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Influence of Residential Combustion on Total Gaseous Mercury (TGM) Levels: a Preliminary Study at an Urban Megacity in Vietnam

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ABSTRACT

Mercury (Hg) is a global pollutant that has gained monumental attention in recent decades due to its neurotoxicity and great bioaccumulation features. The atmosphere plays a key role in the movement of Hg in the environment, and residential combustion is a significant contributor to atmospheric Hg. However, there is a severe lack of research on the impact of Hg emissions from residential combustion in Southeast Asia (SEA). This study focused to illustrate the contribution of residential combustion on the level of total gaseous mercury (TGM) in a tropical megacity in southern Vietnam by conducting a short field campaign in February 2023. The mean TGM concentration during the study period was 2.27 ± 0.81 ng m^{-3}, in which daytime TGM level (7 am – 7 pm, 2.56 ± 0.80 ng m^{-3}) was significantly higher than at night (7 pm – 7 am, 1.58 ± 0.28 ng m^{-3}), which could be attributed to local burning activities. The TGM concentration was also increased because of the increase in burning activities (i.e. burning joss paper and incense) during the Lantern Festival. Backward trajectory analysis suggested the domination of regional background air mass in the study area, implying that TGM concentration variation is mostly associated with local influences. We estimated that residential combustion caused a rise of around 70% in TGM concentration at the sampling site. Overall, this is the first study to illustrate the importance of Hg emissions from residential combustion in urban SEA region, laying a background for upcoming studies in atmospheric Hg in this region.

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1. Introduction

Mercury (Hg) is a toxic trace metal that is recognized as a widespread global pollutant [1]-[4]. When released into the atmosphere, Hg can be transported over long distances and eventually deposit in ecosystems where it can have adverse effects on humans and wildlife [5]-[7]. Mercury exists in three forms in the atmosphere: elemental gaseous mercury (GEM), gaseous oxidized mercury (GOM), and particulate-bound mercury (PBM) [5]-[8]). The sum of GEM and GOM is called total gaseous mercury (TGM). The majority of Hg in the atmosphere is GEM, which has an average lifespan of about 1 year [5], [9]. The negative impacts of Hg on the environment and human health are well-documented, particularly through the consumption of methylmercury in fish [6], [10]. In response to the global concern over Hg pollution, 128 countries and territories, including Vietnam, signed the Minamata Convention on Mercury in 2017, intending to protect human health and the environment from Hg emissions and releases.

Mercury can enter the atmosphere from both natural and anthropogenic sources. Natural sources include things like burning biomass, volcanic eruptions, and emissions from soil and vegetation [2], [6], [11], [12]. Meanwhile, important anthropogenic Hg emission sources have been small-scale gold mining, coal combustion, residential burning, and producing of cement and non-ferrous metals which account for about 30% of total atmospheric Hg emissions [6], [13]. Residential burning, including the burning of fossil fuels and biomass (e.g., coal, wood, agricultural waste), is a common source of atmospheric Hg both indoor and outdoor [6], [14]. In addition, the burning of incense in temples can have a significant impact on the air quality and could contribute to the levels of Hg in the atmosphere [15], [16]. Incense sticks are commonly made from a mixture of herbs, resins, and other materials that may contain Hg [15]-[17]. Mercury is released into the atmosphere during burning processes, leading to a buildup of Hg concentrations in the surrounding environment [2], [7]. The amount of Hg released through residential burning is found to be directly proportional to the amount of fuel burned and type of fuel burned (e.g., burning of coal results in higher Hg emissions compared to burning of wood or other biomass fuels [18]. Moreover, the emission rates from residential burning can be much higher than from other sources such as coal-fired power plants and globally, 2.8 billion people still rely on solid fuel for cooking and heating [18]-[20].

Studies have been conducted on the impacts of residential burning on atmospheric Hg worldwide [2], [21]. GEM, GOM and PBM can be emitted from residential burning, however, GEM is considered to be the main form emitted from this source [18], [19], [22]. For instance, a study found the release of atmospheric Hg from residential coal burning and found that GEM was the dominant form of Hg in the coal exhaust gas, accounting for more than 90%, while GOM and PBM made up less than 10%, respectively [19]. Residential burning is a significant contributor to the global Hg budget, with estimates suggesting that it may account for as much as 50% of the total anthropogenic atmospheric Hg emissions in some regions [2], [23]. The absence of studies on the levels of Hg in the air from residential burning in Southeast Asia (SEA), a region that encompasses many developing countries where such activities as cooking and incense burning for religious purposes are widely practiced, is a matter of concern.

