS smoke can affect different parts of the body such as the eyes, nose, throat, and skin. Burning ICS has been linked to respiratory problems, asthma, allergies, and wheezing, and has been identified as a factor that increases lung cancer, cardiovascular mortality, and stroke mortality [9]. Similarly, the use of mosquito coils has been associated with numerous health issues that vary in severity from irritation and allergies to chronic respiratory problems and cancers [7,10].

In Bamako, Mali, all these household emission sources are common. The primary household energy sources for cooking are wood and charcoal. These represent 90% of the national energy needs [11]. ICS use is an integral part of culture. Used by women in households, ICS is considered a significant part of womanhood, in addition to the reasons mentioned earlier. Additionally, IST is commonly used to prevent mosquito bites and malaria, among the leading causes of mortality. In fact, 1698 deaths were attributed to malaria in Mali as of 2020 [12,13]. Therefore, the population of this urban city is a major factor that affects PM exposure in household microenvironments.

Our previous study provided evidence that the level of PM_{2.5} was above the WHO (World Health Organization) standards in Bamako City. The exposure levels were 3- and 7.5-fold higher than the daily (25 µg/m³) and yearly (10 µg/m³) limits, respectively [6]. Cooking and the use of IST and ICS were identified as the most exposed household activities. The average daily values recorded during these activities were 42 ± 1 µg/m³ (cooking) and 207 ± 115 µg/m³ (IST/ICS). Regardless of toxicity, exposure to PM triggers human health. Toxic elements can also have harmful effects. Heavy metals that are largely detected in PM are persistent; hence, they can enter the food chain and bioaccumulate in organisms. They can cause damage to important organs, such as the brain, lungs, kidney, and liver, and deteriorate muscular and physical functions [14,15]. In addition to knowing the level of exposure to PM, investigating the chemical composition is necessary to evaluate health risks. To our knowledge, there are no published data on the chemical characterization of PM in household microenvironments and related risks in Bamako. Therefore, we initiated this study to advance our understanding of PM emitted from typical daily activities. The key objectives of this research were to determine the chemical composition of PM emitted from activities that produce the greatest exposure and to evaluate the possible health risks.

In this study, we collected PM on quartz and polytetrafluoroethylene (PTFE) filters at specific household emission sources and simulated the conditions of IST and ICS burning using an NILU filter holder system and analyzed the ions, organic carbon (OC), elemental carbon (EC), and metallic components. This study is the first to provide information on the chemical composition of PM from different household emission sources and their associated health risks. Providing evidence based on research is the key to initiating pollution mitigation strategies. Hence, we believe that our findings could be useful in promulgating effective plans for air pollution control in household microenvironments and for public health improvement in this region.

2. Materials and Methods

2.1. Description of Sampling Sites and Sampling Procedure

2.1.1. In Situ Samplings (Bamako)

The purpose of sampling in Bamako City was to gather information on the PM emitted in real-life conditions. We conducted sampling at specific emission sources identified as the main household exposure sources in our previous research [6]. When selecting sampling sites, we considered the emission sources and availability of energy supply. PM emitted from cooking and the use of ICS and IST was collected from a residential area in the neighborhood of ATT Bougou ($12^{\circ}61'15.5''$ N $7^{\circ}99'64.1''$ W). The samplings were conducted in two different households. The involved microenvironments in each household were the living rooms (12.4 m^2) on the second floor and kitchens (semi-open) on the first floor. In Mali, as in many other countries in Western Sub-Saharan Africa, the kitchen is in the house yard. Cooking is performed in an open space or semi-open kitchen. Additionally, an NILU filter holder system was used for the particulate matters collection. Several devices were used as a part of the one-stage NILU filter holder system (Figure S1). This consisted of an open-face inlet NILU filter holder ($47\text{ mm}\phi$), a flow meter (RK1600R-10-S-Air-5, MonotaRO Co., Ltd., Amagasaki, Japan) to control the sampling flow rate, a pump (APN-085, IWAKI Co., Ltd., Saitama, Japan) to pull the air into the filter, and a transformer (DP-100F, Swallow Electric Co., Ltd., Osaka, Japan) to convert the electric voltage from 220 volts (electric power supply in Japan) to 110 volts (electric power supply in Mali). Quartz and PTFE filters were used at each emission source. Eight ICS and IST samples were collected at 0.30 to 0.50 m (Figure S2a) from the emission source. Charcoal and wood represent the main household energy sources in Bamako; hence, samples were collected separately during cooking activity using charcoal as cooking fuel and during cooking activity using wood fuel. Four samples for cooking activities were collected in the kitchens (semi-open). The NILU filter holder system was placed at 0.8 to 1 m from the cooking stove. The distance between the cook and cooking stove was approximately 0.5 to 0.7 m (Figure S2b). All samples were collected at 0.9 to 1 m above the ground. The flow rate was 9 L/min, and the sampling time was proportional to the duration of the activity. The duration of the cooking activity from fire ignition was between 1 h 30 min and 3 h. The ICS and IST products with the windows and door were closed were sampled for 2 h, after which the residents removed the combustion source as considerable amount of the products was burned. The cooking activity and the combustion if ICS/IST were not simultaneous in the households. Hence, only one combustion source was active for each sample collection. Additionally, for the samplings in the household microenvironments, three-day 24 h samplings were performed at a typical cross road to investigate the exposure risks from PM at traffic sites in Bamako City. To compare the health risks from household PM exposure to roadside ones. Samples were collected on the second floor of a building (3.80 m from the ground). The sampling site was influenced by traffic and other common anthropogenic activities such as uncontrolled incineration, street food preparation, construction activities, and livestock.

2.1.2. Laboratory Samplings

Laboratory samples simulating IST and ICS burning were used to confirm the PM chemical composition originating from ICS and IST and to evaluate the difference between the ICS and IST products used in Mali and Japan. Various brands of ICS and IST (mosquito coils) were burned in a tube furnace, and the emitted PM was sampled using a NILU filter holder system similar to that used in situ. The system consisted of a pump (APN-P215MV-1, Iwaki Co., Ltd., Tokyo, Japan), a flowmeter (DC-5, Shinagawa Co., Ltd., Tokyo, Japan), an NILU filter holder ($47\text{ mm}\phi$), a quartz tube (70 cm length and 2.2 cm inner diameter), a tube furnace, and a temperature control unit (DSM83, Shimaden Co., Ltd., Tokyo, Japan). Twenty-two samples were collected. Approximately 1–2 g of ICS or IST sample was placed in a quartz sample boat ($45\text{ mm} \times 20\text{ mm} \times 9\text{ mm}$), which was set at the center of the quartz tube. The ICS and IST burning experiments were conducted in a closed system setting (closed-face inlet) at a flow rate of approximately 3 L/min and temperature varying

between 350 and 450 °C. At the inlet of the quartz tube, indoor air was introduced through a silica gel tube, purafil tube, and active charcoal tube to purify the air. The burning particles were collected on quartz (2500QAT-UP, Pall Co., New York, USA) or PTFE filter (T080A047A, Advantec Toyo, Ltd., Tokyo, Japan) in the filter holder (Figure 1).

