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Physiochemical characteristics of indoor $PM_{2.5}$ with combustion of dried yak dung as biofuel in Tibetan Plateau, China

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Abstract

People inhabiting the Tibetan Plateau rely for survival on the yak, the region's native cattle. One of the important products of yak is dung, which has been served as cooking and heating fuels in the traditional Tibetan pastoralist society for several thousand years. The indoor air quality (IAQ) at eight residential homes with altitudes ranging from 3212 m to 4788 m was investigated in November 2012 to obtain a shot-term profile of emission from combustion of dried yak dung as biofuel in pastoral and agro-pastoral regions on the Tibetan Plateau. The indoor temperature, relative humidity, CO_2 and mass concentrations of $PM_{2.5}$ were monitored for around a 4-h period (5 kg dried fuel was consumed) at each site. Filter-based aerosol samples were also collected to characterize their elemental compositions, water-soluble ions, carbonaceous species and individual particle morphologies. The results showed that combustion of solid biomass fuel in cast-iron stove is the preliminary source of indoor particulate pollution. The average indoor and outdoor $PM_{2.5}$ mass concentrations were 330.7 and 29.1 $\mu g/m^3$, respectively. Individual particle analysis showed that most of the particles in smoke from dung burning were in the submicrometer size range. Regular and irregular organic balls and soot aggregates were the predominant species in the smoke (>90% in numbers). The data set in this study can provide significant basis for IAQ and epidemiology study on the Tibetan Plateau.

Keywords

Indoor air quality, Biofuel, Yak dung, Tibetan Plateau, Individual particle analysis

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Introduction

Air pollutants indoors affecting respiratory health can originate from a range of sources including the combustion of fuel for heating or cooking, the activities of building occupants and other biological sources, and emissions from building materials.[1–3] Infiltration from outside can also be another important source for some contaminants. Indoor air quality (IAQ) has become a matter of growing public concern because humans spend a significant proportion of their time indoors.[4] Consequently, particulate pollutants in the indoor residential microenvironment are the major contributor to personal non-occupational exposure.[5] Small particles

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with aerodynamic diameter less than 2.5 microns (termed as $PM_{2.5}$) are able to penetrate deeply into the lungs and appear to have the greatest adverse health effects.

Although exposure concentrations vary and depend on a number of factors as individuals' behaviour and activities, pollutant sources and geographical locations, the use of solid fuels for cooking and heating is likely to be the largest source of indoor air pollution.[6] On a global basis, it is estimated that approximately half of the world's domestic cooking rely on coal or unprocessed biofuels including wood, crop residues and animal dung.[7] Taking into account dung or crop residues, the human exposures resulting from biofuel combustion can exceed World Health Organization (WHO)-recommended values by factors of 10 to 20 or even more. Concentrations of particulates from the indoor combustion of biomass have been measured at levels of 10–50 times higher than those in urban areas of developed countries.[8]

On the Tibetan Plateau region, biomass (among those about 53% is yak dung) dominates the total energy consumption, accounting for nearly 70% in 2003.[9] Forest is distributed only in the Southeastern Tibetan Plateau, then yak dung serves nearly all heating and cooking needs in this region where people require ample fuel but where wood or coal are scarce or nonexistent. In vast pastoral and agro-pastoral regions on the Tibetan Plateau, yak dung was stored and air-dried outside the permanent dwellings in large heaps or nomadic tents in dung cakes for several thousand years. Yak dung was pounded and moulded into cakes, which were completely desiccated in the sun and symmetrically piled. Outside dwellings, yak dung was always piled intermittently to about 2 m wide, 3–4 m long and 1–2 m high. Each family used a heap every two months in the summer (about 0.133 m^3 per day), one every month in the winter (about 0.260 m^3 per day), or about 10 piles per year.[10] The Tibetan Plateau region was characterized by long duration of winter time with an average of 194 days in a year.[11] In winter, the weather conditions were severe on the Plateau and activities in nomadic tents were limited. Therefore, it is suitable to evaluate the IAQ in traditional Tibetan dwellings during winter time due to the most domestic fuel consumption and limited ventilation. However, limited data are available on the general understanding about present IAQ of residences in the whole Tibetan Plateau region.

The objectives of this study are to investigate IAQ parameters and to characterize the physicochemical properties of indoor aerosols at different residential homes with the combustion of dried yak dung as biofuel in vast pastoral and agro-pastoral regions on the Tibetan Plateau.

Methodology

Site selection

Eight residential homes were selected as sampling sites in the major pastoral and agro-pastoral regions in Tibetan Plateau, among those who had the same type of cast-iron multi-pot stove fuelled with dried yak dung (Figure 1). The three-room dwelling (each room about 7 m long by 5 m wide and 2.5 m high) in this study is the representative architecture style of permanent residences in the Tibetan Plateau region, of which the number and dimension of rooms and their functions were similar.

In most of those dwellings, the living-room or bedroom is also used for cooking, with a series of beds, sofas and cabinets lying with their back against the walls and surrounded the stove in the middle of the room. During winter time, the fire was started from the morning for a whole day's heating, cooking and tea or water boiling until naturally extinguished at night. In this study, the stoves were ignited indoors 2 h before sampling. The smoke from dung burning passed upward through the chimney flue to be vented outside the dwellings (Figure 2(a) and (b)). During the sampling periods, the windows were kept close and the residents were asked to live as usual (cooking and household task) except no smoke in the sampling rooms. All sites were at least 5 km away from the nearby roads. There is no nomadic tent being used in winter season.

