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Household air pollution from, and fuel efficiency of, different coal types following local cooking practices in Xuanwei, China☆

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ABSTRACT

The domestic combustion of smoky (bituminous) coal in the Chinese counties of Xuanwei and Fuyuan, are responsible for some of the highest rates of lung cancer in the world. Cancer rates vary between coal producing regions (deposits) in the area, with coals from Laibin exhibiting particularly high risks and smokeless (anthracite) coal exhibiting lower risks. However, little information is available on the specific burning characteristics of coals from throughout the area. We conducted an extensive controlled burning experiment using coal from multiple deposits in either a traditional firepit or ventilated stove, accompanied by a detailed examination of time-weighted and real-time size-aggregated particle concentrations. Smoky coal caused higher particle concentrations of all sizes than smokeless coal, with variations observed by geological source. Virtually all particle emissions were in the $PM_{2.5}$ fraction (98% - mass based), and 75% and 46% were in the PM_1 and $PM_{0.3}$ fraction respectively. Real-time concentrations of PM_1 and $PM_{0.1}$ peaked after coal was added and declined afterwards. Ventilation reduced particle concentrations by up to 15-fold and increased the coal burning rate by 1.9-fold. These findings may provide valuable insight for reducing exposure and adverse health effects associated with domestic coal combustion.

1. Introduction

Solid fuel (animal dung, crop residue, wood, coal etc.) is a major domestic energy source for around 3 billion people world-wide (World Health Organization, 2006), primarily from low-and-middle income countries. The emissions from these fuels have been recognized as a major source of household air pollution (HAP), exposure to which is responsible for up to 4 million global deaths per year, from a variety of causes including pneumonia, chronic obstructive pulmonary disease, and lung cancer (Gordon et al., 2014). Among different types of solid fuels, coal is particularly noteworthy as indoor emissions from household coal combustion have been designated as Group I carcinogens by the International Agency for Research on Cancer (IARC) (IARC Working Group on the Evaluation of Carcinogenic Risks to Humans, 2012).

Xuanwei and Fuyuan counties, in Yunnan province, China have among the highest lung cancer rates both in China and the world (Lin

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et al., 2015; Chen et al., 2015a). Studies exploring the etiology of lung cancer in the region have shown that domestic combustion of locally sourced "smoky" coal (a late Permian bituminous coal) was primarily responsible for the excessive lung cancer risks (Li et al., 2019; Lan et al., 2008; Chen et al., 2015b; Barone-Adesi et al., 2012). A retrospective cohort study (Barone-Adesi et al., 2012) showed that domestic smoky coal usage would increase lung cancer mortality rates by up to 100-fold (hazard ratio for women: 98.8, 95% CI 36.8–276.6) compared to smokeless coal (a carboniferous anthracite coal available in some parts of the region). In addition to the large difference between coal types, notable variation in lung cancer mortality was observed by geological sources of smoky coal. For example, the odds ratio of lung cancer associated with smoky coal ranged from 7.49 to 33.40, depending on the geological source (Wong et al., 2019).

Stove improvements (e.g. replacing unvented firepits with standing stoves with a dedicated chimney) have been observed to reduce both HAP and disease burden (Chapman et al., 1999; Chapman et al., 2005). In Xuanwei, such stove improvements have been associated with up to 50% reductions in pneumonia (Shen et al., 2009) and lung cancer mortality (Lan et al., 2002; Seow et al., 2014). Exposure assessment studies have shown that ventilated stoves have been associated with substantial reductions in multiple HAP constituents including $PM_{2.5}$, CO, BC, PM_{10}, NO, NO_x, BaP and other PAHs (Downward et al., 2014a; Downward et al., 2016; Hu et al., 2014; Seow et al., 2016; Tian et al., 2009) compared to conventional stoves.

We have previously studied multiple components of the HAP in Xuanwei, including $PM_{2.5}$ and PAHs, in a large-scale field exposure assessment (Downward et al., 2014a; Downward et al., 2017). However, as this was done under "real-world" conditions (i.e. people carried out their day-to-day lives), fuel and stove usage was performed in a non-uniform way. Additionally, exposures were measured over 24 h, meaning that transient but heavy HAP exposures (e.g. during cooking) were not explicitly captured (Hosgood et al., 2012), which may be important for understanding the relationship between domestic coal combustion and lung cancer as duration of cooking is strongly and positively associated with lung cancer incidence in both men and women (Lan et al., 2002; Hosgood et al., 2008). Further, no metric of stove efficiency was reported, impacting the ability to evaluate the broader effectiveness of the stove improvements in the region. Some studies have examined coal emissions under controlled (i.e. laboratory) conditions (Chen et al., 2005; Tian et al., 2008; Venkataraman and Rao, 2001; Buhre et al., 2006). However, testing under extremely confined conditions limits generalizability to "real world" conditions as lab practices (e.g. grinding coal samples) do not occur within peoples' homes. To address these limitations, we performed a series of water boiling tests (based on the protocol developed by the Global Alliance for Clean Cookstoves), where a known quantity of water was brought to the boil and simmered in a controlled and consistent fashion within a typical Xuanwei home.

The aims of the research presented here are to compare the HAP caused by the combustion of different coal types and related burning efficiencies, and to determine whether ventilation can help to decrease HAP and improve burning efficiencies.

2. Methodology

We performed a series of water boiling tests in a typical Xuanwei household using coal from multiple sources (Table S1), using either a ventilated stove or firepit. A total of 31 tests were conducted with several coal samples, including those from areas with particularly high rates of lung cancer. For all tests, the goal was to bring a pot, containing 2.8–3.5 L of water, from room temperature to the boil, before simmering for 45 min. The emissions were measured, and fuel efficiency factors were calculated.