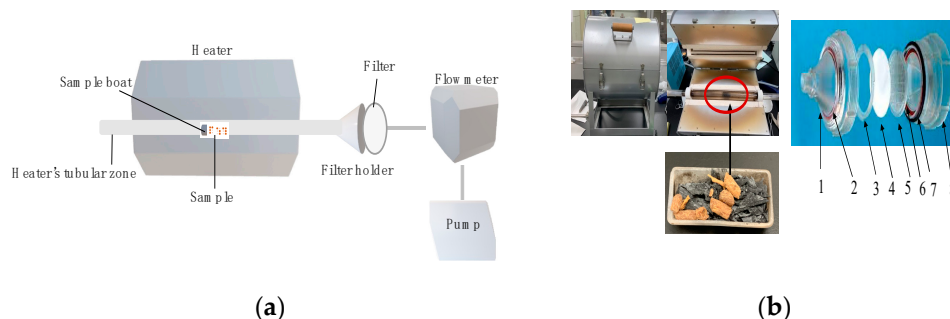


Figure 1. Laboratory sampling system. (a) Schematic diagram; (b) picture of the NILU filter holder system. See supplementary material for nomenclature (1–8).

2.2. Chemical Analysis and Quality Control

A total of 40 samples were analyzed. One-quarter of each filter was used to identify and quantify ionic and metallic compositions. After 30 min of extraction in 20 mL of distilled water with an electric shaker (SR-2w, Taitec Co., Saitama, Japan) and filtration using a pre-washed membrane filter (A045A025A, 25 mm in diameter and 0.45 μm pore size, Advantec Toyo Ltd., Tokyo, Japan), the samples were introduced in ion chromatography systems (Thermo Scientific Dionex TM ICS, 2100 and Dionex TM ICS, 1100, Inc., Waltham, MA, USA) to identify the anions (SO_4^{2-} , NO_3^- , Cl^- , F^- , PO_4^{3-} , NO_2^- , and Br^-) and cations (NH_4^+ , Na^+ , K^+ , Mg^{2+} , PM Ca^{2+}). The carbonaceous OC and EC in the PM samples were analyzed using a thermal and optical carbon analyzer from the Desert Research Institute (DRI 2001A, Desert Research Institute, Nevada, USA). The IMPROVE protocol is based on the liberation of carbon compounds at different temperatures and oxidation environments [16]. Only small portions of quartz filters were used for the carbonaceous measurements. A punch of 0.505 cm^2 was sectioned from each filtered sample. This was directly loaded into the instrument. An inductively coupled plasma mass spectrometer (ICP-MS; Thermo Fisher XSeries 2, Waltham, MA, USA) was used to determine metallic elements (Al, Li, Be, Na, Mg, K, Ca, Sc, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Y, Mo, Ag, Cd, In, Sb, Cs, Ba, Hg, Ti, Pb, Bi, Th, and U). One-quarter of the filters was digested in acids using a microwave oven (ETHOS EASY, Milestone General Inc., Shelton, CT, USA) and then dissolved in 15 mL of HNO_3 (electronic grade) at 1 mol/L. The sample preparation for chemical analysis has been described in detail elsewhere [17,18].

To ensure that the analytical laboratory instruments and procedures were effective, instrument checks and calibrations were regularly performed following the Acid Deposition Monitoring Network in East Asia (EANET) quality control and quality assurance procedures [19]. The reference material of artificial rainwater with different sample concentrations was tested by ion chromatography for anion and cation analysis. To evaluate the analytical protocol for metallic elements, the values from the measured elements were compared with those of the certified reference material (CRM) of urban particulate matter. The analytical results were in good agreement with the certified reference values within analytical error [17]. After the analysis, the values obtained from the blank filters were subtracted from the values obtained from the collected filter samples. Values under the detection limit were considered undetected (nd).

2.3. Health Risk Assessment

Health risk assessment is a useful tool for determining the possibility of harmful effects that might occur from exposure to heavy metals. In this study, the equations for

health assessment are adopted from the US EPA (United States Environmental Protection Agency) health risk assessment models [20].

2.3.1. Non-Carcinogenic Risks

A non-carcinogenic risk assessment determines health risks that do not include cancer. The health risk from metals not identified as carcinogenic was evaluated by determining the hazard quotient (HQ) for a single metal and/or the hazard index (HI) for multiple metals.

$$HI = \sum HQ \quad (1)$$

$$HQ = EC / (RfC \times 1000) \quad (2)$$

where HI is the hazard index, HQ is the hazard quotient, EC is the exposure concentration ($\mu\text{g}/\text{m}^3$), and RfC (mg/m^3) is the reference inhalation concentration.

$$EC = (CA \times ET \times EF \times ED) / AT$$

where

$$AT = ED \times 365 \text{ days} \times 24 \text{ h/day} \quad (3)$$

where EC is the exposure concentration, CA is the concentration of a metal ($\mu\text{g}/\text{m}^3$), ET is the exposure time (hours/day) proportionally to the duration of each activity, EF is the exposure frequency assumed to be 350 days (days/year), and ED is the exposure duration (years), and AT is the average time (hours).

Non-carcinogenic impacts may occur when the HI and/or the HQ are greater than 1, while harmful health impacts are not expected when the value is ≤ 1 [21].

2.3.2. Carcinogenic Risks

Carcinogenic risk (CR) assessment determines the risk of cancer development from exposure to toxic metals. CR for health purposes should range from 10^{-6} to 10^{-4} . A value higher than 10^{-6} represents a risk, and a value equal to or greater than 10^{-4} is unacceptable [22].

$$CR = EC \times IUR \quad (4)$$

where CR is carcinogenic risk, EC is exposure concentration ($\mu\text{g}/\text{m}^3$), and IUR is inhalation unit risk ($\mu\text{g}/\text{m}^3$).

The RfC and IUR values for each metal were obtained from the EPA Integrated Risk Information System (IRIS) [23].

According to the EPA, at least five metals are expected to be carcinogenic to humans: As, Cd, Cr (VI), Be, and Ni. Pb has been identified as a probable carcinogen. Therefore, in this study, the carcinogenic risks of these six metals were estimated. The non-carcinogenic risks of Cr, Mn, Ni, As, Cd, and Be were assessed. Pb was excluded from the non-carcinogenic risk assessment calculation because the RfC was not available.

3. Results

The total suspended particulate (TSP) collected on the quartz and PTFE filters was used to provide information on the chemical composition of the PM emitted from typical household activities.

Figure 2 shows the PM constituent and mass concentrations for different emission sources. The filters were weighed before sampling in the laboratory, in contrast to the in-situ sampling. Therefore, the mass concentrations of TSP are not shown in Figure 2a. Figure 2a illustrates the mass concentrations of the different PM constituents in the samples collected from household microenvironments in Bamako City. IST presented the highest PM concentration, followed by cooking using wood fuel, ICS, and charcoal fuel. Figure 2b presents the PM constituents, mass concentrations, and mass concentrations of TSP in ICS and IST samples collected in the laboratory. The mass concentration of TSP was highest

for the Japan IST, followed by the Mali IST, Mali ICS, and Japan ICS. Figure 3a,b show the contribution of the PM constituents in the samples collected in situ and in the laboratory, respectively. Carbonaceous compounds, especially OC, were the dominant constituents of all the samples. Although carbonaceous matter had the highest contribution, a visible difference was observed in the distribution of PM constituents for different emission sources. The distributions of ionic compounds, carbonaceous elements, and metallic elements are discussed in the following sections.