Among the eight sampling sites, five (Amdo, Nagqu, Damxung, Dulan and Gangcha) were distributed in pastoral regions where animal husbandry (yaks, sheep, goats and horses) is booming in the area. The other three (two in Ngamring and one in Shigatse) were in agro-pastoral regions where the dominant crop was highland barley. Amdo, Nagqu and Damxung counties have a harsh, alpine climate, with long, very cold and dry winters. Dulan and Gangcha counties have a subalpine semi-arid climate, with long, very cold winters and warm summers. The Gangcha site is situated ~4 km to the west shore of the Qinghai Lake, a closed-basin saline lake and the largest water body in China. Vegetation around the Lake comprises steppe, desert shrub, alpine shrub and alpine meadow. Ngamring county and Shigatse city have a monsoon-influenced, alpine version of a humid continental climate, with frosty and very dry winters. The residential homes at the five sites in pastoral regions adopted dried yak dung as fuel, with dung cakes piled up (Figure 2c) or placed along the wall (Figure 2d) and air-dried outside the house, whereas biofuel used in residential homes at agro-pastoral regions as Shigatse site and Ngamring sites were dried yak dung mixed with highland barley straw and moulded to dung cake on the ground (Figure 2(e) and (f)).

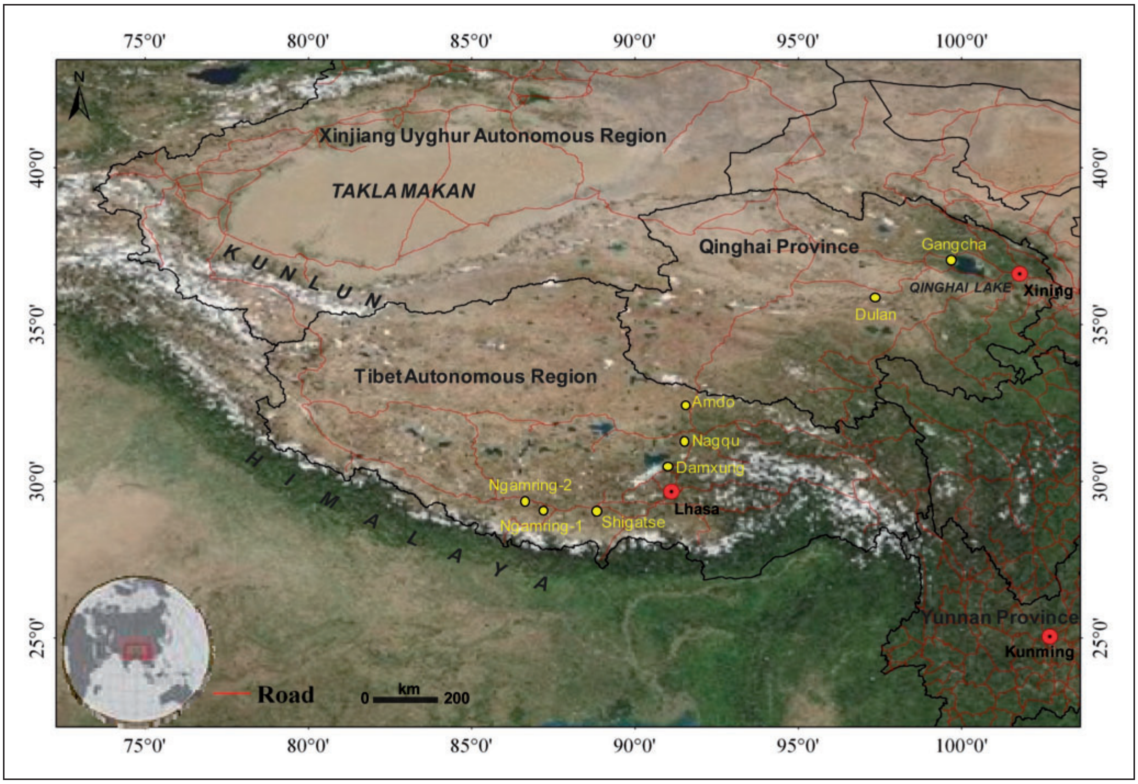

Figure 1. The locations of sampling sites at eight residential homes with combustion of dried yak dung as biofuel on the Tibetan Plateau.

Sampling methods

The sampling was performed from 17 November to 28 November 2012 (Table 1). At each site, the sampling duration was about 4 h during daytime while 5 kg dried fuel was added into the fire. Two portable Q-Trak Plus IAQ monitors (Model 7565, TSI Inc., Shoreview, MN, USA) were used to obtain the 1-min average CO_2 concentrations, air temperature (T) and relative humidity (RH) records indoors and outdoors, respectively. The monitor is able to detect CO_2 based on the mechanism of non-disperse infrared detection.[2] This monitor is also equipped with a thermistor and a thin film capacitive sensor for temperature and relative humidity measurements. Two Dust-Trak Handheld Aerosol Monitors (Model 8532, TSI Inc., Shoreview, MN, USA) with $PM_{2.5}$ inlets were placed at each indoor and outdoor location to measure $PM_{2.5}$ mass concentrations. Based on the method of light scattering, the monitor measured $PM_{2.5}$ at a 1-min interval with a flow-rate of $0.102\,m^3$/min. Before sampling, a separate calibration test was carried out to convert the results given by the Dust-Trak monitor into corresponding concentrations obtained by gravimetric methods. The temperature in the middle of the stove combustion chamber was measured using a thermocouple sensor during sampling.