2.1. Description of the study site, stoves and fuels

The current study was performed in a typical Xuanwei kitchen, located in the village of "Laibin" at an altitude of 1943 m. The house was located away from direct pollution sources (e.g. major roads) and approximately 6 km away from the nearest major town. All tests were performed using either a traditional style unvented firepit or a pre-existing ventilated stove which was installed over 10 years prior and represented a typical ventilated stove for the area. The kitchen was 3.69 m × 5.15 m × 1.92 m (width × depth × height) and contained a door at each end. There was one window in the room, 1.6 m × 1.0 m (width × height), located by the ventilated stove. The firepit was 0.5 m × 0.7 m × 0.5 m (width × length × height) and constructed from brick (Figure S1 A). The ventilated stove (Figure S1 B) had a separate burning chamber with a passage beneath to facilitate air flow and had a dedicated chimney (which was over 4 m in height) to vent smoke outside (Figure S1 C). The heating surface of the ventilated stove was round with an external diameter of 25 cm and an internal diameter of 20 cm. The fuel area was 17 cm below the surface and was 50 cm in both width and length, with an ash chamber (50 cm × 50 cm × 65 cm width × height × length) beneath it.

Table S1 shows the different coals used for the tests. Coal samples consisted of ten smoky and two smokeless coal samples that had been collected recently (2010–2017) and two smoky and one smokeless coal that had been collected in the 1980s and subsequently stored. There is a potential risk that, due to changes in mining depth or position, coals produced today are not reflective of those previously used – thus limiting generalizability to historical lung cancer rates. Analyzing the historical coal samples allows for direct comparisons between HAP from coal collected in the present with those from the 1980s. Figure S2 shows the locations of the mines where the coal samples were collected from (Wong et al., 2019). As with our previous studies, we used the standard classification by the State Standard of China Coal Classification to divide smoky coal into the following subtypes: 1/3 coking coal, coking coal, gas fat coal and meager lean coal (Downward et al., 2017). Many coal subtypes came from more than one deposit, in which case individual deposits were recorded for later comparison.

2.2. Description of the adapted water boiling tests

All tests were performed with the assistance of a local resident to ensure burning practices adhered to the local "normal" manner. In all tests, wood and dried corncob were used as kindling (ignition stage), with coal added when judged appropriate by the local resident (coal stage). A pot containing a known quantity of room-temperature water was put onto the stove and was brought to the boil as fast as possible (high power stage) and then left to simmer for 45 min (low power stage). Additional fuel was added as needed to maintain the fire (all additions were documented). The doors were kept open, and the window was shut during each test.

The weights of all fuels at the start and end of each test, alongside the weight of any ash, were recorded. Water temperature was measured by a digital thermometer in combination with a visual inspection of the water to document the time taken to bring the water to boil. To calculate evaporation losses, the quantity of water in the pot was recorded before it was added to the fire, once it was boiling and after simmering.

2.3. Household air pollution from coal combustion

HAP was measured using multiple real-time and time-weighted devices, including a micro-orifice impactor (MOI Model 100-NR, TSI Incorporated), a DustTrak DRX Aerosol Monitor 8533 (DRX, TSI Incorporated) and a TSI P-Trak 8525 (P-trak, TSI Incorporated), these devices were placed at equal distances (2 m) from the stoves in each test. Another DRX (TSI Incorporated) was used to measure outdoor particulate matter (PM) concentrations and placed on the roof of another residence (approximately 4-stroties tall) which was approximately 30 m away from the residence where the tests were performed. All equipment underwent daily calibration and maintenance.

Time-weighted and size differentiated PM was collected with a cascade impactor (MOI), which was equipped with 5 pre-weighed Teflon filters (PTFE membrane with PMP ring, 47 mm diameter, 2.0 μm pores) to collect PM with an aerodynamic diameter <0.3 μm ($PM_{0.3}$), 0.3–1 μm ($PM_{0.3\text{-}1}$), 1–2.5 μm ($PM_{1\text{-}2.5}$), 2.5–10 μm ($PM_{2.5\text{-}10}$) and >10 μm (PM > 10 μm) separately. In order to prevent filter and pump from overloading in a high exposure setting, the MOI was optimized to a "1min on, 4min off" sampling approach, beginning up to 10min before the tests to achieve a stable flow rate and turned off once the simmering ended. All times and flow rates (pre- and post-test) were recorded. After sample collection, filters were packed into petri slides and stored in zipped amber plastic bags. Until weighing, filters were stored at 4 °C. Filters were weighed in duplicate before and after sampling using a microbalance. The particle mass concentrations were calculated by dividing mass collected on filters by the total air volume drawn through the filters.

Real time measurements were collected with a DRX and a P-Trak every second. Mass concentrations for both indoor and outdoor PM_{10}, respirable particle (RESP), $PM_{2.5}$ and PM_1 were measured by DRX. Indoor and outdoor measurements were compared to evaluate whether the indoor pollution levels were influenced by outdoor air pollution. For the current paper, we will focus on PM_1 mass concentrations (reasons given below), but full results are available on reasonable request. The number concentration of ultrafine particles ($PM_{0.1}$) was measured with P-Trak. To examine changes in HAP concentrations in real-time, measurements were broken down into different burning stages: Pre (5 min before ignition), ignition stage (adding wood and corncob), coal stage (after adding coal and before putting pot on fire), high power stage (HP, after placing the pot on the fire until the water came to the boil), low power stage (LP, 45-min-simmering) and post (up to 30 min after simmering ended).

2.4. *Fuel and stove efficiency*

Two efficiency metrics are reported - burning rate (g/min) and temperature & weight corrected time to boil (T.W_time to boil, min/L). Burning rate represents the speed of coal being burned after it has been added to the fire, the mass of burned coal is the difference between weight of coal at the start and end of each test (Table S1). T.W_time to boil indicates the time it takes to boil 1 L of water with a temperature rise of 75 °C (25 °C → 100 °C). Equations describing these calculations are presented in the supplement.

The amount of water evaporated during the high-power stage was monitored for QAQC standards as excessive evaporation negates the efficiency metrics. Our results (median 4%, range 1%–6%) are within the acceptable limits as stipulated by the Global Alliance for Clean Cookstoves.

2.5. *Statistics*

Preliminary examination of the data indicated that the MOI, DRX, P-Trak measurements, and the efficiency metrics tended towards log-normality. Therefore, results were summarized by geometric means (GM) and geometric standard deviations (GSD). In addition, median and interquartile range were summarized for contribution of different size-fractions. Correlations between indoor and outdoor PM_1 concentrations; between measurements from DRX and MOI; and between particle of different size fractions were examined by the Spearman's correlation test. Data was analyzed in R (version 3.62) with the Chron and Envstats packages (James et al., 2020; Millard, 2013; RStudio Team, 2020).