Vietnam is a prosperous economic center in Southeast Asia (SEA) with a rapidly growing population of around 100 million, especially in large cities like Ho Chi Minh City (HCMC). The rate of urbanization in Vietnam has dramatically increased over the past two decades, leading to a rise in air pollution, which poses a serious threat to health [12], [24]. The occurrence of residential burning is widespread in the central urban district of HCMC, which is suggestive of its role in elevating the Hg levels in the air. Due to the limited knowledge of atmospheric Hg in HCMC, this study was conducted with the major aim to characterize the levels of TGM in a residential area where burning (i.e. outdoor cooking, incense burning, joss paper burning) occurs frequently. To evaluate the effect of burning on various locations, both indoor and outdoor TGM levels were measured at the same time. This is the first study in Vietnam and the SEA to examine the impact of residential burning on atmospheric Hg levels both indoor and outdoor, laying the groundwork for future studies and could help to enhance the understanding of atmospheric Hg in the SEA region.

2. Materials and methods

2.1. Site description

Atmospheric Hg (i.e. TGM) sampling was conducted on the 1st floor of an apartment building in the Cho Lon area (CL, 10 45'10.66"N 106 39'29.81"E, Figure 1), a hub for commerce and trade in HCMC. In this area, cooking and temple activities are a central part of daily life and both are deeply rooted in Vietnamese culture. The sampling location is located in an area affected by multiple sources of residential combustion, including the use of coal for grilling and cooking in several restaurants located along with the busy street and the burning of incense in a temple located 20 meters away (Figure 1). In addition, the sampling site is approximately 1 km away from the Kim Bien and Binh Tay markets, both of which have a long history and are bustling with trade activity, especially the Kim Bien market, which is the biggest chemical trading center in HCMC. The sampling campaign was carried out for 4 days (February 4-7, 2023), falling into the Lantern Festival period (i.e. February 5). As a result, there were numerous religious ceremonies taking place at the nearby temple. In addition, Cho Lon is a region inhabited by a significant population of Chinese descent, thus the festivities during the Lantern Festival period are highly bustling. This study designed the simultaneous sampling of atmospheric Hg from three different locations (3 sampling systems were used) - one indoor (ID) and two outdoor (OD1 and OD2) - based on the characteristics of the sampling site (i.e. CL). OD1 and OD2 are situated roughly 20 meters away from each other, with OD1 positioned in front of the apartment's main door and OD2 situated in the open space of the apartment staircase.

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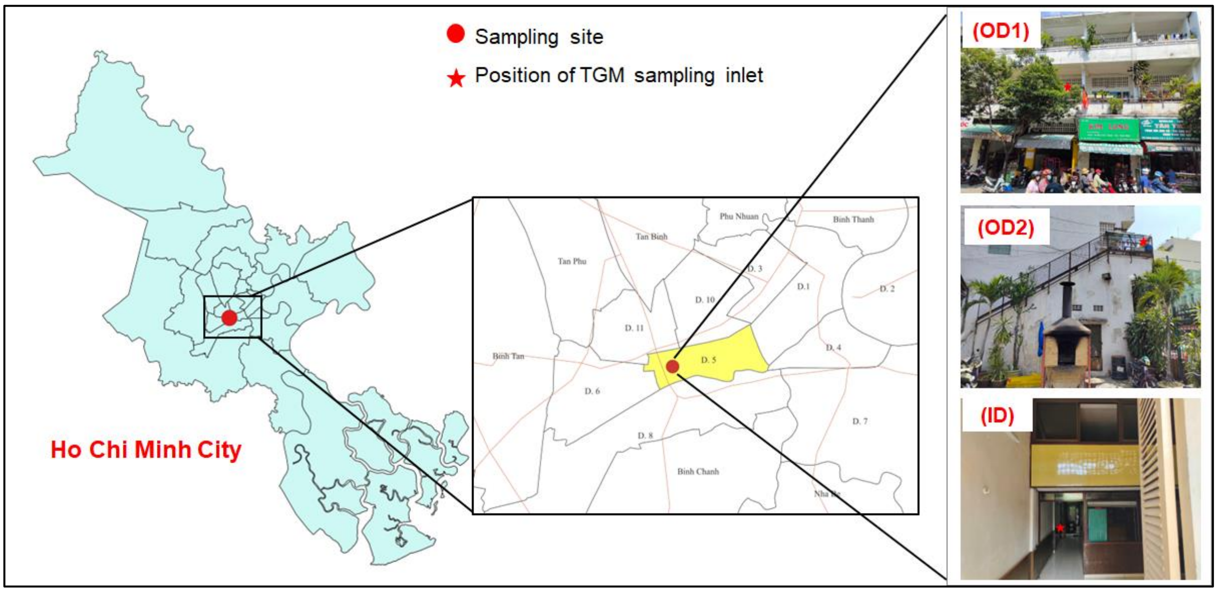