Collocated with Dust-Trak monitoring, one Minivol portable sampler with $PM_{2.5}$ impactors (Airmetrics, Eugene, OR, USA) was used to collect 4-h airborne particulate matters onto 47 mm quartz microfiber filters (Whatman, Maidstone, Kent, UK) for subsequent chemical analysis.[12] Another sampler was used to collect 20-min particle samples onto Nuclepore polycarbonate filters (0.2 µm in porosity, Whatman International Ltd., Maidstone, UK) for individual particle analysis.[13] The operating flow rate of the samplers was $0.3\,m^3$/h. All monitors and samplers were placed 0.5 m away from the stove and 1 m above the ground. The quartz filters were preheated before sampling at 900°C for 3 h to remove the contaminants. After collection, the loaded filters were stored in a refrigerator at 4°C until analysed. All the instruments are listed in Table 2.

Sample analysis

The quartz filters were analysed gravimetrically for $PM_{2.5}$ mass concentrations by an electronic microbalance with 1 µg sensitivity (MC5, Sartorius, Göttingen, Germany) after 24-h equilibration at temperature between 20 and 23°C and RH between 35 and 45%.[12]

A punch of $0.5\,cm^2$ from each quartz filter was analysed for organic carbon (OC) and elemental carbon

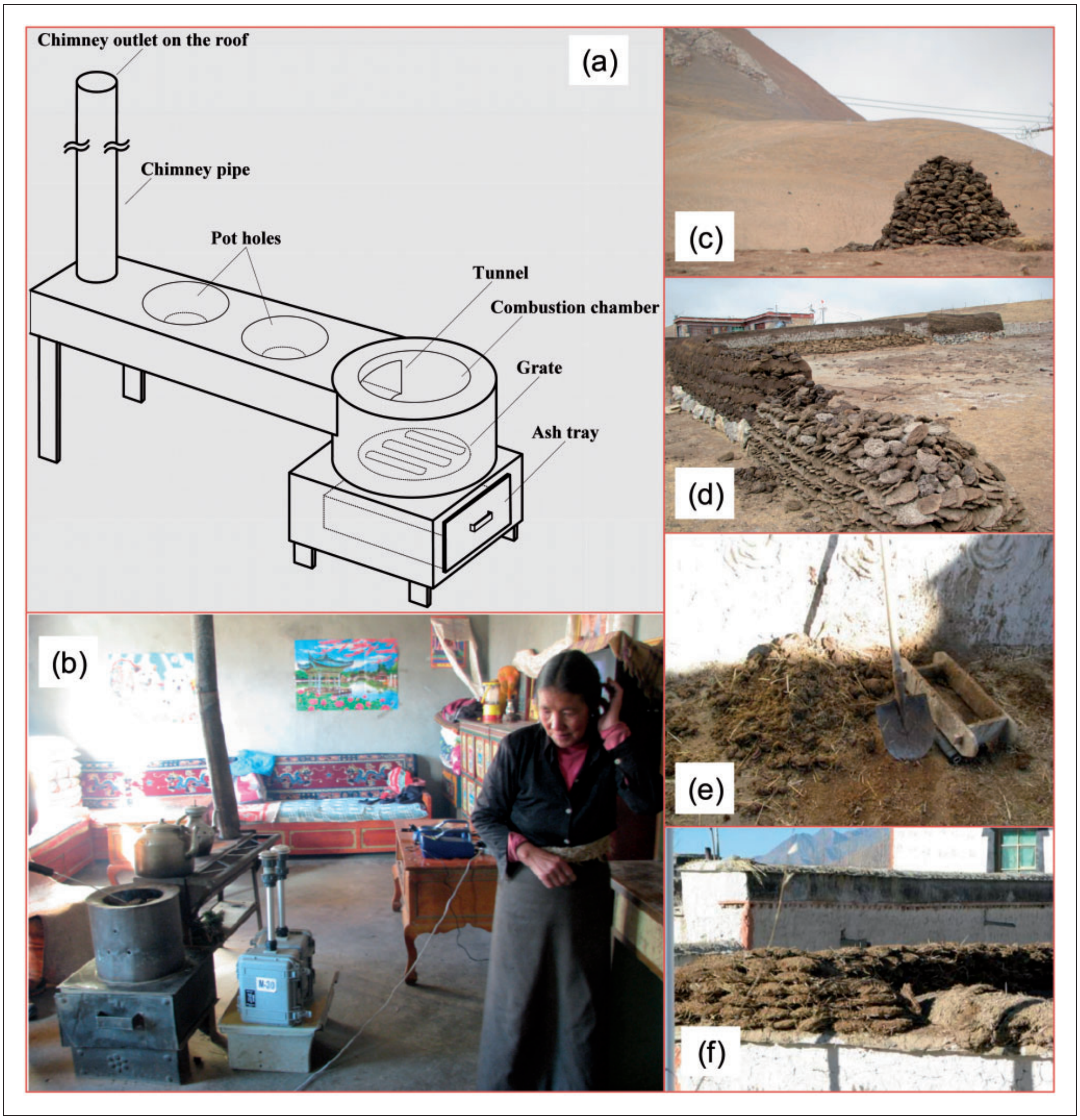

Figure 2. The schematic diagram of typical cast-iron multi-pot stove (a), its position at a residential home (b). In the pastoral regions, yak dung cakes were piled up (c) or placed along the wall (d) and air-dried outside the house. In the agro-pastoral regions, yak dung was mixed with highland barley straw and moulded (e) to dung cake (f).