3. Results and discussion

3.1. *Burn description*

Of the 31 water boiling tests, 26 successfully brought water to boil and were retained for further analysis (Table S2; Figure S3). Among these 26 tests, three had incomplete MOI measurements (machine failure) and one test with an extreme outlier was excluded (over 20 folds higher than another experiment using the same stove and coal samples, and over 20 folds higher than all other smoky coal experiments using the same stove). Complete DRX and P-Trak measurements are available for 24 tests (Figure S3). The median weight of coal used across the experiment was 1.12 kg (IQR: 1.02, 1.42. Table S1). Of the five tests which did not bring water to the boil, four were from smokeless coal samples and one coking coal sample, the water temperature was raised to over 70 °C (data not shown).

3.2. *PM concentrations*

3.2.1. *Size aggregated concentrations of particulate material from MOI measurements*

Regardless of stove type, the majority of the particles generated during the burning of both smokeless and smoky coal were of a smaller size. Of the particles collected from 22 water boiling tests with full MOI measurements, we found that $PM_{2.5}$ contributed to approximately 98% (IQR 96%–99%) of the total particle mass, PM_1 to 75% (IQR 67%–83%) and $PM_{0.3}$ 46% (IQR 38%–53%) (Table 1, Fig. 1); the four failed tests also showed similar size-distributions (Figure S4). These findings are also in accordance with previous studies (Tian et al., 2008; Mumford et al., 1987) where 51% and 90% of the particulate mass in Xuanwei and Fuyuan was reported to be in the PM_1 fraction. Chen et al. (2005) previously reported that 94% of the particles emitted from five Chinese coal samples (one sub-bituminous, four bituminous and one anthracite coal) were in the <0.95 μm fraction, and 85% were in the <0.49 μm fraction. Buhre et al. (2006) also reported that the PM_1 fraction constituted up to 74% of $PM_{2.5}$ during the combustion of five Australian bituminous coals. It is also noteworthy to mention that the historical smoky coal samples (collected in the 1980s) showed identical size-distribution patterns as recent smoky coal samples (collected between 2010 and 2017) (Figure S5; Table S3). In the current study, correlation analysis (Fig. 2) found that $PM_{0.3}$ highly correlated with $PM_{0.3\text{-}1}$ and $PM_{1\text{-}2.5}$, while moderately with $PM_{2.5\text{-}10}$ and PM over 10 μm (Spearman's correlation coefficients 0.81, 0.75, 0.47 and 0.51 respectively). Similarly, $PM_{0.1}$ correlated moderately with $PM_{0.3}$, $PM_{0.3\text{-}1}$ and $PM_{1\text{-}2.5}$ while poorly to $PM_{2.5\text{-}10}$ and PM over 10 μm (Spearman's correlation coefficient: 0.63, 0.69, 0.64, 0.38 and 0.23 respectively), the relatively weaker correlation might be partially explained by the differences between number concentration and mass concentration. Our earlier research (Hosgood et al., 2012) reported that coal burning contributed to indoor nanoparticle levels, and the aggregated surface area of nanoparticles was only moderately correlated with $PM_{2.5}$ ($r^2 = 0.43$, $p = 0.11$). Collectively, smaller size fractions contributed the majority of the particles in coal smoke, and household coal combustion contributed to indoor nanoparticle levels.

Among the 22 tests with complete MOI data, one was smokeless coal (burned in the ventilated stove) and 21 were smoky coal (6 burned in the ventilated stove and 15 in the firepit). In general, the particle concentrations from the smoky coal experiments were higher, both overall and in different size fractions, than smokeless. As shown in Table 1, the highest concentrations (for the ventilated stove) within the $PM_{2.5}$, PM_1 and $PM_{0.3}$ fractions came from coking coal (1.76 mg/m^3 for $PM_{2.5,}$ 1.52 mg/m^3 for PM_1 and 1.00 mg/m^3 for $PM_{0.3}$) and the lowest from smokeless coal (0.42 mg/m^3 for $PM_{2.5,}$ 0.38 mg/m^3 for PM_1 and 0.23 mg/m^3 for $PM_{0.3}$). Gas fat coal was the second highest in particulate concentrations (1.69 mg/m^3 for $PM_{2.5,}$ 1.47 mg/m^3 for PM_1 and 0.95 mg/m^3 for $PM_{0.3}$) followed by meager lean coal (0.51 mg/m^3 for $PM_{2.5,}$

Table 1
Time-weighted measurement of concentration (mg/m^3) of particles of different sizes by MOI.

		N		PM > 10		$PM_{2.5\text{-}10}$	
Ventilation		Yes	No	Yes	No	Yes	No
smokeless coal		1	0	NA (NA)	NA	0.02(NA)	NA
Smoky coal		6	15	0.02 (1.56)	0.03 (2.25)	0.05 (1.66)	0.09 (2.22)
	1/3 Coking coal	1	1	0.01(NA)	0.06(NA)	0.02(NA)	0.12(NA)
	Coking coal	2	10	0.02 (1.68)	0.02 (1.85) (n = 9)	0.07 (1.10)	0.07 (2.16)
	Gas fat coal	1	1	0.02(NA)	0.12(NA)	0.08(NA)	0.19(NA)
	Meager lean coal	2	3	0.02 (1.50)	0.04 (3.13)	0.04 (1.27)	0.15 (2.34)

Size aggregated concentration of MOI (mg/m^3) measurements

		N		$PM_{1\text{-}2.5}$		$PM_{0.3\text{-}1}$		PM < 0.3	
Ventilation		Yes	No	Yes	No	Yes	No	Yes	No
smokeless coal		1	0	0.04 (NA)	NA	0.15 (NA)	NA	0.23 (NA)	NA
Smoky coal		6	15	0.21 (2.09)	3.19 (3.17)	0.35 (1.61)	4.05 (2.34)	0.70 (1.74)	5.17 (2.13)
	1/3 Coking coal	1	1	0.08 (NA)	4.21 (NA)	0.18 (NA)	4.00 (NA)	0.25 (NA)	6.64 (NA)
	Coking coal	2	10	0.24 (1.10)	2.26 (3.54)	0.52 (1.01)	2.98 (2.32)	1.00 (1.01)	4.93 (1.91)
	Gas fat coal	1	1	0.22 (NA)	10.41 (NA)	0.52 (NA)	8.87 (NA)	0.95 (NA)	9.11 (NA)
	Meager lean coal	2	3	0.27 (3.59)	6.24 (1.41)	0.27 (1.29)	8.70 (1.65)	0.71 (1.44)	4.62 (3.96)