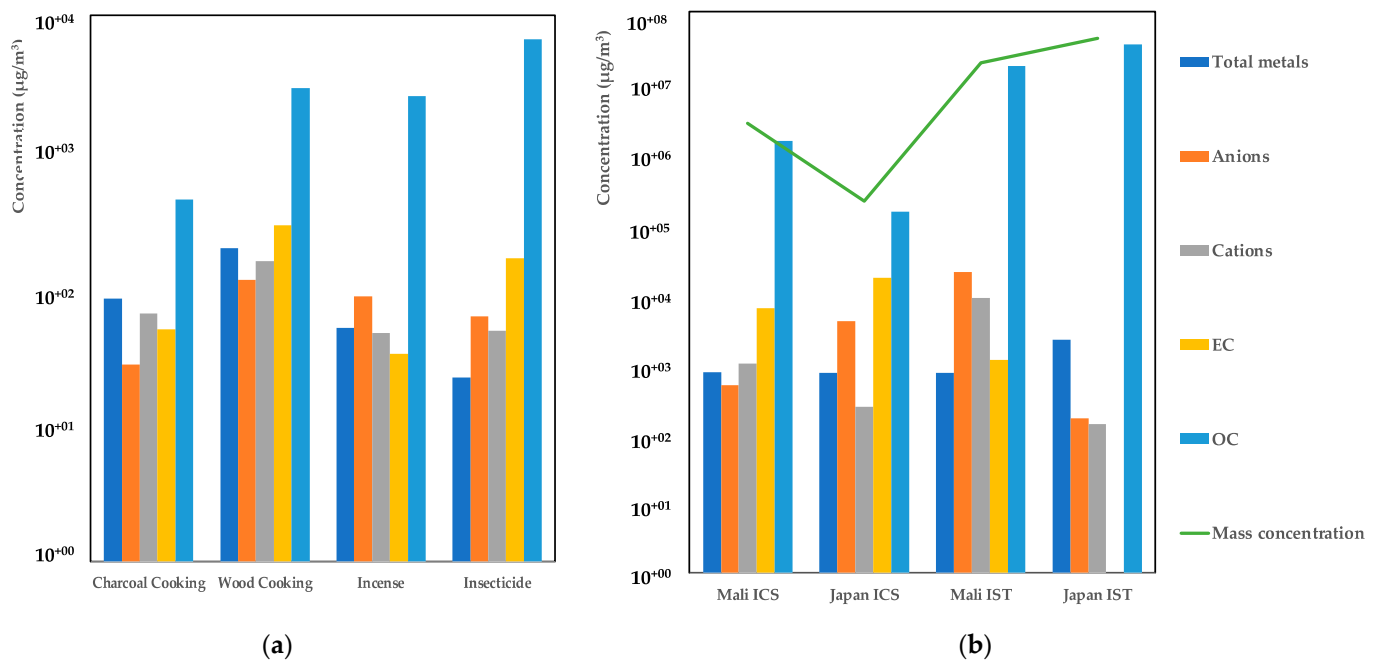


Figure 2. Average concentration of particulate matter constituents. (a) in situ samples (N = 18); (b) laboratory samples (N = 22). ICS: incense; IST: insecticide.

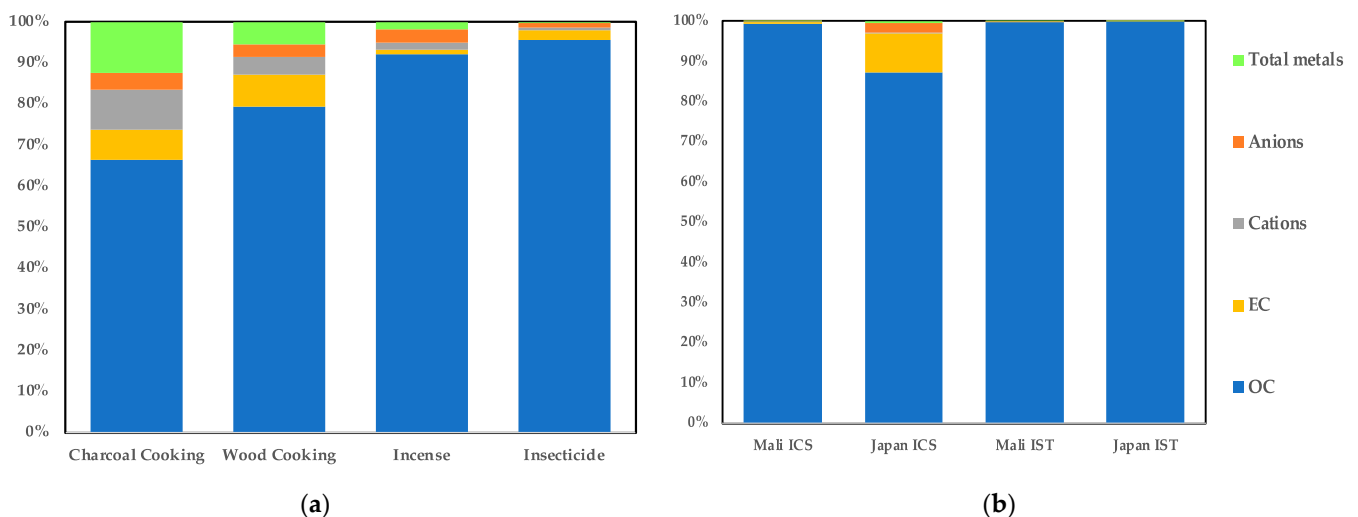


Figure 3. Contribution of particulate matter constituents in different samples. (a) in situ samples (N = 18); (b) laboratory samples (N = 22). ICS: incense; IST: insecticides.

3.1. Ionic Composition

In this study, the concentrations of seven anions (SO_4^{2-} , NO_3^- , Cl^- , F^- , PO_4^{3-} , NO_2^- , and Br^-) and five cations (NH_4^+ , Na^+ , K^+ , Mg^{2+} , and Ca^{2+}) were determined for different emission sources. The PO_4^{3-} concentration was below the detection limit for all samples;

thus, it was excluded from the data analysis. Figure 4 shows the average mole fraction of the ions in different PM samples obtained by in situ sampling in Bamako and the laboratory. The distribution differed for each emission source.