(EC) concentrations by IMPROVE A, which is a thermal/optical protocol method using a DRI Model 2001 thermal/optical carbon analyser (Atmoslytic Inc., Calabasas, CA, USA). Quality assurance and quality control procedures were described by Cao et al.[12]

One-fourth of each quartz filter sample was used for gravimetric determinations of water-soluble ion mass concentrations.[13] Three anions (Cl^-, NO_3^- and SO_4^{2-}) and five cations (Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) were measured by a Dionex-600 Ion Chromatograph (Dionex Inc., Sunnyvale, CA, USA). Field blank levels were averaged, subtracted and standard deviations were propagated to the measurement precisions.[14] Minimum detection limits were 15 µg/L for Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+} and NO_3^-; 0.5 µg/L for Cl^-; and 20 µg/L for SO_4^{2-}. Ten per cent of the samples were submitted for replicate analyses.

Concentrations of elemental K, Ca, Ti, Cr, Mn, Fe, Ni, Zn, As, Br, Mo, Cd and Pb collected on the $PM_{2.5}$ quartz filters were determined using an Epsilon 5 ED-XRF (PANalytical B. V., the Netherlands).[15] The X-ray source is a side-window X-ray tube with a gadolinium anode, operated at an accelerating voltage of 25 to 100 kV and a current of 0.5 to 24 mA. A spectrum of X-ray counts versus photon energy was acquired during analysis, with the individual peak energies matching to

Table 1. Sampling information with the weather conditions, outdoor wind direction (WD), prevailing wind speed (WS, m/s), resident population and the temperature in the middle of the stove combustion chamber (stove T, °C).

Date	Province	County	Economics	Latitude	Longitude	Altitude (m)	Weather	WD	WS	Resident population	Stove T
2012.11.17	TAR	Nagqu	Pastoral	31°18.184′	91°48.289′	4552	Sunny	–	≤5.4	Two Tibetans	550
2012.11.18	TAR	Damxung	Pastoral	30°29.906′	91°06.481′	4283	Sunny	–	≤5.4	Five Tibetans	600
2012.11.20	TAR	Ngamring	Agro-pastoral	29°12.432′	87°21.143′	4354	Sunny	–	≤5.4	Four Tibetans	380
2012.11.21	TAR	Ngamring	Agro-pastoral	29°19.433′	87°12.217′	4345	Sunny	–	≤5.4	Three Tibetans	320
2012.11.22	TAR	Shigatse	Agro-pastoral	29°19.803′	89°23.538′	3806	Sunny	–	≤5.4	Five Tibetans	600
2012.11.24	TAR	Amdo	Pastoral	32°20.540′	91°43.092′	4788	Snowy	W	≤20.7	Five Tibetans	388
2012.11.27	Qinghai	Dulan	Pastoral	36°18.062′	97°06.045′	2766	Cloudy	W	≤7.9	Two Mongolians	630
2012.11.28	Qinghai	Gangcha	Pastoral	37°00.258′	99°48.050′	3212	Sunny	N	≤7.9	Three Tibetans	560

Note: "TAR" means Tibet Autonomous Region; "-" means no sustained wind.

specific elements, and peak areas corresponding to elemental concentrations.[15]

Small rectangular sections (approximately 10 × 10 mm) were cut from the polycarbonate filters and mounted on the holder with conductive adhesive carbon tape. Samples were coated under vacuum with platinum and analysed using a JEOL JSM-6460 LV SEM (Japan Electron Optics Laboratory Co. Ltd., Tokyo, JP) under 25 kV accelerating voltage at a work distance of 10 mm. Individual particles larger than 0.2 μm were selected for elemental analyses manually using NORAN SYSTEM SIX Si-Li EDX detector with an ultra thin window (Thermo Electron Corporation, Waltham, MA, USA).[13]

Microclimate

Indoor and outdoor air temperature, relative humidity and levels of CO_2 during the sampling at all sites are listed in Table 3. The fluctuations of indoor air temperature and relative humidity did not follow the trend of those outdoors due to the protection of thick walls and passive solar house. The average CO_2 concentrations inside the eight sampling homes ranged from 436 to 915 ppm with an average of 583 ppm. These CO_2 concentration levels did not exceed China's IAQ standard recommended value of 1000 ppm. On the other hand, the mean CO_2 concentrations outdoors ranged from 372 to 442 ppm with an average of 394 ppm. All the stoves did not extinguish during daytime for heating, cooking and tea or water boiling, resulting with an elevated CO_2 levels observed inside all the monitored homes than those outdoors.

Results and discussion

$PM_{2.5}$ mass concentration

Figure 3 illustrates the mean indoor and outdoor $PM_{2.5}$ mass concentrations at the eight sampling sites. The average $PM_{2.5}$ mass concentrations inside the residential homes ranged from 69.5 to 713.0 μg/m^3 with an average of 330.7 μg/m^3, whereas the average $PM_{2.5}$ mass concentrations outside the residential homes ranged from 9.8 to 61.0 μg/m^3 with an average of 29.1 μg/m^3. As shown in Figure 3, all the monitored residential homes had higher levels of indoor $PM_{2.5}$ mass concentrations than those outdoors. The pattern of indoor $PM_{2.5}$ mass concentrations varied from site to site during the sampling periods, with large peaks corresponding to the condition of cooking stove and refuelling schedules. Indoor airborne particles were mainly emitted through the narrow cracks of the iron-cast stove or from the pot holes during adding fuel. The indoor/outdoor (I/O) ratios of $PM_{2.5}$ mass

Table 2. Indoor and outdoor instruments used in the residential homes at the sampling sites.