PM > 10, $PM_{2.5\text{-}10}$, $PM_{0.3\text{-}1}$ PM < 0.3 μm are particles with aerodynamic diameter over 10 μm, between 2.5 μm and 10 μm, between 0.3 μm and 1 μm and smaller than 0.3 μm respectively. Geometric means of concentration values are presented. Data are shown as "geometric mean (geometric standard deviation)". N: number of burns. NA: data not available. MOI: micro-orifice impactor.

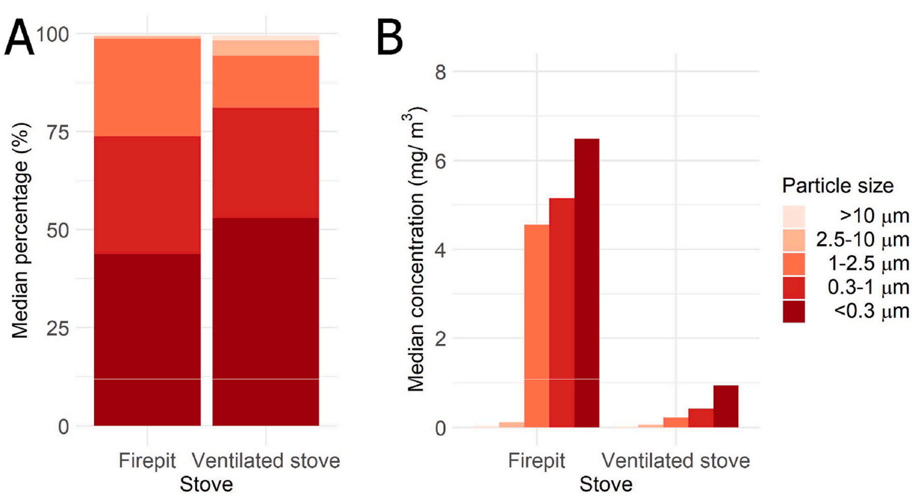

Fig. 1. Mass based size distribution of MOI measured particles from water boiling tests with smoky coal in ventilated stove (n = 6) and firepit (n = 15). A shows contribution of different size fractions to total particle mass, B shows the absolute concentrations of different size fractions.

0.43 mg/m^3 for PM_1 and 0.25 mg/m^3 for $PM_{0.3}$).

Specific coal sources (i.e. deposit numbers (Wong et al., 2019)) for the coking and gas fat coal subtypes were examined by comparing burns performed in the firepit (Table S4). In general, variation in measurements below the 2.5 μm fraction was observed for coking coal. Within coking coal, samples from deposit 16 had the highest $PM_{1\text{-}2.5}$ concentration (6.52 mg/m^3), while samples from deposit 9 had the highest concentration of $PM_{0.3\text{-}1}$ (4.76 mg/m^3) and $PM_{0.3}$ (7.37 mg/m^3), followed by samples from deposit 4 and deposit 10. Samples from deposit 8 had the lowest concentration of $PM_{1\text{-}2.5}$ (0.11 mg/m^3), $PM_{0.3\text{-}1}$ (0.68 mg/m^3), $PM_{0.3}$ (1.05 mg/m^3). We have previously reported that the geographic source of smoky coal is important for both chemical composition and lung cancer risk (Wong et al., 2019; Downward et al., 2014b). Similarly, in the current paper, when comparing PM concentrations caused by different smoky coal subtypes, we observed notable heterogeneity in findings. For example, the MOI measurements derived from gas fat coal were generally the highest while the lowest measurements were from 1/3 coking coal (in the ventilated stove) and coking coal (in the firepit), which parallels earlier findings that gas fat coal had the highest hydrocarbon content (Downward et al., 2014b). However, when looking at specific water boiling test stages, we found that coking coal led to the highest PM_1 and $PM_{0.1}$ concentrations during both the HP and LP stages (further discussed below). Similarly, in a previous study (Hu et al., 2014), we observed significant variation between the four designated smoky coal subtypes (coking coal, 1/3 coking coal, gas fat coal, meager lean coal) for indoor $PM_{2.5}$ in Fuyuan.

In addition to differences between coal types and deposit, using the ventilated stove reduced particle concentrations. For smoky coal overall, concentrations of $PM_{1\text{-}2.5}$, $PM_{0.3\text{-}1}$, $PM_{0.3}$ were reduced by 15 (3.19 mg/m^3 →0.21 mg/m^3), 12 (4.05 mg/m^3→0.35 mg/m^3) and 7 (5.17 mg/m^3→0.70 mg/m^3) folds respectively (Table 1), Despite a consistent effect of ventilation across all experiments, the degree of effect varied across particle sizes, with the largest reduction in absolute concentration found in smaller particles. Understanding the impact of these smaller particles may help understand the etiology of coal smoke induced lung cancer as well as promoting health improvements. In addition, the patterns of particle size distribution also differed between the ventilated stove and firepit (Fig. 1). Among particles emitted from smoky coal, the majority of the particles collected from the ventilated stove (median 53%) were in the <0.3 μm fraction, which is higher than that of the