Figure 1. *Geographic location and overview of the sampling site (HCMC)*

2.2. TGM sampling and analysis

This study utilized a manual sampling technique following method IO-5 (US-EPA) to determine the atmospheric Hg concentration at CL. Specifically, air from the surroundings was drawn through a gold trap (NIC, Japan) at a flow rate of 0.5 L min^{-1}. The sampling flow rate was controlled by the mass flow controllers (MFC, KOFLOC, Japan). To eliminate interfering substances, a soda-lime trap (Sigma-Aldrich) was positioned upstream of the gold trap. This soda-lime trap was able to remove PBM and a portion of GOM, therefore, the term TGM (GEM + GOM) was used to describe the atmospheric Hg data in this study [25], [26].

For OD1, the TGM sample was collected during three distinct periods in a day (i.e. 3 samples/day), covering the hours of 7 am - 1 pm, 1 pm - 7 pm, and 7 pm - 7 am of the following day. For both OD2 and ID, two TGM samples were taken daily, simultaneously with the sample collected at OD1 (i.e. from 7 am - 1 pm and 1 pm - 7 pm. Moreover, burning activities (i.e. burning vs no burning) were also recorded and we noticed that most of the burning activities are stopped at nighttime. This sampling schedule allowed us to study the influence of residential combustion on the levels of atmospheric Hg by contrasting samples gathered during “no burning" and "burning" periods. Additionally, comparing TGM levels found between OD1 and ID could provide insight into the relationship between indoor and outdoor atmospheric Hg pollution. Finally, we attempted to illustrate the influence of the festival's activities (i.e. Lantern Festival) on the TGM level in the study area.

A comprehensive examination of the method and QA/QC protocol to determine TGM concentrations in HCMC has been provided in our previous study [27]. In brief, the gold traps were cleaned by heating them at 600^0C to eliminate any remaining Hg prior to sampling. To prevent contamination during the sampling process, travel blanks were conducted. The results indicated that the amount of Hg in the blank samples was always lower than MDL, demonstrating that contamination was negligible. The samples and blanks were analyzed at the University of Science, VNUHCM. The Hg was quantified using a WA-5F instrument and TC-WA automatic sample changer from NIC, Japan. A calibration curve was created before sample analysis using the MB-1 Hg standard vaporizer from NIC, Japan, with injection volumes from 0.02 to 2.5 ng Hg vapour. The standard curve must reach $R^2>0.9999$ and have a mean deviation of the response factor (RF) of less than 3% before sample analysis could proceed. Moreover, this study also compared the TGM concentrations obtained from 3 sampling systems when collected under the same condition to evaluate the accuracy of the measurement. The results showed that there was no significant difference in TGM concentrations taken by the 3 sampling systems (ANOVA test, $p>0.2$, triplicate samples n = 5), showing the reliability of the sampling method.