Item	Instrument	Site	Analytical data
Microclimate	Q-Trak Plus IAQ monitors	Indoor and outdoor	1 record/min T, RH, and CO_2
$PM_{2.5}$ mass concentration	Dust-Trak Handheld Aerosol Monitor	Indoor and outdoor	1 record/min $PM_{2.5}$
Chemical analysis	MiniVol sampler, 47 mm quartz filter	Indoor and outdoor	IC, XRF, OC/EC
Single particle analysis	MiniVol sampler, 47 mm polycarbonate filter	Indoor and outdoor	SEM-EDX
Combustion condition	Thermocouple sensor	Indoor	Stove T

Table 3. Indoor and outdoor air temperature (T, °C), relative humidity (RH, %) and CO_2 concentrations (CO_2, ppm) in the residential homes at different sampling sites.

		T		RH		CO_2	
Site	n	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
Nagqu	247	8.5±2.1	14.8±4.6	36.9±3.6	13.5±1.4	915±139	403±37
Damxung	306	9.8±1.2	17.1±8.5	25.7±5.8	11.8±2.6	540±251	374±25
Ngamring-1	143	13.9±0.6	1.5±3.7	31.2±3.2	13.6±2.7	679±96	379±14
Ngamring-2	237	14.9±3.5	−1.9±1.3	15.4±6.1	21.4±2.5	436±29	408±5
Shigatse	242	12.8±0.7	−0.9±0.6	14.1±2.4	13.5±0.7	587±146	442±25
Amdo	226	9.2±3.1	8.7±0.6	13.5±1.9	6.8±0.6	443±57	372±9
Dulan	194	20.1±5.8	0.5±4.1	4.5±2.8	25.2±12.3	442±60	381±7
Gangcha	206	19.8±1.7	9.5±3.4	25.2±5.4	6.8±3.1	625±45	392±9

concentration ranged from 2.8 to 31.0, implying insufficient ventilation in dwellings all over the Tibetan Plateau.

Indoor $PM_{2.5}$ mass concentrations measured in this study were compared with those levels of particulate matters in residential homes using cattle, yak and sheep dung as fuel in the major pastoral regions in China (Table 4). Despite the variability in measurements and size differentiation of samples, the indoor winter $PM_{2.5}$ in this study are much higher than the value monitored in tent with flue and much lower than that in tent without flue, implying that the smoke exhaust efficiency and ventilation condition were the critical factors affecting the indoor particle level.

Material balance for indoor $PM_{2.5}$

The material balance approach has been used to estimate mass enclosure[20] and to evaluate the potential health hazards of indoor smoke from yak dung burning. By multiplying a characteristic coefficient, Fe is used to estimate the upper limit of geological material.[21] Previous study has shown that Fe accounts for 4% of Asian dust.[22] Thus, the concentration of geological material was considered as the mass concentrations of elemental Fe multiplied by 1/0.04. The amount of organic material was determined by multiplying the amount of OC by 1.6.[23,24]

The material balances for geological materials, organic materials, EC, ions (sum of Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) and uncertain materials (as the difference between the measured mass and the sum of the major components) for indoor $PM_{2.5}$ at all sampling sites are shown in Figure 4. Distinct differences were found in the chemical compositions of $PM_{2.5}$ emitted from combustion of dried yak dung and yak dung mixed with straw. In all of the residential homes in pastoral regions using dried yak dung as biofuel, carbonaceous species, especially OC, are the major component (70.0%, average) of indoor $PM_{2.5}$ mass. OC, including polycyclic aromatic hydrocarbons (PAHs) and other components with both primary and secondary origins, can have possible mutagenic and carcinogenic effects. On the other hand, in the three residential homes at Shigatse site, Ngamring-1 site and at Ngamring-2 site, geological materials dominated in indoor $PM_{2.5}$ with the weight percentage of 50.3%, 29.3% and 58.1%, respectively. In those agro-pastoral regions, yak dung was mixed with highland