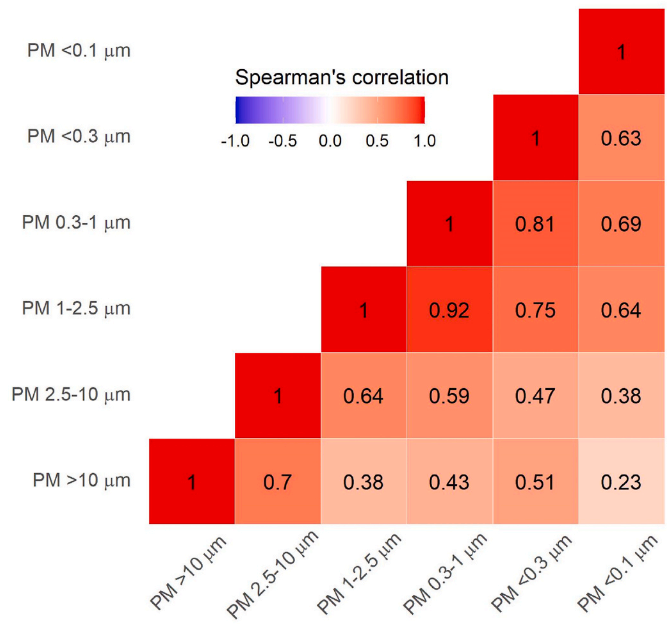

Fig. 2. Correlation matrix for different size fractions from 22 water boiling tests. PM < 0.1 μm, PM < 0.3 μm, PM 0.3–1 μm, PM 1–2.5 μm, PM 2.5–10 μm and PM > 10 μm represent particulate matter with aerodynamic diameter of < 0.1 μm, <0.3 μm, 0.3–1 μm, 1–2.5 μm, 2.5–10 μm and >10 μm respectively. PM < 0.3 μm, PM 0.3–1 μm, PM 1–2.5 μm, PM 2.5–10 μm and PM > 10 μm are from time-weighted measurements by MOI. PM < 0.1 μm is from real-time measurements by P-Trak. The 22 water boiling tests includes one test with smokeless coal and 21 with smoky coals.

firepit (44%). By contrast, the 1–2.5 μm fractions contributed less to the total emissions of the ventilated stove than those of the firepit (median 12% vs 25%). The effect of ventilation also appeared to differ between the different coal subtypes, although we must acknowledge relatively small sample sizes for this comparison. For example, the HAP from gas fat coal had the greatest reduction for $PM_{1\text{-}2.5}$ (10.41 mg/m^3→0.22 mg/m^3), $PM_{0.3\text{-}1}$ (8.87 mg/m^3→0.52 mg/m^3) and $PM_{0.3}$ (9.11 mg/m^3→0.95 mg/m^3). By contrast, coking coal had the lowest reduction for $PM_{1\text{-}2.5}$ (3.16 mg/m^3→0.24 mg/m^3), $PM_{0.3\text{-}1}$ (3.76 mg/m^3→0.52 mg/m^3) and $PM_{0.3}$ (5.69 mg/m^3→1.00 mg/m^3).

Improved stoves are a common strategy against HAP and its associated health risks not just in Xuanwei and Fuyuan, but also around the world. Previous studies have also shown that the use of improved cookstoves reduced pollution levels (Shen et al., 2009; Downward et al., 2016; Hu et al., 2014). Such reduction in pollution may be reflected in health studies. For example, when compared to firepits, stoves with a chimney reduced lung cancer risks in Xuanwei for both men and women by 50% (Lan et al., 2002). The reduction was also found to be associated with 50% reduction in pneumonia deaths (Shen et al., 2009). While these studies have reported that stove improvements have reduced lung related diseases in Xuanwei and Fuyuan, it remains unclear which size fractions are more important to these health gains. It has been argued that smaller particles might impose higher health risk than their bulkier counterparts due to increased potency (Miller and Poland, 2020; Nho, 2020), suggesting that there is still room for health improvements by further targeting the small-sized household air pollutants.

3.2.2. PM_1 and $PM_{0.1}$ generated during different burning stages

As PM_1 constitutes 76% of the total particle mass and $PM_{0.3}$ 46% (in time-weighted measurements), we examined real time emissions of PM_1 from the DRX and $PM_{0.1}$ from the P-trak (the size fraction measured by P-track), with concentrations broken down into different burning stages (Table 2). In general, regardless of fuel and stove type, PM_1 emissions increased when transitioning from the ignition to coal stage and declined during the HP and LP stages. For example, the PM_1 concentration for smokeless coal tested in the ventilated stove were:0.02 mg/m^3 (background) → 0.16 mg/m^3 (ignition stage) →7.10 mg/m^3 (coal stage) →0.11 mg/m^3 (HP stage) →0.02 mg/m^3 (LP stage). The PM_1 and $PM_{0.1}$ concentrations for smoky coal samples tested in firepits also showed similar temporal transitions (Fig. 3). Concentrations and the temporal transition patterns of PM_1 and $PM_{0.1}$ were also similar between historical and recent smoky coal samples (Figure S6; Table S5-6).

The indoor measurements for PM_1 from the MOI and DRX were moderately correlated (Spearman's correlation coefficient: 0.56, p-value: <0.01). However, indoor PM_1 concentrations (as measured by DRX) were not correlated with outdoor PM_1 concentrations across all burning stages (Figure S7), suggesting outdoor air pollution levels had minimal influences on indoor levels during these tests.

Ventilation shortened the ignition and coal stages (Table 2). For the ventilated stove, very little time (in most cases none) was judged necessary for the coal stage before proceeding to the HP stage. By contrast, the amount of time judged necessary for the coal stage in the firepit could be as long as 50 min (29 min on average). Within the smoky coal tests, the HP stage was slightly longer for the ventilated stove than the firepit (41 min vs 35 min), however the amount of time varied between different fuel types. Stove ventilation also reduced PM_1 and $PM_{0.1}$ concentrations across all burning stages (Table 2). For example, PM_1 from smoky coal during the HP stage were reduced from 37.89 mg/m^3 to 3.79 mg/m^3, and from 6.00 mg/m^3 to 0.69 mg/m^3 in the LP stage. There was variation in the magnitude of reduction (from 0.9 fold to 116 fold) depending on fuel types and stages, for example, during the HP stage, PM_1 concentrations were reduced by 25 fold (from 69.71 mg/m^3 to 2.99 mg/m^3) for coking coal and by 2.5 fold (from 10.26 mg/m^3 to 4.05 mg/m^3) for meager lean coal; during the LP stage, PM_1 concentrations were reduced by 116 fold (from 10.46 mg/m^3 to 0.09 mg/m^3) for 1/3 coking coal and by 2.7 fold (from 1.74 mg/m^3 to 0.64 mg/m^3) for meager lean coal. Similar reductions were seen for $PM_{0.1}$ emissions of smoky coal tested in firepits, with reductions from 1.63×10^5 #/cm^3 to 0.71×10^5 #/cm^3 in the HP stage, and from 1.33×10^5 #/cm^3 to 0.40×10^5 #/cm^3 in the LP stage.