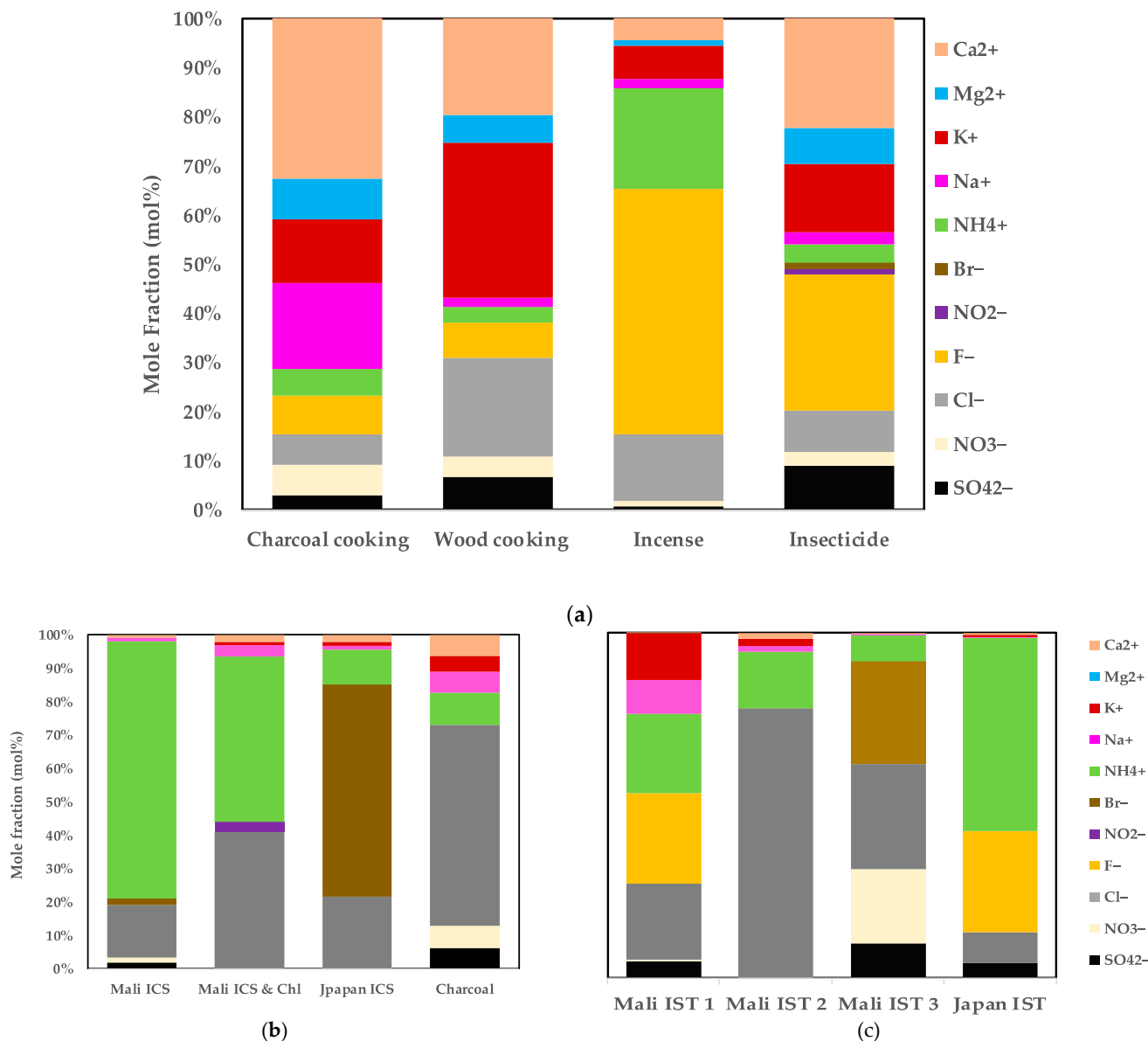


Figure 4. Average mole fraction of the ions in different particulate matter samples. (a) collected by in situ samplings; N = 18, (b) and (c) comparison of mole fraction of the ions in different PM samples collected in the laboratory; N = 22. Mali ICS: incense in Mali; Mali ICS & Chl: incense in Mali mixed with Charcoal; Japan ICS: incense in Japan; Mali ICS 1, 2 and 3: insecticide in Mali (different brands); Japan IST: insecticide in Japan.

Sampling at the cooking sites was performed using two different types of cooking fuels (charcoal and wood). The ion content for charcoal cooking was dominated by Ca²⁺ (33 mol%), Na⁺ (17 mol%), and K⁺ (13 mol%), and the order was Ca²⁺ > Na⁺ > K⁺ > Mg²⁺ > F⁻ > NO₃⁻ > Cl⁻ > NH₄⁺ > SO₄²⁻. The order of cooking using wood as cooking fuel was K⁺ > Cl⁻ > Ca²⁺ > F⁻ > SO₄²⁻ > Mg²⁺ > NO₃⁻ > NH₄⁺ > Na⁺. K⁺ accounted for 32 mol%, while Cl⁻ and Ca²⁺ accounted for 20 mol%. Cl⁻ showed a minor contribution to charcoal fuel compared with wood fuel. Na⁺ showed a high contribution to charcoal fuel but was minor for wood fuel. K⁺ and Ca²⁺ levels were significant in both samples. K⁺ is among the

main products detected during biomass combustion, especially wood combustion. Ca^{2+} and Na^+ have also been reported to occur during biomass burning [24]. The presence of Cl^- in the wood cooking sample may be associated with biomass burning and cooking ingredients. Cl^- has also been detected in vegetables cooked with a high chlorine content, such as garlic and onions [24–26].

The order of the ICS samples was $\text{F}^- > \text{NH}_4^+ > \text{Cl}^- > \text{K}^+ > \text{Ca}^{2+} > \text{Na}^+ > \text{NO}_3^- > \text{Mg}^{2+} > \text{SO}_4^{2-}$. F^- , NH_4^+ , and Cl^- represent 50, 21, and 13 mol%, respectively. The IST samples were predominant in anions and cations: F^- (28 mol%), Ca^{2+} (22 mol%), and K^+ (14 mol%). The order of the ions was $\text{F}^- > \text{Ca}^{2+} > \text{K}^+ > \text{SO}_4^{2-} > \text{Mg}^{2+} > \text{NH}_4^+ > \text{NO}_3^- > \text{Na}^+ > \text{Br}^- > \text{NO}_2^-$. F^- highly contributed to ICS and IST in situ and was identified as the main product of biomass burning [27]. The main ingredients of traditionally made ICS in Mali are wood residues or tree roots impregnated in perfumes. Mosquito coils (ISTs) are wood based [28]. This could explain the high contribution of F^- for ICS and IST, but there might be another reason, as the cooking activity using wood and charcoal showed a lower F^- contribution. Figure 4b represents the ionic composition of the traditional Malian ICS (ICS only and ICS mixed with charcoal) and the Japanese ICS stick. In households in Mali, as in other cities, ICS is combusted using charcoal; therefore, the combustion experiments included ICS samples without charcoal and ICS samples mixed with charcoal. Only charcoal burning was used as a reference. Figure 4c shows the three most popular brands of IST used in Mali (IST1 to IST3 for the most to least popular) and a typical Japanese IST obtained by laboratory sampling. From figure, the Mali IST1 and Japan IST seemed to have a similar ionic composition. They shared the particularity of emitting F^- ions, similar to the in-situ samples collected in Bamako (Figure 4c). In fact, IST1 was the most popular brand in Bamako City. The fact that F^- was not detected in the ICS combustion experiment (Figure 4b), but was highly loaded (28 mol%) in the in-situ ICS samples (Figure 4a), could be explained by the burning of ICS in the traditional clay burner in Bamako (Figure S2). This is because F^- is naturally abundant in clay. It has been detected in clay combustion experiments in previous studies [29,30]. Although F^- was detected in both in situ ICS and IST samples, it was not detected in ICS by laboratory sampling, in contrast to IST. This suggests that F^- might be a specific product of IST combustion.