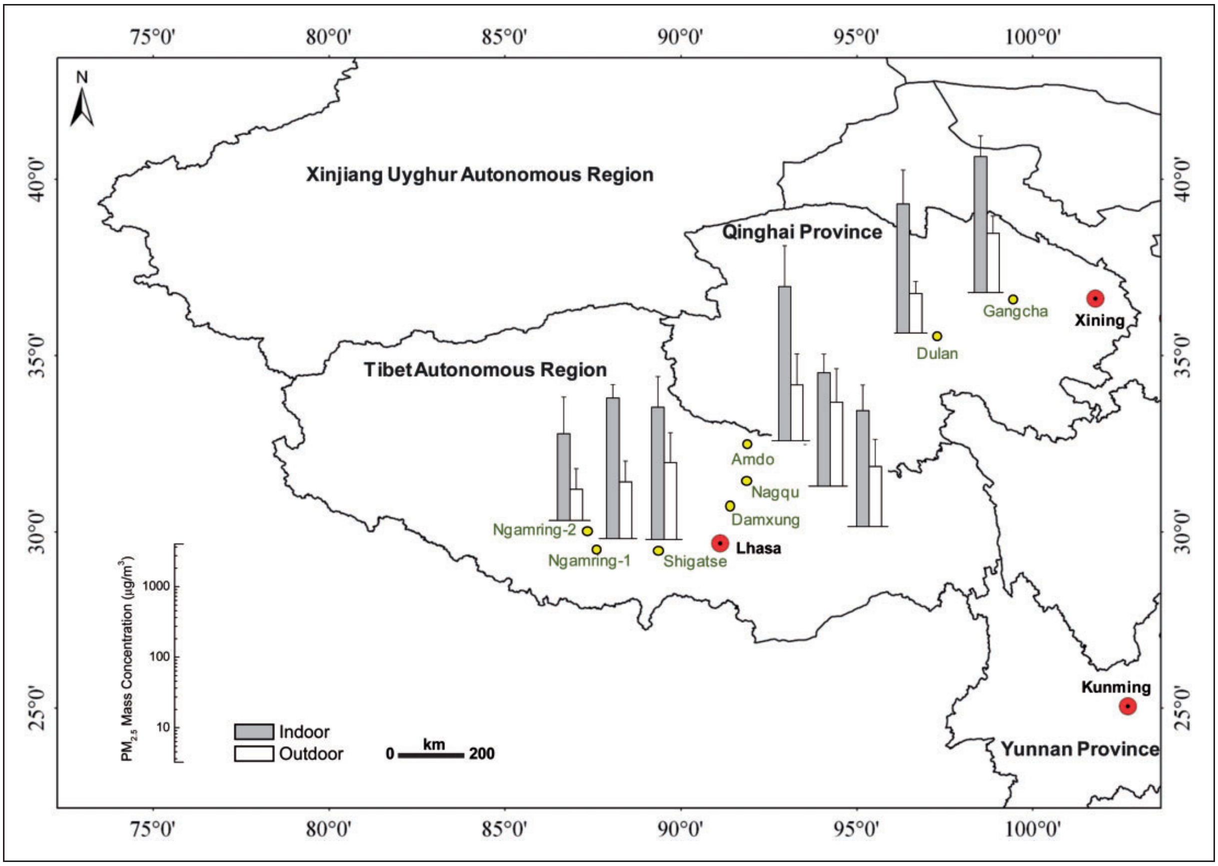

Figure 3. Indoor and outdoor average $PM_{2.5}$ mass concentrations and their standard deviations at residential homes in pastoral and agro-pastoral regions on the Tibetan Plateau.

Table 4. Indoor mass concentrations of particulate matters in the residential homes using animal dung as fuel at different sampling sites in pastoral regions in China.

Province	Item	Housing type	Fuel type	Avg. time	Sampling location	Avg.±SD (μg/m³)	Source
TAR	$PM_{2.5}$	Residences	Yak dung	Avg.4 h at 1-min interval	Stove with flue	359.5±306.5	This study
TAR	$PM_{2.5}$	Residences	Yak dung & straw		Stove with flue	291.5±200.6	This study
Qinghai	$PM_{2.5}$	Residences	Yak dung		Stove with flue	346.5±562.7	This study
TAR	$PM_{2.5}$	Herders' tent	Yak dung	Personnel sampler	Tent without flue	1272	[16]
					Tent with flue	97	
Qinghai	TSP	Residences	Yak dung	6×/day hourly filter weighing avg.	Kitchen & living room avg.	2204	[17]
	PM_{10}					1573	
Inner Mongolia	TSP	Residences	Cow & sheep dung	Morning & afternoon avg.		Win. 1939 Sum. 1061	[18]
	PM_{10}					Win. 1674 Sum. 830	
Gansu	PM_{10}	Residences	Yak dung	Morning & afternoon cooking time avg.	Hallway & bedroom avg.	3765±1504	[19]