For tests in the ventilated stove, we focused on HAP during the HP and LP stages (Table 2) as the coal stage was relatively short. Concentrations of PM_1 from smoky coal were over 30 times higher than smokeless coal in both the HP (3.79 mg/m^3 vs 0.11 mg/m^3) and LP stage (0.69 mg/m^3 vs 0.02 mg/m^3). However, the relative difference was less for concentrations of $PM_{0.1}$, in which smoky coal was less than twice as high than smokeless coal in both the HP (0.71×10^5 #/cm^3 vs 0.40×10^5 #/cm^3) and LP stage (0.40×10^5 #/cm^3 vs 0.30×10^5 #/cm^3). Regarding the variations within smoky coal sub-types, the highest PM_1 concentration came from gas fat coal in the HP stage (8.22 mg/m^3) and in the LP stage (2.20 mg/m^3). The highest $PM_{0.1}$ concentration came from gas fat coal (0.98×10^5 #/cm^3) in HP stage and from coking coal (0.62×10^5 #/cm^3) in LP stage.

We found that at virtually all stages of the water boiling tests, smoky coal had much higher PM concentrations than smokeless and that using a ventilated stove can greatly reduce indoor air pollution and improve stove efficiency. In our previous coal composition publication (Downward et al., 2014b), we reported that smoky coal bore 5–15 times the hydrocarbon content than smokeless coal. Such compositional differences might explain the increased PM concentrations. Smaller controlled burning studies have also reported the difference in emissions between smoky and smokeless coal. For example, Zhang et al. (2008) reported that the $PM_{2.5}$ emission factors of bituminous coal was 7-fold that of anthracite coal. Our earlier study, based on "real world" exposures (Hu et al., 2014), also reported smoky coal was associated with higher personal and indoor $PM_{2.5}$ concentration than smokeless coal.

Deposit specific variation of coking and gas fat coal was examined by comparing burns performed in the unvented stove (Table S7-8). Within coking coal samples, the highest PM_1 concentration came from deposit 4 (116.67 mg/m^3) in the HP stage and from deposit 10 (12.60 mg/m^3) in

Table 2
Time (min) for each burning stage, real-time measurements for PM_1 (mg/m^3) and $PM_{0.1}$ ($\times 10^5$ #/cm^3) by different burning stages.

	Ventilation		N		Pre		Ignition		Coal	
			Yes	No	Yes	No	Yes	No	Yes	No
Time	Smokeless coal		1	0	5 (0)	NA	7(NA)	NA	6(NA)	NA
	Combined smoky coal		7	18	5 (0)	5 (0)	2 (2)	7 (3)	0 (1)	29 (13)
		1/3 Coking coal	1	1	5 (0)	5 (0)	1(NA)	5(NA)	0(NA)	43(NA)
		Coking coal	3	11	5 (0)	5 (0)	3 (3)	7 (3)	0 (0)	26 (14)
		Gas fat coal	1	2	5 (0)	5 (0)	3(NA)	8 (4)	0(NA)	28 (2)
		Meager lean coal	2	4	5 (0)	5 (0)	2 (1)	9 (2)	2 (2)	34 (12)
PM_1	Smokeless coal		1	0	0.02(NA)	NA	0.16(NA)	NA	7.10(NA)	NA
	Combined smoky coal		7	16	0.07 (2.31)	0.18 (5.04)	0.55 (5.19)	6.51 (3.54)	2.63(NA)	59.12 (3.08)
		1/3 Coking coal	1	1	0.05(NA)	0.02(NA)	0.11(NA)	0.83(NA)	NA (NA)	100.62(NA)
		Coking coal	3	10	0.04 (2.14)	0.15 (3.33)	1.87 (12.59)	8.06 (4.13)	NA (NA)	88.04 (1.81)
		Gas fat coal	1	1	0.08(NA)	0.32(NA)	0.95(NA)	7.37(NA)	NA (NA)	48.23(NA)
		Meager lean coal	2	4	0.16 (1.69)	0.41 (11.47)	0.28 (1.50)	6.19 (1.85)	2.63(NA)	20.12 (5.80)
$PM_{0.1}$	Smokeless coal		1	0	0.03(NA)	NA	0.34(NA)	NA	3.15(NA)	NA
	Combined smoky coal		7	16	0.07 (0.00)	0.08 (0.34)	0.31 (0.00)	1.28 (0.00)	3.15(NA)	NA
		1/3 Coking coal	1	1	0.11(NA)	0.02(NA)	0.25(NA)	0.36(NA)	0.89(NA)	2.25 (0.00)
		Coking coal	3	11	0.08 (0.00)	0.11 (0.31)	0.73 (0.00)	1.58 (0.00)	NA (NA)	1.75(NA)
		Gas fat coal	1	2	0.11(NA)	0.03 (0.41)	0.46(NA)	0.78 (0.00)	NA (NA)	2.66 (0.00)
		Meager lean coal	2	2	0.05 (0.00)	0.05 (0.47)	0.13 (0.00)	1.20 (0.00)	NA (NA)	1.62 (0.00)