Both Malian and Japanese ICS were significantly loaded with 16 mol% and 22 mol% Cl^- , respectively (Figure 4a). Cl^- was identified as among the main compounds in different ICS samples [31]. The dominant species in the Malian ICS was NH_4^+ (77 mol%) (Figure 4b). The contribution of NH_4^+ (Figure 4a) in the in-situ samples was significantly lower (21 mol%). After mixing the samples with charcoal, the contribution of NH_4^+ ions decreased to 50 mol% and that of Cl^- increased from 16 to 41 mol%. The simultaneous combustion of ICS and charcoal might have decreased the quantity of combusted ICS samples, resulting in a decrease in NH_4^+ compared to the combustion of only ICS. The addition of charcoal might have increased the contribution of Cl^- . In fact, charcoal is dominated by Cl^- . Japanese ICS showed a different ionic composition with a dominance of Br^- ions (63 mol%) reported from biomass burning [32,33]. This was followed by Cl^- (22 mol%) and NH_4^+ (11 mol%). The ionic contribution was different for different IST brands, but Cl^- and NH_4^+ were detected in all samples. Cl^- had a larger contribution to the Malian IST, while NH_4^+ had a larger contribution to the Japanese IST. This has been previously detected in biomass combustion [24,34,35]. They may have been produced from the ICS and IST biomass content. Br^- was detected only in Mali IST3 and contributed significantly (30 mol%).

Ca^{2+} is mostly associated with soil or crustal sources, and its contribution was significant for nearly all emission sources in Bamako, but had a very low contribution to the combustion experiment. This suggests that, in addition to Ca^{2+} from biomass combustion, the outdoor dust resuspension of the dry soil might have affected the air quality in the household microenvironments in Bamako.

3.2. Carbonaceous Composition

OC is either directly emitted from primary sources or produced by secondary atmospheric reactions of gaseous organic precursors. The EC is mainly formed from incomplete fuel combustion at high temperatures [36]. Numerous studies worldwide have identified activities such as biomass combustion (wood, leaves, and charcoal) and motor vehicle emissions as the main sources of OC and EC [36,37]. In this study, the combustion of IST was higher than that of charcoal or wood (Figure 5).

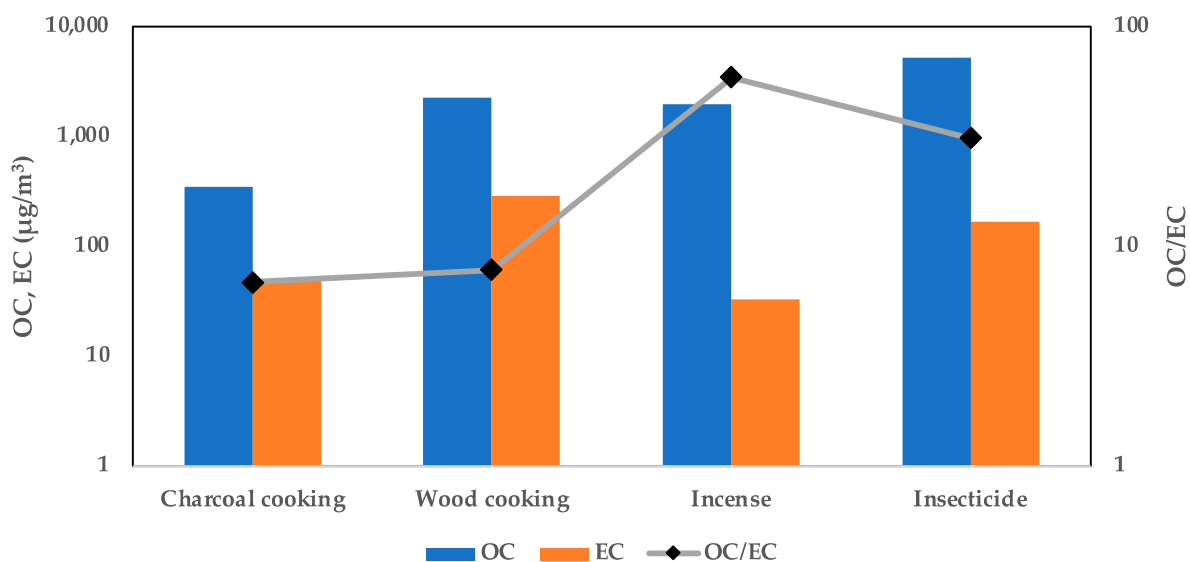


Figure 5. Average elemental carbon and organic carbon concentrations in different particulate matter samples collected by in situ samplings (charcoal cooking, wood cooking, IST: insecticide, and ICS: incense).

High carbonaceous concentrations were observed in household combustion sources. OC emitted from cooking activities was 2271.4 and 346.2 $\mu\text{Cg}/\text{m}^3$ for wood and charcoal cooking, respectively, and EC was 290.5 and 50.2 $\mu\text{Cg}/\text{m}^3$, respectively. The IST concentrations were 5172.5 $\mu\text{Cg}/\text{m}^3$ (OC) and 188.2 $\mu\text{Cg}/\text{m}^3$ (EC). In average, ICS concentrations were 1972.3 $\mu\text{Cg}/\text{m}^3$ (OC) and 33.43 $\mu\text{Cg}/\text{m}^3$ (EC). This high concentration might be attributed to the combustion conditions, including biomass content and aeration conditions. In fact, reported OC and EC values largely varied over previous studies for different biomass types, cooking styles, and burning conditions. Table 1 presents the OC and EC values obtained from this study and previous research. Alves et al. [38] reported much higher values for various cooking types compared with this study. OC was up to 62 mg/m^3 and EC was 0.35 mg/m^3 for cooking with charcoal grills. Wood ovens presented higher OC (178 mg/m^3) and EC (9.24 mg/m^3). Similarly, higher OC values of 3540 $\mu\text{g}/\text{m}^3$ were observed during cooking using wood fuel in a typical rural village in northern China. The various cooking fuels and styles in Bangladesh and Singapore presented different OC and EC values (Table 1). Popovicheva et al. [39] reported OC and EC values of up to 901 $\mu\text{g}/\text{m}^3$ and 73 $\mu\text{g}/\text{m}^3$, respectively, for wood cooking. Furthermore, high OC (up to 3802 $\mu\text{g}/\text{m}^3$) and EC (up to 203 $\mu\text{g}/\text{m}^3$) have been reported for different types of biomasses burning [39].

OC accounted for the largest fraction of carbonaceous substances in the households. The largest contributions occurred during the combustion of ICS and IST (96 and 75 wt%). The OC in PM collected from charcoal and wood cooking were 75 and 74 wt%, respectively. This suggests that although emission sources differed, they had the common characteristic of containing highly volatile carbonaceous compounds. Similarly, OC was the dominant carbonaceous material in the simulated combustion experiments. EC represented 10 wt% of Japanese ICS but was negligible in Malian ICS. The contribution was 1 wt% in the Mali ICS after the addition of charcoal. This suggests that charcoal might be among the ingredients

used in Japanese ICS, in contrast to Mali ICS. Charcoal is a common ingredient in various Japanese ICS sticks [31,43,44]. The EC was negligible in all IST products.

Table 1. Concentration of particulate matter constituents in this research (in bold) and previous researches. Numbers in [] are references. Emission sources are in italic letters. CHL: charcoal cooking; Wood: wood cooking; ICS: incense; IST: insecticide; B burning: biomass burning; amb: ambient air; OC: organic carbon; EC: elemental carbon. The values are in $\mu\text{g}/\text{m}^3$.