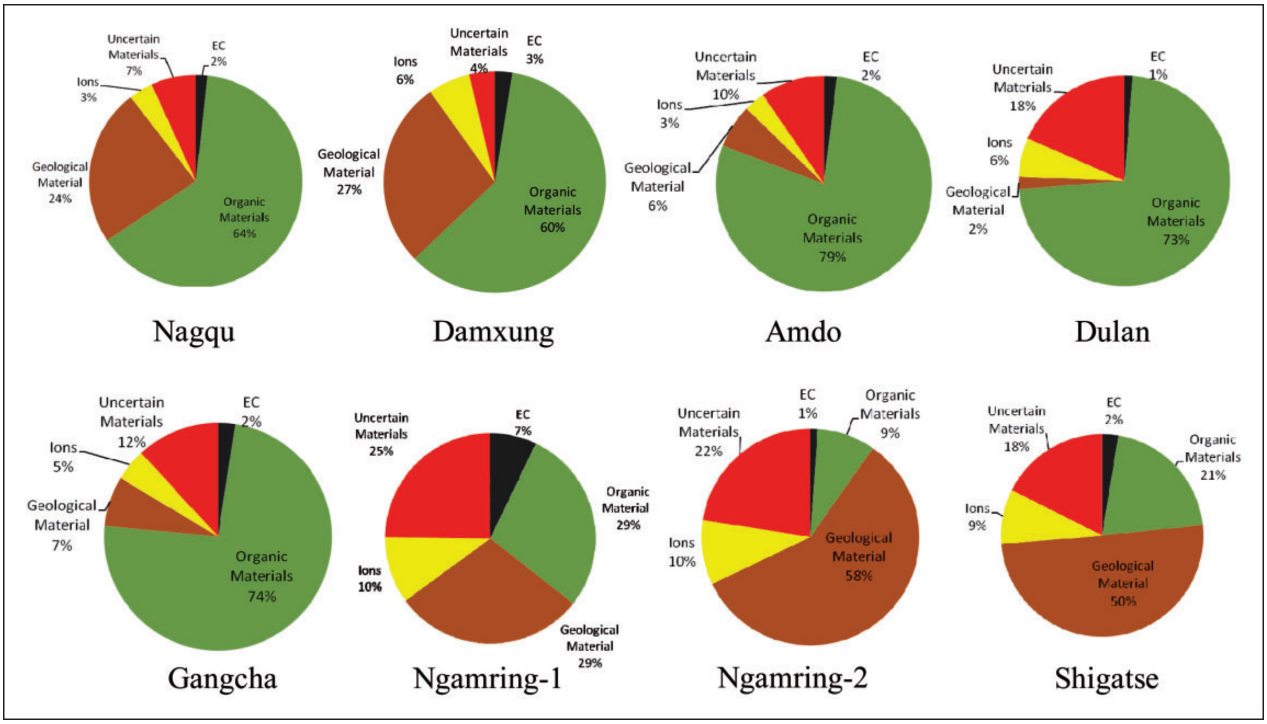

Figure 4. Material balance charts determined by chemical bulk analyses for $PM_{2.5}$ at residential homes in pastoral and agro-pastoral regions on the Tibetan Plateau.

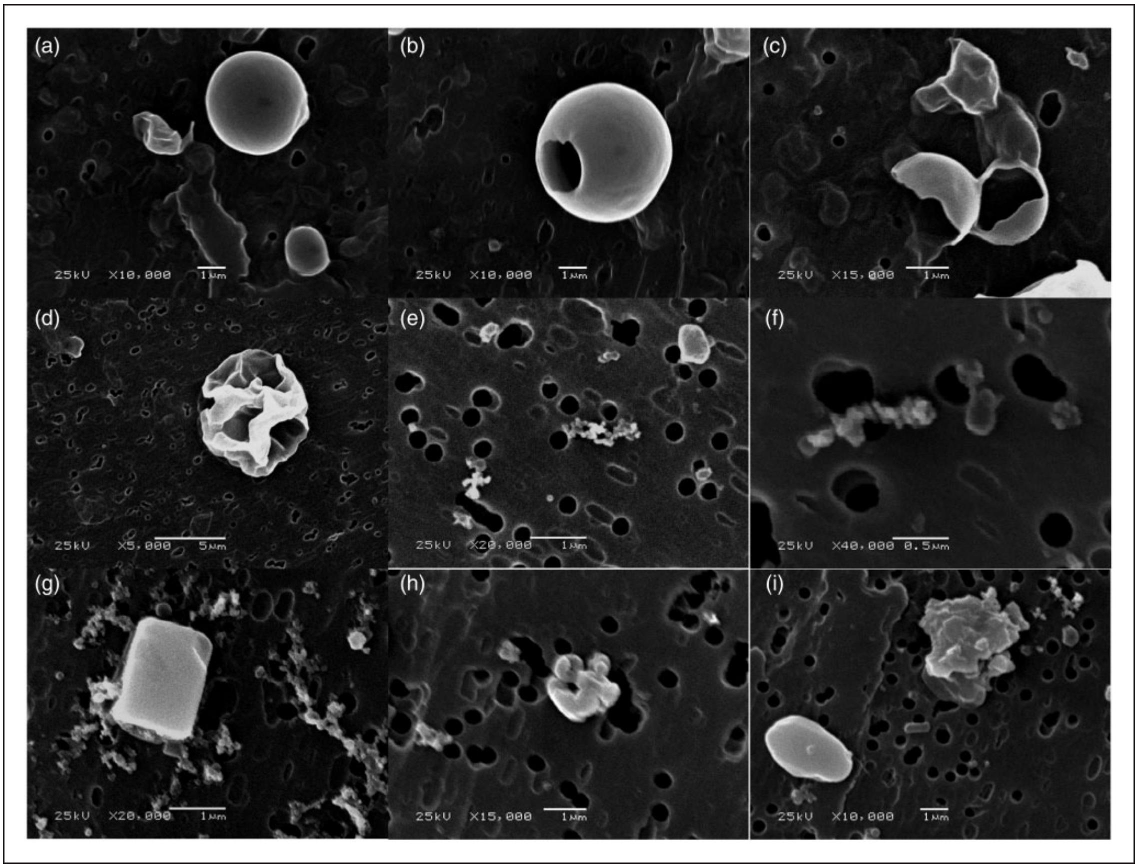

Figure 5. The morphologies of typical indoor particles: (a–d) organic balls; (e) and (f) soot aggregates; (g) potassium chloride particle; (h) calcium sulphate particle and (i) mineral dust.