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3. Results and discussion

3.1. General characteristics of TGM data

Throughout the sampling period, a total of 24 TGM samples were collected, with 18 taken outdoor and 6 taken indoor. The mean (±S.D.) of TGM concentrations for outdoor and indoor were 2.27 ± 0.81 ng m^{-3} and 1.95 ± 0.15 ng m^{-3}, respectively. The TGM concentrations ranged from 1.18 to 3.84 ng m^{-3} for outdoor samples and from 1.79 to 3.14 ng m^{-3} for indoor samples. The S.D. value of outdoor TGM concentrations was about 5 times higher than that of indoor, implying the strong influence of potential Hg emission sources in the outdoor environment. The outdoor TGM concentration at CL was significantly higher than the background TGM level in the northern hemisphere (1.4-1.6 ng m^{-3}; Sprovieri et al., 2016). In addition, the outdoor TGM value in this study was comparable to or higher than values reported in Chuncheon, Korea (2.12 ± 1.47 ng m^{-3} [28]), Jhongli, Taiwan (2.61 ± 6.47 ng m^{-3}, [29]), and Fukuoka, Japan (2.33 ± 0.49 ng m^{-3}, [30]). On the other hand, the TGM level at CL was significantly lower than those reported in Chinese cities such as Shanghai (4.19 ± 9.13 ng m^{-3}, [31]), Nanjing (7.9 ± 7.0 ng m^{-3}, [32]), and Guiyang (9.72 ± 10.2 ng m^{-3}, [31]). A recent Hg emission inventory revealed that China is the largest anthropogenic source of Hg emissions in the world (e.g., coal combustion [33]) accounting for approximately 30% of the total anthropogenic Hg emissions. Consequently, atmospheric Hg concentrations observed at sites in China tend to have high atmospheric Hg concentrations. These comparisons highlighted the important roles of anthropogenic emissions on atmospheric Hg levels, in which combustion activities are an important source that needs attention.

Figure 2 shows the daytime (7 am – 7 pm) TGM concentrations observed at 3 sampling sites CL during the sampling campaign. On days 4 and 5 of the campaign, the TGM concentration in the outdoor samples (OD1 and OD2) was greater by more than 50% compared to the levels on the other days. This can be explained by the fact that the Lantern Festival is celebrated heavily on February 4-5 (i.e., major days of the festival) with a variety of festival events taking place, including the burning of votive papers at nearby temples. Moreover, increasing the act of burning votive papers and coal for cooking from neighboring households during this period also played a role in increasing the TGM level. A sharp decrease in TGM concentrations was found after the major event days (Figure 2) suggesting the strong impacts of local residential burning on atmospheric Hg in the study area. Compared to outdoor, the indoor TGM concentration showed less variation, with the TGM concentration on February 5 only slightly higher than the rest of the days (Figure 2). Detail regarding the comparison between indoor and outdoor TGM concentrations will be presented in the following section.

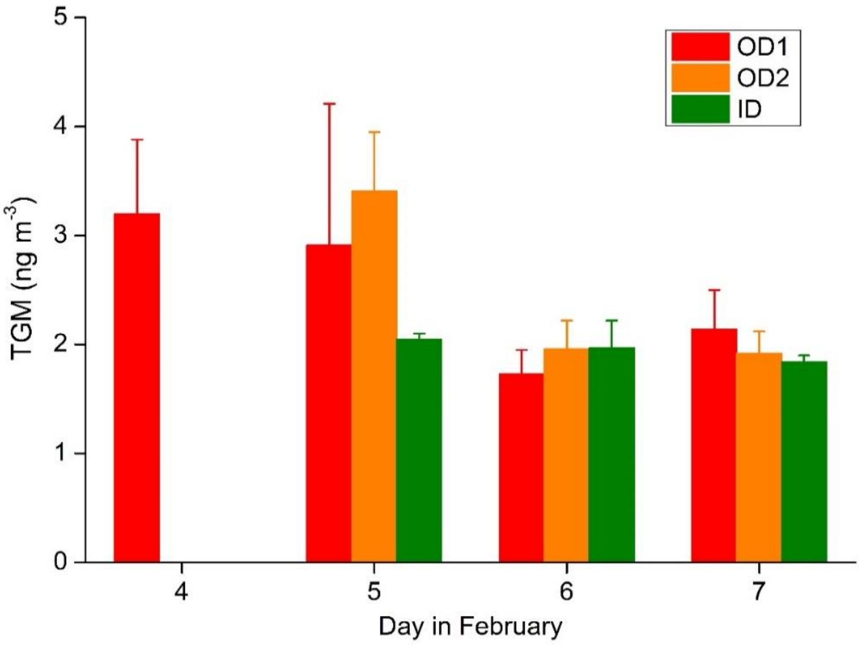

Figure 2. *Daytime (7 am – 7 pm) atmospheric Hg concentrations at CL during February 4 – 7. OD1 and OD2 are the outdoor samples and ID is the indoor samples. Error bar represent +S.D*

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3.2. Outdoor vs indoor TGM concentrations.