	Ventilation		N		HP		LP	
			Yes	No	Yes	No	Yes	No
Time	Smokeless coal		1	0	69(NA)	NA	45(NA)	NA
	Combined smoky coal		7	18	41 (18)	35 (29)	45 (0)	45 (1)
		1/3 Coking coal	1	1	27(NA)	33(NA)	45(NA)	45(NA)
		Coking coal	3	11	53 (17)	41 (35)	45 (0)	46 (2)
		Gas fat coal	1	2	18(NA)	14 (1)	46(NA)	45 (0)
		Meager lean coal	2	4	42 (9)	30 (6)	46 (1)	45 (0)
PM_1	Smokeless coal		1	0	0.11(NA)	NA	0.02(NA)	NA
	Combined smoky coal		7	16	3.79 (2.49)	37.89 (4.45)	0.69 (3)	6.00 (3.08)
		1/3 Coking coal	1	1	3.13(NA)	78.78(NA)	0.09(NA)	10.46(NA)
		Coking coal	3	10	2.99 (1.29)	69.71 (1.81)	1.00 (2.16)	9.33 (1.96)
		Gas fat coal	1	1	8.22(NA)	7.59(NA)	2.20(NA)	NA (NA)
		Meager lean coal	2	4	4.05 (7.51)	10.28 (10.53)	0.64 (1.15)	1.74 (3.74)
$PM_{0.1}$	Smokeless coal		1	0	0.40(NA)	NA	0.30(NA)	NA
	Combined smoky coal		7	16	0.71 (0.00)	1.63 (0.25)	0.40 (0.19)	1.33 (0.20)
		1/3 Coking coal	1	1	0.90(NA)	0.94(NA)	0.16(NA)	0.58(NA)
		Coking coal	3	11	0.74 (0.12)	2.18 (0.18)	0.62 (0.17)	1.50 (0.21)
		Gas fat coal	1	2	0.98(NA)	0.63 (0.92)	0.48(NA)	1.35(NA)
		Meager lean coal	2	2	0.49 (0.16)	1.12 (0.21)	0.29 (0.19)	1.07 (0.19)

Time is presented as "arithmetic mean (standard deviation)", PM_1 and $PM_{0.1}$ are presented as "geometric mean (geometric standard deviation)". Unit for time is "minute", unit for PM_1 is "mg/m^3", unit for $PM_{0.1}$ is "10^5 #/cm^3". Pre: 5 min before ignition; Ignition: from adding wood to adding coal; Coal: from adding coal to putting pot and water on fire; HP: high power stage, from putting pot and water on fire to water boils; LP: low power stage, water simmering. N: number of burns. NA: data not available.

the LP stage. The lowest concentration in the HP (53.10 mg/m^3) and LP stages (4.00 mg/m^3) both came from deposit 16; the highest $PM_{0.1}$ emission came from deposit 8 in both HP stage (4.00×10^5 #/cm^3) and LP stage (3.52×10^5 #/cm^3), the lowest $PM_{0.1}$ emissions were also from deposit 16 in both HP (0.94×10^5 #/cm^3) and LP stage (0.77×10^6 #/cm^3).

Despite being associated with the highest cancer rates in Xuanwei (Wong et al., 2019), samples from deposit 9 did not have the highest levels of $PM_{2.5}$ or $PM_{0.1}$ (Table S4; Table S8). These findings may imply that particle loading alone does not explain the excessive risk, and that there may be additional factors such as particle bonded chemicals or gaseous chemicals which are associated with the excessive lung cancer risks (Hu et al., 2014; Chen et al., 2005; Mumford et al., 1989; Mumford et al., 1990). Organic extracts of the emission particles from smoky coal combustion in Xuanwei have been shown to be the most active in tumor initiation (Mumford et al., 1990). A previous study (Lu et al., 2017) has reported that over 60% (number based) of the emitted particles from Xuanwei and Fuyuan coal samples were carbonaceous particles, whereas PAH bearing particles only contributed 4%–14% of the particles. Additionally, the carbonaceous and PAH-bearing particles are enriched in the size range below 0.56 μm, suggesting that the smaller particles should be given more attention when investigating the etiology of smoky coal related lung cancer.

In our previous exposure assessment study (Hu et al., 2014), we reported 24 h personal exposure to $PM_{2.5}$ as high as 277 $\mu g/m^3$ for smoky coal users. However, as this period includes times with relatively low exposures (e.g. overnight), peak exposures were omitted. In the current study, $PM_{2.5}$ levels of 12.4 mg/m^3 and 1.3 mg/m^3 for firepit and ventilated stove respectively were found during smoky coal combustion, which are substantially higher than the personal exposure levels. These extremely high "peak" exposures may provide insights in health risks not captured in time-weighted measurements. For example, it has previously been reported that the duration of cooking is strongly and positively associated with lung cancer incidence in Xuanwei and Fuyuan region (Lan et al., 2002; Hosgood et al., 2008).

Due to the long latency period of lung cancer, there are concerns that carcinogenic exposure from coal emissions over previous decades may have already changed. However, several lines of research have indicated that there have been no major compositional or quality change for coal being mined in the region for the past several decades as the research has compared Xuanwei coal from 1980s and coal collected more recently (Large et al., 2009; Downward, 2015). These findings are further

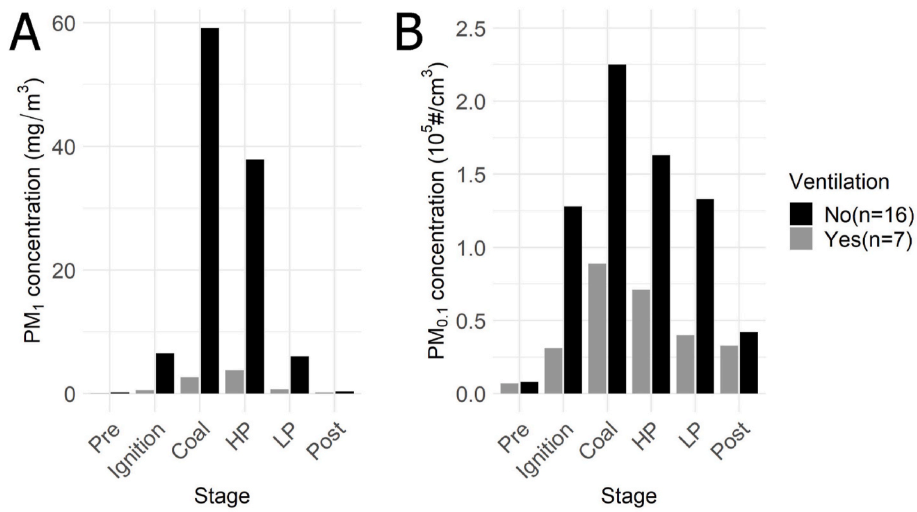

Fig. 3. Concentration of indoor PM_1 (A) and $PM_{0.1}$ (B) across different stages for water boiling tests in firepit with smoky coals. Pre, 5 min before fire was lit; ignition, from lighting the fire to adding coal; coal, from adding coal to putting water on fire; high power stage (HP), from putting water on fire to water was boiled; low power stage (LP), simmering stage; Post, up to 30 min after the simmering stage ended.

supported in current paper, where we report similar emissions between smoky coal from the 1980s and from 2010 to 2017 in terms of particle emission level and transition across the different burning stages, as well as the size-distribution of particle emissions.