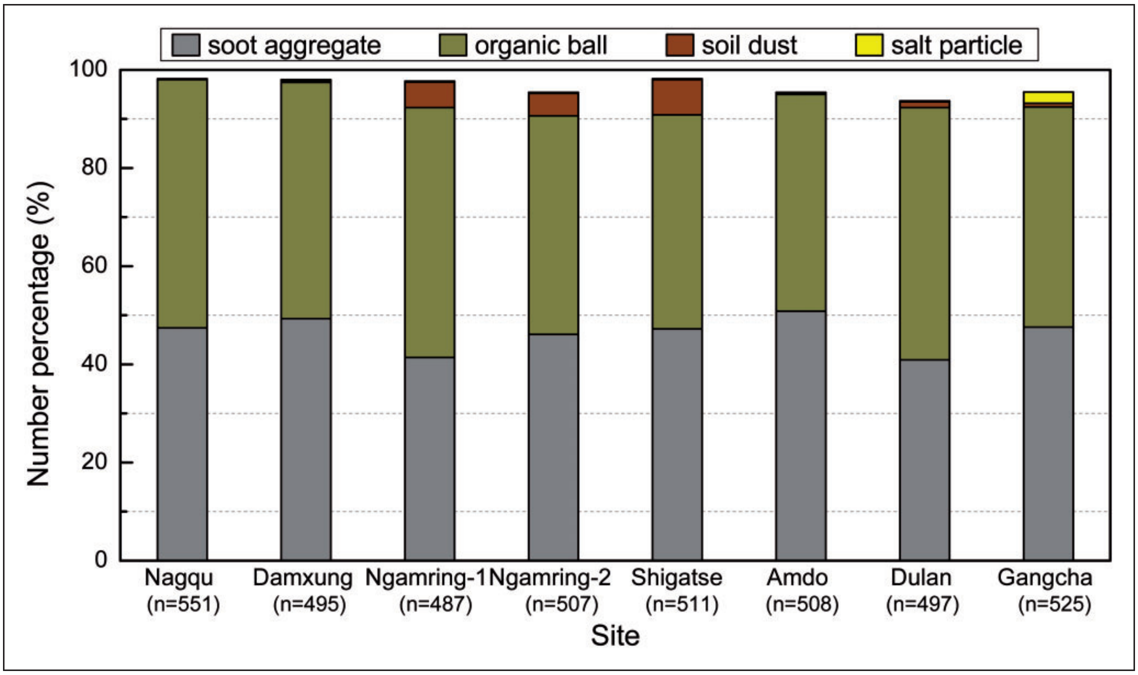

Figure 6. Classification and number percentage of each group in airborne particles collected in the residential homes at different sampling sites.

barley straw and moulded to dung cake during which crustal materials were brought into the solid biofuels. The water-soluble K^+ in airborne particulate matters was considered as biomass burning marker. Compared with the water-soluble ionic component concentrations measured during straw combustion events in western China during 2006–2007,[25,26] the K^+ concentrations in $PM_{2.5}$ from yak dung burning ($1.0 \pm 0.5\,\mu g/m^3$) were much lower than those from straw combustion ($14.3 \pm 3.6\,\mu g/m^3$ in $PM_{2.5}$ and $17.6 \pm 5.4\,\mu g/m^3$ in PM_{10}), probably due to the yak's digestion of potassium.

Morphology of indoor particles

The individual indoor $PM_{2.5}$ particles were grouped into four dominant types according to their morphologies and elemental compositions as organic ball, soot aggregate, salt particle and soil dust (Figure 5). The major elements of spherical organic balls detected by EDX were C and O with four typical morphologies, including sphere with smooth surface (Figure 5a), sphere with holes (Figure 5b), broken balls (Figure 5c) and shrunk balls (Figure 5d) with diameters from less than one micron to several microns. Nearly all soot particles were observed as chain-like aggregates (Figure 5(e) and (f)) with submicron diameters. Several salt particles, including potassium chloride particle (Figure 5g) and calcium sulphate particle (Figure 5h) with diameters around one micron were also found. Soil dusts were classified according to their elemental composition enriched by Al and Si, and minor Na, Mg, Ca, Mn and Fe.

The number percentages of each group in airborne particles collected in the residential homes at different sampling sites are illustrated in Figure 6. A distinct difference was found by comparing the number percentages and mass fractions of major constituents in $PM_{2.5}$ samples. As for the particle number concentrations, carbonaceous aerosols (almost equal number of organic balls and soot aggregates) dominated among the four particle types. Those two types of particles were added up to >90% (in numbers) in indoor $PM_{2.5}$ particles emitted from combustion of dried yak dung in stoves at all sampling sites. Soot aggregate was always the dominating component in numbers but EC occupied an obviously small percentage in the total $PM_{2.5}$ mass due to their small size and loose structure. Soil dust was composed of silicate, quartz, sulphate, carbonate and aluminosilicate from natural sources, which were non-spherical particles with large size and high density. Although the contributions of soil dusts to $PM_{2.5}$ mass were significant in samples collected at agro-pastoral regions, their number concentrations were only 7.2% at Shigatse site, 5.2% at Ngamring-1 site and 4.6% at Ngamring-2 site, respectively. Since aerosol adverse health effects are related to particle numbers,[27] results from bulk analysis techniques may have overestimated the human health risk associated with geological materials and underestimated the human health hazard associated with organic materials.

Conclusion

Although yak dung has been used as the most widespread solid fuel in traditional Tibetan household use for thousands of years, little is known about the indoor emission of dung combustion at a regional scale. In this study, the IAQ at eight residential homes was investigated to obtain a shot-term IAQ profile of emission from combustion of dried yak dung as biofuel in pastoral and agro-pastoral regions on the Tibetan Plateau. The indoor/outdoor ratios of $PM_{2.5}$ mass concentration ranged from 2.8 to 31.0 with an average of 14.1 at all sampled homes. The real-time results also showed that particles emitted through cracks of the stove or from the pot hole during refuelling were the main source of indoor $PM_{2.5}$. Moreover, high content of submicron size carbonaceous particles, including OC (in mass concentration) and EC (in number concentration) in indoor solid fuel smoke may contain and/or have adsorbed toxic and hazardous substances, that could cause adverse health effects on human health with limited ventilation.

Authors' contribution

TH and Junji Cao conceived the basis of the research and drafted the manuscript, SL, KH and XL commented and discussed on the research, SL and Ji Chen performed the sampling and experiments.

Declaration of conflicting interests

The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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