	Combustion Type	OC	EC
This research	CHL	346.6	50.2
	Wood	2271.4	290.5
	ICS	1972.3	33.43
	IST	5172.5	188.2
Amazon region (2011) [38]	CHL (grilling)	62,000	350
	Wood (oven)	178,000	9240
China (2012) [40]	Wood	3540	45.1
Bangladesh (2012) [41]	Wood and/or rice husk (unimproved oven)	210	176
	Wood and/or rice husk (improved oven)	182	152
Singapore (2008) [42]	Gas (steaming)	29.3	6.16
	Gas (boiling)	36.1	8.11
	Gas (stir-frying)	62.6	14.5
	Gas (pan-frying)	71.6	14.7
	Gas (deep frying)	121.5	15.8
North Vietnam (2016) [39]	Wood (indoor)	901	73
	Wood oven	825	203
	Wood (outdoor)	850	192
	B burning (cassava root)	3802	203
	B burning (corn and bushes)	166	28
	B burning (longan leaves)	1030	52

Biomass burning, including household fuel combustion, is a major source of carbonaceous compounds in PM. This emits a higher OC than EC. In the present study, the OC/EC ratio fluctuated significantly from one emission source to another (Figure 5). The highest OC/EC ratios for ICS (59) and IST (30) were observed in the indoor microenvironments of households. This may be due to the smoldering combustion of the products, which is characterized by a high OC/EC ratio [45–47]. In contrast, cooking activity with charcoal and wood fuel presented much lower ratios of 7 and 8, respectively. In this study, the average ratio from the cooking activity using wood fuel presented a higher value compared to those reported by He et al. [48] (1.24 to 5.26), Atiku et al. [49] (0.41 to 1.46), and Popovicheva et al. [39] (4.1 to 4.3). However, the ratio in this study was lower than those reported by Coffey et al. [50] (14 to 15), Keita et al. [51] (12.7), and Popovicheva et al. [39] (12.3). The difference in wood quality and species combined with the difference in combustion conditions could have resulted in the high variability in the ratio obtained in different studies. A lower value (2.7) for charcoal has been reported by Keita et al. [51].

Figure S4 shows the Char EC, Soot EC, and Char EC/Soot EC ratio. The Char EC, which is characterized by incomplete combustion, was obtained by subtracting the pyrolyzed organic carbon (POC) value from the EC1 value. Soot EC, characterized by high-temperature combustion, was calculated as the sum of EC2 and EC3. Char EC/Soot EC is a good indicator of biomass burning and incomplete combustion [52–54]. Previous studies have reported a high Char EC in samples collected near biomass combustion sites. The ratio of Char EC to Soot EC varied for different emission sources. Char EC/Soot EC was re-

ported to be 6.0 ± 4.6 and 4.8 ± 2.2 for biomass burning and 4.3 ± 1.9 and 1.3 ± 0.8 for coal combustion [53]. In this study, the highest Char EC/Soot EC ratio was observed for wood combustion (19.6), followed by charcoal (3.6), IST (1.6), and ICS (1.4). The cooking stoves used in Bamako were not adapted for efficient combustion. This results in an incomplete combustion [55]. Our results showed higher Char EC values for all samples. This suggests that the emission sources were characterized by incomplete combustion.

3.3. Metallic Elements Composition

The concentrations of 35 metallic elements in the PM collected by in situ sampling were measured. Hg was below the detection limit for all samples. Average concentrations ($\mu\text{g}/\text{m}^3$) are listed in Table S1. Except for a few elements (Ni, Ti, Bi, U, and Mo), PM emitted by cooking with wood samples presented the highest metallic concentrations.

Figure 6 shows the average weight fraction of the metallic elements in the different PM samples. In general, the same constituents of major elements were detected for the four emission sources with slightly different distributions. Al and Fe were the dominant constituents of all emission sources, with ICS and charcoal cooking presenting the highest contribution. Al and Fe are dust tracers but have been detected in previous studies during biomass combustion and cooking fumes [56]. Similarly, Zn was detected in all the emission sources. Other trace elements, such as Mn, Rb, and Ba, had negligible contributions and are represented by other metals, as shown in Figure 6a.

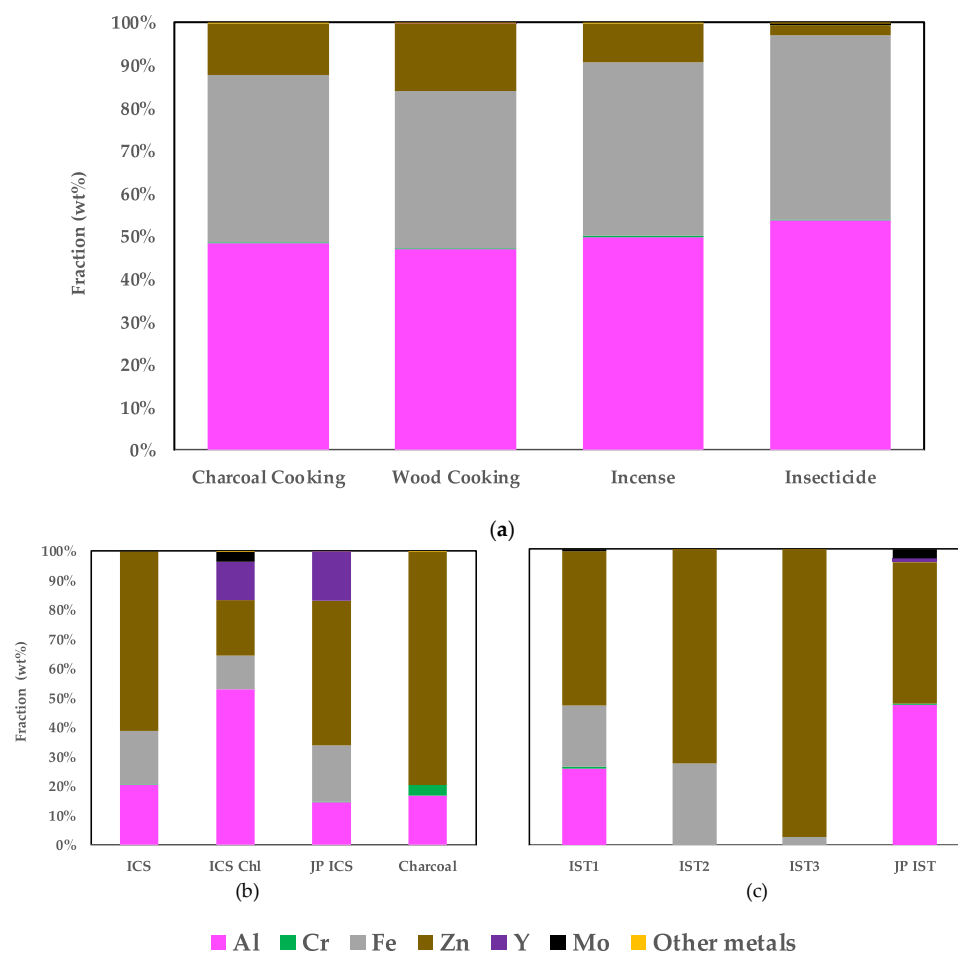


Figure 6. Average fraction of the major metallic elements in different particulate matter samples (a) collected by in situ samplings, (b) ICS collected by laboratory samplings, and (c) IST collected by laboratory samplings. Mali ICS: incense used in Mali; Mali ICS & Chl: incense used in Mali mixed with Charcoal; Japan ICS: incense used in Japan; Mali ICS 1, 2 and 3: insecticide used in Mali (different brands); Japan IST: insecticide used in Japan.