In this section, we focus to compare TGM levels obtained from OD1 and ID locations. The OD1 is situated in front of the apartment's main door, which allows for the characterization of outdoor TGM concentrations and is a good site to make comparision with ID. Six pairs of ID vs OD1 TGM samples were collected simultaneously during February 5-7 (for ID, 2 samples/day from 7 am – 7 pm, and for OD1 the data collected in the same period was used) and the concentration variations are shown in Figure 3. In general, ID and OD1 TGM concentrations were relatively similar except for sample number 2 where OD1 TGM concentrations were more than 90% higher than that of ID. The similarity in TGM concentrations between ID and OD1 for most of the pair samples could be explained by good air circulation from outdoor to indoor environment because the main door and window are open. On the other hand, pair sample No. 2 was collected during the major period of Lantern Festival activities (i.e. 12 am – 7 pm February 5), which resulted in a notable rise in outdoor TGM levels. However, this did not have a significant impact on ID TGM concentration, as indicated by the slight increase in TGM concentration (Figure 3). This can be explained by the fact that during the sampling time of No. 2, the main door and window of the apartment are closed for most of the sampling time and thus restricting the air circulation leading to limiting the influence of OD1 TGM on the ID environment. It should be noted that no people was living in the apartment building where the samples were taken, and thus the potential indoor Hg source was excluded. A better agreement between ID and OD1 TGM concentrations was observed when we kept the window and main door open (pair samples 4 and 6, Figure 3) while a greater discrepancy was found when keeping them closed (pair samples 3 and 5, Figure 3). In general, based on our results, it could be concluded that with no potential ID Hg source, the outdoor TGM plays an important role in determining the ID TGM concentration. In addition, based on our data, we speculated that the capacity for spreading air or ventilating, known as diffusion mode, is a crucial factor in determining the level of atmospheric Hg concentration within the household.

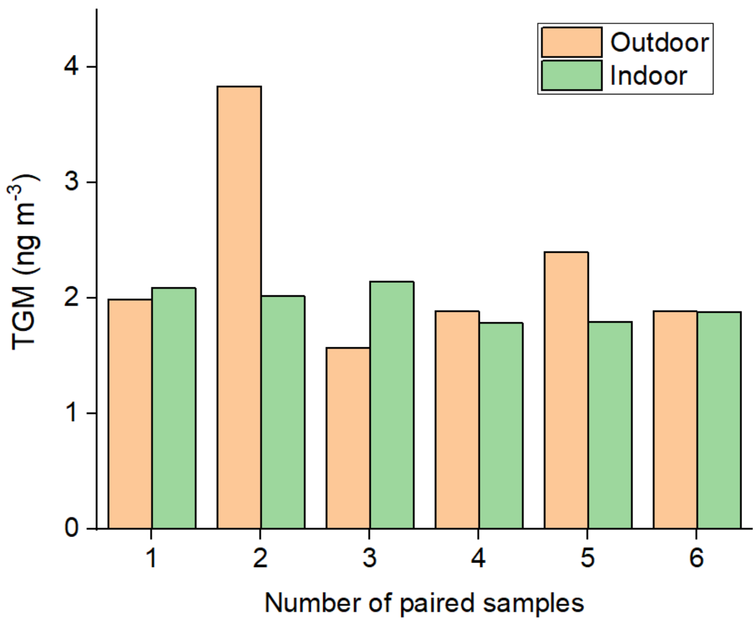

Figure 3. *Outdoor vs Indoor TGM concentrations during the sampling period*

3.3. Daytime vs nighttime TGM concentrations and implication for the influence of residential burning.

Both local and regional (i.e. long-range transport) sources of air pollutants could influence the air quality, and TGM concentrations as a result, HCMC has been well-documented by previous studies [12], [16], [34]. With the major aim of assessing the contribution of residential burning (local source) on the TGM concentration, we initially investigated the role of air mass origins (regional source) affecting the study area. The HYSPLIT backward trajectories (BWTs) model was employed to simulate