3.3. Fuel efficiency and burning rates

Among the water boiling tests in the ventilated stove (Table 3), smokeless coal had the lowest burning rate (in g/min) and boiled water slower (in min/L) than smoky coal (10.58 g/min vs 16.78 g/min; 27 min/L vs 13 min/L). Within the smoky coal samples tested in the ventilated stove, gas fat coal had the highest burning rate (21.08 g/min) and fastest boiling time (6 min/L). In contrast, meager lean coal had the lowest burning rate (14.98 g/min), and coking coal had the slowest boiling time (18 min/L).

Within the smoky coal tests, ventilation increased the coal burning rate from 8.85 g/min to 16.78 g/min and decreased T.W_time to boil (21 min/L → 13 min/L). However, the associated changes differed per coal subtypes. For example, the burning rate increased by approximately 3.28-fold for 1/3 coking coal (16.78 g/min → 8.85 g/min) but only 1.42-fold for meager lean coal (14.98 g/min → 10.57 g/min). For T.W_time to boil, the effect of ventilation varied from 0.35-fold in 1/3 coking coal (26 min/L → 9 min/L) to 0.81-fold in coking coal (22 min/L → 18 min/L). Despite being associated with relatively lower PM concentrations, smokeless coal might be less efficient for cooking than smoky coal given the lower burning rate and prolonged T.W_time to boil. For the smoky coal tests, the improved burning rate and shortened T.W_time to boil from ventilation could be explained by the improved oxygen supply in ventilated stove, which had a dedicated chimney to vent smoke outside and a dedicated ash chamber which could also function as an extra tunnel for oxygen. Among the four smoky coal subtypes, gas fat coal had much better cooking efficiency as it had the highest burning rate, the shortest T.W_time to boil among the four smoky subtypes in both ventilated stove and firepit. Such observations could be supported by the fact that gas fat coal ranked the highest among the four smoky coal subtypes and contains relatively higher hydrocarbon content (Downward et al., 2014b; PengChen, 2000). Among the coking coal and gas fat coal samples tested in the firepit, we also observed geological variations regarding burning rate and T.W_time to boil. These might be explained by the heterogeneity in coal composition, including hydrocarbon contents and organic carbon as reported in our previous publication (Downward et al., 2014b). Such compositional differences exist in different coal types and deposits, and would result in heterogeneity in calorific values and in turn influence the performance for cooking and heating.

Apart from reducing indoor PM concentrations, ventilation also improved cooking efficiency, as observed by the shortened T.W_time to boil. Replacing traditional firepits with efficient cookstoves is part of the global movement to improve solid fuel related health while capacity for moving populations up the energy ladder is developed.

In addition to the variation associated with coal subtypes, deposit specific variation was also observed for smoky samples tested in the firepit (Table S9). Among the coking coal samples, the highest burning rate was found from deposit 7 (16.77 g/min) and the lowest from deposit 4 (4.93 g/min); the longest T.W_time to boil was found from deposit 4 (48 min/L) and shortest from deposit 10 (18 min/L). Among the gas fat coal samples, the sample from deposit 13 had lower burning rate and similar T.W_time to boil when compared to sample from deposit 12 (16.74 g/min vs 13.24 g/min; 14 min/L vs 15 min/L).

Table 3
Fuel efficiency metrics for burnings with water boiled.

Ventilation		N		Burning rate		T.W_time to boil	
		Yes	No	Yes	No	Yes	No
Smokeless coal		1	8.85 (1.62)	10.58 (NA)	NA	27 (NA)	NA
Smoky coal		7	6.00 (NA)	16.78 (1.23)	8.85 (1.62)	13 (2)	21 (1)
	1/3 Coking coal	1	7.72 (1.72)	19.68 (NA)	6.00 (NA)	9 (NA)	26 (NA)
	Coking coal	3	14.89 (1.18)	15.89 (1.31)	7.72 (1.72)	18 (2)	22 (1)
	Gas fat coal	1	10.57 (1.06)	21.08 (NA)	14.89 (1.18)	6 (NA)	14 (1)
	Meager lean coal	2	4	14.98 (1.01)	10.57 (1.06)	14 (1)	21 (1)

Burning rate, g/min; temperature and weight corrected time to boil (T.W_time to boil), time to boil 1 L of water with a temperature increase of 75 °C, min/L. Data is presented as "geometric mean (geometric standard deviation)". N: number of burns. NA: data not available.

3.4. Study strengths and limitations

The current study represents one of the most extensive experimental burning projects carried out in Xuanwei to date, with multiple water boiling tests conducted and a detailed examination of size-aggregated particle concentrations. By incorporating local co-operation, we were able to ensure that experiments were conducted in a consistent manner while still following the normal practices within the region. However, the relatively small sample size limits the overall statistical power. An additional limitation was that several burns, especially for smokeless coal, failed. Visual inspection of these coals revealed that some, especially smokeless coal, were small and fragile, easily breaking into smaller pieces, which were difficult to burn. A common practice can be to use mud or other packing material to make coal “cakes” in an effort to make a more burnable compound. Future studies will need to consider this practice.

4. Conclusion

In this extensive study of controlled burning experiments in Xuanwei, China, smoky coal was found to have caused higher indoor air pollution levels than smokeless coal samples, which varied by geological source. Stove improvements can not only reduce indoor concentrations of particulate material of all sizes from coal emission, but also promote cooking efficiency. The majority of particulates emitted during combustion of the Xuanwei and Fuyuan coal samples were $PM_{2.5}$ and smaller fractions. Understanding the impact of these smaller particles may help understanding the etiology of coal smoke induced lung cancer as well as promoting health improvements.

Author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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