Zinc (Zn), which is a volatile metal [57], had a low contribution to the ICS and IST collected in Bamako, but was the predominant element in the ICS and IST samples collected during the former combustion experiment. The Malian IST, especially IST2 and IST3, was highly loaded with Zn (Figure 6b,c). The higher contribution of this metal in the combustion experiment samples compared to the in-situ samples might be due to the difference in the sampling conditions. The combustion temperature using charcoal as heat source in oxygen rich conditions was higher compared to the one using the ceramic heater with a restricted oxygen condition. Aerosol Zn was sampled upon liberation in the closed sampling system, whereas the gas-phase Zn evaporated into the room air, when at a higher combustion temperature using charcoal as a combustion source, resulting in a lower contribution in the aerosol samples collected in situ compared to the ones collected during the combustion experiment.

3.4. Health Exposure Assessment

PM emitted into the atmosphere can affect human health. They can enter organisms via diverse pathways. The main exposure occurs during respiratory processes. Inhaled PM can be deposited in the deepest body systems, including the respiratory, cardiovascular, and nervous systems, which can cause great damage. Hence, in this study, we estimated the health risks of PM exposure via inhalation. The HQ for a single heavy metal and the HI for multiple heavy metals were calculated to estimate the potential non-carcinogenic health impacts caused by exposure to a single heavy metal and a mixture of heavy metals, respectively. The CR was calculated to estimate the carcinogenic risks.

3.4.1. Non-Carcinogenic Risks

The HQ and the HI were estimated for different typical daily household activities based on in situ sampling (Table 2). As mentioned earlier, the HQ and/or the HI should not exceed the recommended value of 1 for health safety purposes. The HQ for Mn exceeded the exposure limit for cooking activities using wood in both adults and children. Cooking (with charcoal and wood) and the use of ICS presented unsafe values for Ni in children. The HI exceeded the safe exposure limit in adults and children for all emission sources except for the use of IST. Cooking activity represented the greatest risk for both adults and children, followed by ICS and IST. The HI for charcoal cooking was more than 6- and 1.5-fold higher than the recommended value in children and adults, respectively. Similarly, the HI for charcoal cooking was more than 5- and 2-fold higher than the limit for adults and children, respectively. The ICS presented an HI more than 2-fold higher than the limit in children and exceeded the limit in adults. The IST presented values below the recommended limit, in contrast to ICS. This could be attributed to the fact that IST are manufactured in industries where health safety regulations are adapted. In contrast, the ICS used in Mali is homemade; hence, no regulations are applied. In this study, non-carcinogenic risks were higher in children than in adults.

Table 2. Non-carcinogenic risks of toxic metals in different particulate matter samples collected by in situ samplings for adults and children. IST: insecticides; ICS: incense.

		Charcoal Cooking		Wood Cooking		IST		ICS	
		Adults	Children	Adults	Children	Adults	Children	Adults	Children
HQ	Cr	0.07	0.07	0.11	0.11	0.02	0.02	0.10	0.10
	Mn	0.99	0.99	1.68	1.68	0.13	0.13	0.70	0.70
	Ni	0.48	4.91	0.31	3.16	0.02	0.25	0.18	1.82
	As	0.04	0.04	0.06	0.06	0.03	0.03	0.03	0.03
	Cd	0.02	0.02	0.04	0.04	0.06	0.06	0.02	0.02
	Be	0.02	0.02	0.10	0.03	0.08	0.02	0.08	0.01
	HI	1.61	6.04	2.31	5.09	0.35	0.51	1.11	2.69

3.4.2. Carcinogenic Risks

The highest carcinogenic risk was attributed to the exposure to PM emitted from wood cooking, followed by those from the use of ICS, charcoal cooking, and the use of IST. The Total-CR was above the lower limit of 10^{-6} but below the unacceptable level of 10^{-4} in adults. In children, Total-CR exceeded the unacceptable limit for wood cooking activity and the use of ICS. The values of 1.45×10^{-4} and 1.21×10^{-4} were obtained for cooking using wood fuel and the combustion of ICS, respectively. Other activities presented values higher than the safe exposure limit, but lower than the unacceptable limit in children (Table 3). Children are considered sensitive. Owing to their young age, they are more vulnerable. Nevertheless, carcinogenic risk was higher in children than in adults.

Table 3. Carcinogenic risks of toxic metals in different particulate matter samples collected by in situ samplings for adults and children. IST: insecticides; ICS: incense.

	Charcoal Cooking		Wood Cooking		IST		ICS	
	Adults	Children	Adults	Children	Adults	Children	Adults	Children
Cr	2.76×10^{-5}	8.04×10^{-5}	4.70×10^{-5}	1.37×10^{-4}	9.28×10^{-6}	2.71×10^{-5}	4.04×10^{-5}	1.18×10^{-4}
Ni	5.87×10^{-7}	1.71×10^{-6}	3.78×10^{-7}	1.10×10^{-6}	2.95×10^{-8}	8.60×10^{-8}	2.18×10^{-7}	6.35×10^{-7}
As	8.46×10^{-7}	2.47×10^{-6}	1.25×10^{-6}	3.64×10^{-6}	6.94×10^{-7}	2.02×10^{-6}	6.69×10^{-7}	1.95×10^{-6}
Cd	1.06×10^{-7}	3.09×10^{-7}	2.73×10^{-7}	7.96×10^{-7}	3.78×10^{-7}	1.10×10^{-6}	1.34×10^{-7}	3.89×10^{-7}
Pb	4.34×10^{-8}	1.27×10^{-7}	1.35×10^{-7}	3.95×10^{-7}	2.94×10^{-8}	8.56×10^{-8}	2.49×10^{-8}	7.26×10^{-8}
Be	6.08×10^{-6}	9.97×10^{-7}	6.85×10^{-6}	1.52×10^{-6}	3.86×10^{-6}	1.13×10^{-6}	3.87×10^{-6}	1.10×10^{-7}
Total-CR	3.52×10^{-5}	8.60×10^{-5}	5.59×10^{-5}	1.45×10^{-4}	1.43×10^{-5}	3.15×10^{-5}	4.53×10^{-5}	1.21×10^{-4}

Although the toxic metal concentrations were lower than the non-toxic ones in this study (Table S1), the risk assessment indicated health hazards. This implies that accumulated and long-term exposure can trigger health.

In addition to the household PM exposure risk assessment, the risks from exposure to ambient PM sampled at a typical roadside in Bamako city were determined. Similarly, the HI and Total-CR were higher in children than in adults (Table 4). The HI was within the recommended limit in adults (0.55) but exceeded the limit in children (1.35). The CR was higher than the acceptable limit for adults (2.63×10^{-5}) and children (7.19×10^{-5}). These risks were lower than those from typical household activities. Except for the use of IST, all household emission sources presented a greater non-carcinogenic and carcinogenic risk compared to the roadside PM sample. We spend most of our time in indoor microenvironments. Indoor microenvironments are generally perceived as safer than outdoor microenvironments. However, indoor pollution is sometimes higher than what is believed. Household activities, including cooking and the combustion of ICS and IST, emit pollutants that have harmful effects on health. This is enhanced in developing countries owing to the type of cooking fuel and aeration conditions. This study highlighted the elevated risks associated with indoor exposure. Hence, more attention should be paid to PM exposure in indoor microenvironments, particularly during typical daily activities.