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backward trajectories of air masses. This model is frequently used in air pollution research to gain insight into the origins and paths of air masses, as demonstrated in previous studies[9,12,35]. By utilizing meteorological data from the National Centers for Environmental Prediction Global Data Assimilation System (GDAS 1°×1°), the 72-hour backward trajectories were calculated for each TGM sample (i.e. 3 BWTs/day at the starting collection time of each TGM sample, i.e. 7 am, 12 am, 7 pm local time) starting from the sampling site (CL) at an altitude of 200 m a.g.l. The choosing starting time allowed us to illustrate the air masses influencing the TGM samples as well as track changes in the air mass of the following TGM sample [3], [12], [25]. Figure 4 shows the BWTs arriving at CL during the study period, in which the air masses affecting CL mainly come from the West and Northwest of HCMC, passing vast areas of southern SEA including Laos, Cambodia, and Thailand. More specifically, the BWTs can divide into two sub-groups, in which group 1 is associated with BWTs mainly from continental SEA (from 4 – early 6) and group 2 (6-7) is associated with BWTs passing the Gulf of Thailand before reaching HCMC (Figure 4). Recent global anthropogenic Hg emission inventory [2] indicated that there are no intensive anthropogenic Hg emission sources over the passing areas of air masses (Figure 4), particularly over the Gulf of Thailand. Therefore, we could expect the limited contribution of regional Hg sources to TGM concentrations at HCMC during the study period. This conclusion is also supported by $PM_{2.5}$ data observed at the HCMC US embassy during the study period with average $PM_{2.5}$ concentrations of 16.3 ± 6.1 μg m^{-3}, significantly lower than that of annual $PM_{2.5}$ concentration (36.3 ± 13.7 μg m^{-3}, [24]). More importantly, air mass transport patterns are similar between daytime and nighttime samples for each sampling day (Figure 4), suggesting the same regional air mass affects the sampling site for each day-long period. Consequently, this allowed us to evaluate the contribution of local Hg sources quantitatively by assuming nighttime data is free from local activities (i.e., limited residential burning).

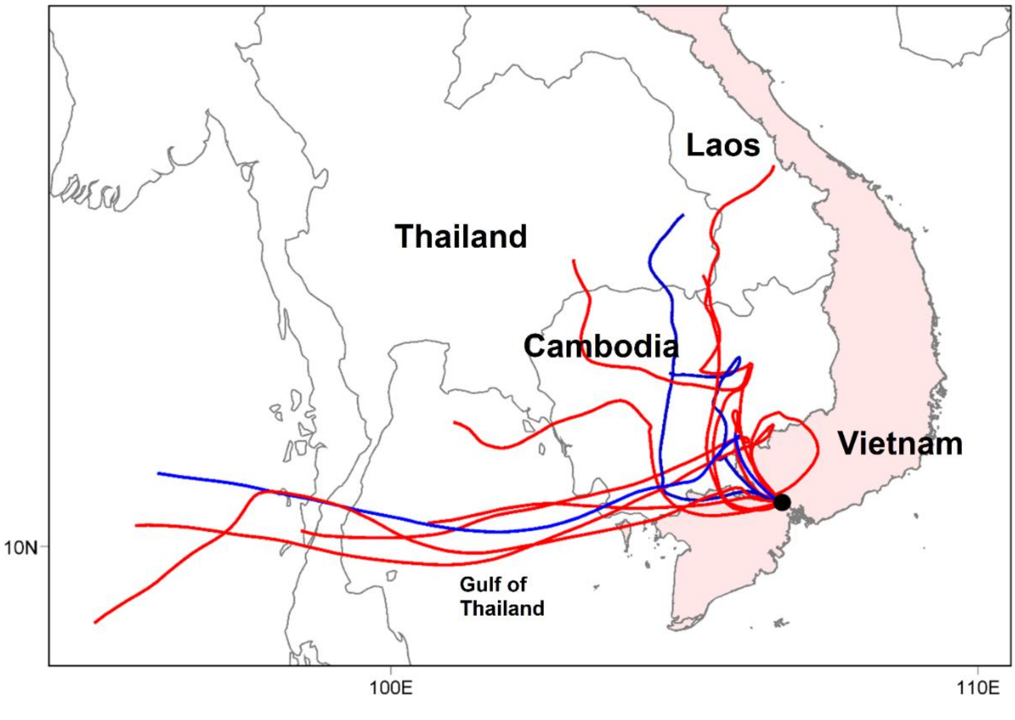

Figure 4. *72-hour backward trajectories (BWTs) started from CL (black dot) during the sampling periods. Nighttime (7 pm – 7 am) BWTs are in blue color and daytime (7 am – 7 pm) BWTs are in red.*