Additionally, we compared our health risk assessment results with those of previous studies in China, Ethiopia, and India (Table 4). The HI in this study during the cooking activities was higher compared to the value of 2.55 in children reported by Yang et al. [58] (2014) in Chinese urban household environments. This indicates higher non-carcinogenic risks for children in the urban household environment in Bamako than in Beijing. This could be attributed to the difference in cooking fuels (natural gas was used in China, and wood and charcoal were used in Mali). The Total-CR was lower (3.35×10^{-4} in adults and 8.38×10^{-5} in children) [38]. This could be attributed to smoking activity, which is known to emit carcinogenic metals, including As, Cd, Cr, Ni, and Pb [59–61]. The risk assessment results from the exposure to cooking emissions conducted in Ethiopia by Embiale et al. [62] and the one conducted in Indian rural households by Chowdhury et al. (2022) [63] were assessed using the exposure concentration of metallic elements contained in PM₁₀ and PM_{2.5}, respectively. As in this study, we conducted a risk assessment using TSP and multiplied the results presented by Emiale et al. [62] and Chowdhury et al. [63] by a

factor of 1.2 and 1.3, respectively, assuming that PM₁₀ and PM_{2.5} represent approximately 80 and 70% of the TSP, respectively (Table 3). Cooking activities in Ethiopia and using charcoal, kerosene, and electricity presented lower values than ours. This indicates the risks in the microenvironments of households in this study compared to those in Ethiopia during cooking of traditional food. The values presented for cooking activities using wood and leaves in rural India presented much higher risks compared to the microenvironments of households in the urban city of Bamako (Table 3). These differences could be due to differences in the sampling conditions, including the distance between the emission source and the sampling instrument, and the biomass type. More studies are needed to provide information on the health risks of exposure to PM emitted from typical indoor activities, including cooking using different fuel types and other indoor combustion sources (such as ICS and IST).

Table 4. Non-carcinogenic and carcinogenic risks of toxic metals in adults and children from this research (in bold) and previous researches of toxic metals in different particulate matter samples collected by in situ samplings for adults and children. Charcoal: charcoal cooking, Wood: wood cooking, IST: insecticides, ICS: incense, Rds: roadside, and N-gas: natural gas.

	This Study		China (2014) [58]		Ethiopia (2020) [62]		India (2022) [63]	
	Adults	Children	Adults	Children	Adults	Children	Adults	Children
HI	Charcoal	1.61	6.04		0.04	0.06		
	Wood	2.31	5.09					
	ICS	1.11	2.69					
	IST	0.35	0.51					
	Rds	0.55	1.35					
	Biomass							37.79
	N-gas/smoking			2.55	2.55			
	Kerosene					0.17	0.29	
	Electricity					0.02	0.04	
	Total-CR	Charcoal	3.52 × 10⁻⁵	8.60 × 10⁻⁵		2 × 10 ⁻⁷	5 × 10 ⁻⁷	
Wood		5.59 × 10⁻⁵	1.45 × 10⁻⁴					
ICS		4.53 × 10⁻⁵	1.21 × 10⁻⁴					
IST		1.43 × 10⁻⁵	3.15 × 10⁻⁵					
Rds		2.63 × 10⁻⁵	7.51 × 10⁻⁵					
Biomass								7 × 10 ⁻³
N-gas/smoking				3.35 × 10 ⁻⁴	8.38 × 10 ⁻⁵			
Kerosene						8 × 10 ⁻⁷	2 × 10 ⁻⁷	
Electricity						2 × 10 ⁻⁷	3 × 10 ⁻⁸	

4. Conclusions

In this study, the composition of PM from specific household emission sources was characterized. The use of ICS and IST was previously identified as the most personal exposure source in households [6]. PM samples were collected on filters at specific household emission sources in Bamako City and simulated under conditions of IST and ICS burning in a laboratory.

The obtained results indicate that the mole fraction of the PM component varies significantly from one emission source to another. The characteristics observed in the different microenvironments mark the exposure. The ion distribution is varied. The cooking samples is dominated by cations, ICS is highly loaded with anions, and IST is equally loaded with cations and anions. OC is the dominant carbonaceous material. The Char EC/Soot EC ratio indicates that the emission sources are mainly characterized by incomplete combustion. Dust and biomass tracers are the most prominent metallic elements. A comparison between the Japanese and Malian products indicate that the mass

percentages of the elements are different. The dominant ions in Malian ICS are NH_4^+ and Br^- for the Japanese ICS. Furthermore, except for the Japanese ICS, the EC has a negligible contribution to the ICS and IST carbonaceous components. Zn and Al are the dominant metals in the ICS and IST samples, respectively. Our experiment showed that different IST brands had different PM component distributions. In addition, the burning conditions might have influenced the liberated elements from the ICS and IST. A health risk assessment study indicated that exposure to PM is unsafe. The HI varied between 0.35 and 6.04. Only the IST presented a value below the HI limit of one in adults and children. However, the values exceeded the safety limit for cooking and ICS use. The CR exceeded the lower limit of exposure of 10^{-6} for all activities and microenvironments and exceeded the unacceptable level of 10^{-4} in children for cooking using wood fuel and the combustion of ICS. Overall, cooking activities had the highest health risks, followed by ICS and IST. Non-carcinogenic and carcinogenic risks were higher in children than in adults.

The findings of this study indicate that it is crucial to control household emission sources. Both the local government and the inhabitants should participate in this effort. Exposure to cooking is an important issue. Gradually switching from inefficient cooking stoves to improved cooking stoves can considerably reduce exposure. ICS is an important part of the culture, and the use of IST is a common practice in Bamako to prevent malaria. Both the ICS and ITS are combusted indoors, where air exchange is very limited. Ensuring that room aeration is adequate during the use of ICS and IST, and reducing the frequency of use can be a good way to reduce exposure. Moreover, previous studies have reported that mosquito coils may be inefficient in preventing malaria and may have harmful effects. According to Hogarth et al. [10], mosquito coils only have 24 to 64% efficiency [64–66]. Given this and the possible health risks, the use of mosquito nets would be the best way to protect health.

To our knowledge, this is the first study to characterize the chemical composition and related health effects of PM emitted from diverse household activities, including cooking and the combustion of ICS and IST in Western Sub-Saharan Africa. The findings provide scientific data that are essential as a preliminary step in promulgating air pollution mitigation strategies and reducing household PM exposure to protect health. Further studies on particulate matter emitted from household activities are needed to better understand the chemical composition and the health effects.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/atmos13081290/s1>. Figure S1. In situ sampling system. (a) Schematic of the NILU filter holder system used in Bamako. (b) Photograph of the sampling system. Figure S2. In situ sampling of images. (a) Insecticide and incense combustion sampling and (b) cooking period sampling. Figure S3. The filter samples were collected in plastic boxes and zip logs. Figure S4. Different IST and ICS products: (a) most popular IST in Bamako; (b) traditional ICS in Bamako. Figure S5. Average Char elemental carbon and Soot elemental carbon concentrations and Char elemental carbon/Soot elemental carbon ratios in different particulate matter samples collected by in situ sampling (charcoal cooking, wood cooking, IST: insecticide, and ICS: incense). Table S1. Average concentration of the particulate matter components for different emission sources (values are in $\mu\text{g}/\text{m}^3$). ICS: incense; IST: insecticides; CHL: charcoal cooking; wood: wood cooking. nd was used for samples with lower values than blank samples.

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