The average outdoor nighttime TGM concentration at CL was 1.58 ± 0.28 ng m^{-3} with day-by-day TGM values shown in Figure 5. This TGM value was comparable to the background TGM level in the northern hemisphere (1.4-1.6 ng m^{-3}; [36]), implying that background air dominated during nighttime at CL during the campaign. This is also supported by the analysis of BWTs and $PM_{2.5}$ concentrations mentioned in the above paragraph. By assuming burning activities are the major contributor to the elevation of TGM levels at the sampling site, we proposed equation 1 to further estimate the contribution of local residential combustion activities (%), and the results were shown in Figure 5. This approach has also been employed in the previous study to estimate the contribution of well-known sources on specific air pollutants [7], [11], [34]. The results suggested that, during the study period, residential combustion contributes about 70% (from 9-147%) to TGM concentration at CL. A larger contribution was obtained on major Lantern Festival days (February 4-5) as compared to normal days (February 6-7), corresponding to the increase in festival activities (i.e., burning of joss paper). Our results, therefore, indicated that residential combustion is an important source of atmospheric Hg, contributing to the high levels of TGM in the urban area of HCMC. In addition, residential burning for heating purposes is typically more prevalent in temperate regions during the winter [37], [38] while in HCMC, it is mainly for cooking and other activities including spiritual practices. Such burning activities in HCMC take place all year round and can therefore be considered as a continuous and significant anthropogenic Hg emission source here as well as in similar tropical urban areas in Southeast Asia.

$$\text{Contribution (\%)} = 100 \times \left(\frac{\text{Cday}}{\text{Cnight}} - 1\right) \quad (1)$$

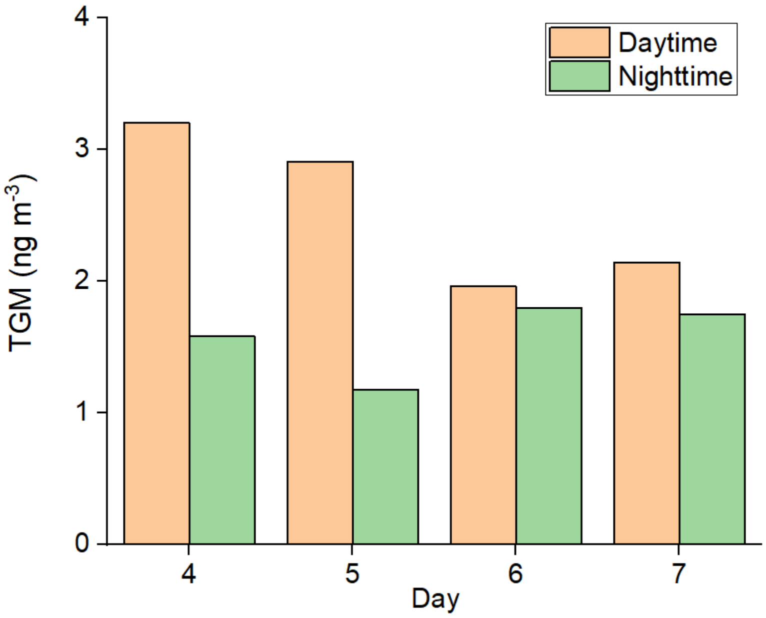

Figure 5. *Daytime vs nighttime outdoor TGM concentrations during the sampling period*

4. Conclusions

This study was conducted to assess the impact of residential burning (e.g., cooking, joss burning) on atmospheric Hg (TGM) levels in an urban area of HCMC, Vietnam. A total of 24 TGM samples (18 outdoor and 6 indoor) were collected during 4-7 February 2023 at Cholon, a busy market in the central HCMC. The average TGM concentration for outdoor samples was 2.27 ± 0.81 ng m^{-3}, in which daytime TGM concentration (2.56 ± 0.80 ng m^{-3}) was significantly greater than that of nighttime (1.58 ± 0.28 ng m^{-3}) attributing to the contributions from local burning activities. In addition, the increase in residential burning due to Lantern Festival activities significantly contributed to the elevation in the concentration of TGM at CL. Under well-ventilated conditions, the indoor TGM concentration is determined by the outdoor air. BWTs analysis was performed to investigate the contribution of regional Hg sources to the study area, suggesting the domination of background air mass during the sampling period. We estimated that residential burning promoted an increase in atmospheric Hg concentrations by about 70% at Cholon during the study period. This study indicates the important role of local burning in atmospheric Hg while further studies are crucial to better understand the role of this source in atmospheric Hg emission inventory in HCMC as well as southern Vietnam.

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