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Characteristics and potential health risk of rural Tibetans' exposure to polycyclic aromatic hydrocarbons during summer period

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ABSTRACT

Biomass fuels remain main energy sources in many remote rural regions, but potential health hazards from exposure to biomass combustion fumes have not been adequately assessed. Combustion of biomass fuels generates abundant polycyclic aromatic hydrocarbons (PAHs); hence residential exposure to PAHs can be used to evaluate the potential health risk to remote rural populations. The present study selected rural Tibetans to address the above-mentioned issue. Samples of indoor air and dust, human urine and local foods (Tsampa flour and buttered tea) were collected from five rural households in Langkazi County, an agricultural and pasturing region in Tibet of China in the summer season, which represented the best-case scenario as no heating was required. Residential exposure to PAHs by adults amounted to benzo[*a*]pyrene equivalent (BaP_{eq}) dosages of 110–760, 1.2–50 and 0.5–23 ng d^{-1} for ingestion, inhalation and dermal contact, respectively. Daily intakes of naphthalene, fluorene, phenanthrene and pyrene estimated from urinary monohydroxy PAH metabolites and from diet and inhalation exposure to PAHs were comparable (3.9, 1.9, 12 and 3.3 μg d^{-1} versus 9.5, 2.5, 5.1 and 1.1 μg d^{-1}), indicating the utility of external exposure in assessing daily intake of PAHs. The median incremental lifetime cancer risk was 32×10^{-6} (95% confidence interval: 0.7–73 $\times 10^{-6}$) for ingestion and 2.4×10^{-6} (95% confidence interval: 0.02–12 $\times 10^{-6}$) for inhalation and dermal contact combined, indicating moderate to slight potential cancer risk. Diet is the dominant source of health hazards for rural Tibetans, but cooking fumes also present a meaningful concern. The present study demonstrates that the pristine lifestyles of remote rural residents may be of global health concern, and merit further investigations.

1. Introduction

Biomass fuel is used by almost 3 billion people around the globe as the main source of domestic energy (IEA, 2014), due to limited access to newer types of energy (Guta, 2014). For example, solid biomass is believed to account for over 89% of the daily energy use in Ethiopia (Guta, 2012) and > 90% of domestic energy is solid biomass fuel such as fuel wood and dry dung cake in Tibet, China (CNBS, 2002). The relatively pristine lifestyle of rural households of Nepal and Tibet relies on biomass fuel for cooking and/or heating, largely within indoor settings (Gao et al., 2009). Biomass combustion for cooking and heating exposes people to increased concentrations of contaminants, such as particulate matter and gases, up to 10–20 times higher than outdoor concentrations (Hu et al., 2016). Many remote rural regions are distant from industries and traffic (Goldsmith, 2008; Yang et al., 2013), cooking and heating are expected to be the prevailing sources of indoor air pollution. Available literature however has largely overlooked the issue of health risk to remote rural populations from exposure to this form of environmental contamination.

Biomass fuel combustion has long been recognized to generate abundant polycyclic aromatic hydrocarbons (PAHs), which are genotoxic and carcinogenic to humans (Abdullahi et al., 2013; Li et al., 1994; Seea and Balasubramaniana, 2006), in addition to particulate matter, nitrogen oxides, carbon monoxide and sulfur dioxide (Ge et al., 2004). Numerous studies have identified PAHs as the dominant contributors to potential human health risk (Liu et al., 2014; Liu et al., 2012) in various environmental settings of China, e.g., urban areas (Yu et al., 2015), rural residency (Chen et al., 2015; Shen et al., 2014) and e-waste recycling zones (Luo et al., 2015) compared to other organic contaminants such as halogenated and organophosphate flame retardants, even in urban indoor environments. As one of the most remote regions, atmospheric pollutants in Tibet have been seriously

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investigated, but most studies mainly focused on toxic elements, $PM_{2.5}$ and CO (Kang et al., 2009; Li et al., 2012b). Therefore, it is reasonable to use residential exposure to PAHs in assessing the potential health risk to the population residing in remote rural regions.

To address the above-mentioned knowledge gap, we used rural Tibet Autonomous Region as a case study and assessed the potential health risks for rural Tibetans' exposure to PAHs generated from different sources, such as combustion of biomass fuels and foods. Besides inhalation and dermal contact, dietary intake was also examined in the present study for comparison as it has been demonstrated to be the dominant route of exposure to PAHs (Chen et al., 2015; Ma and Harrad, 2015; Suzuki and Yoshinaga, 2007). Samples of suspended particulate matter, gaseous phase, indoor dust, local foods (Tsampa flour and buttered tea; Supplementary data Text S1; "S" designates text, tables and figure in the Supplementary data thereafter) and residents' urine were collected. All samples except urine were analyzed for PAHs, while urine samples were processed for monohydroxy PAH metabolites (OH-PAHs), biomarkers for internal exposure to PAHs (Jongeneelen et al., 1985). Based on these measurements, we estimated the daily uptake of PAHs by rural Tibetans via inhalation, dermal contact and dietary consumption and assessed the potential health risk related to exposure scenarios. The results from the present study are expected to provide some baseline information for stimulating further investigations into the potential health risk of remote rural populations around the world, as these populations mostly lead a similar pristine lifestyle as the rural Tibetans.

2. Materials and methods

2.1. Materials

All calibration standards for target analytes, as well as internal and surrogate standards, and materials used in the present study are described in detail in Text S2.

2.2. Sample collection

Sampling was conducted on August 8–29, 2015 at rural Tibetan households located in Langkazi County (Fig. 1), an agricultural and pasturing region in Tibet Autonomous Region of China located at an altitude of 4400 m with a per capita gross domestic product of 12,000 RMB ($1750 USD). It well represents a remote rural population with a pristine lifestyle and with minimal influences of industrialization and urbanization. Target residences were selected randomly because the residences in this region have similar architecture styles and comparable air ventilation conditions (Fig. 1). During the sampling period, residents' daily schedules were not disrupted and yak dung was not used for heating in the evening but used only for cooking. Daily cooking and other activities were conducted inside the integrated room in each household. Most local residents used the same type of cast iron stove with chimney for cooking and heating. A second sampling was planned in the winter season, but was abandoned as we failed to secure willing participants. Because no heating is required in summer, exposure to cooking fume only represents the best-case scenario.

Also due to the difficulty in securing willing participants, only five households were selected for collection of air, indoor dust, food and urine samples. In non-smoking residences, gaseous and particle samples were collected with QCD-3000 air samplers (Yancheng Galaxy Science and Technology, China) in outdoor, kitchen, bedroom and living room (at ~1 m above the ground) at different time points, representative of different exposure scenarios. Polyurethane foam and 47 mm diameter glass microfiber filters (Whatman International, Maidstone, England) were used to collect gaseous and particle samples, respectively. Sampling was conducted daily for two days during 11:00–13:00 (cooking time) and 13:00–11:00 next day (non-cooking time). Tsampa flour and butter tea samples were collected from five households. Indoor dust samples were also collected from kitchens, bedrooms, living rooms and blankets. In addition, urine samples were collected during 7:00–8:00 and 18:00–19:00 from 18 local residents. Demographic groups are divided into children (1–11 years old), adolescents (12–17 years old) and adults (> 17 years old). Detailed information is presented in Table S1. Overall, 30 air and particle samples (300 and 3300 L during cooking and non-cooking periods, respectively), 15 indoor dust (from 1 m^2), 10 food (30 g for Tsampa flour and 20 mL for Butter tea) and 36 urine (3 mL) samples were collected.

2.3. Sample extraction and instrumental analysis

The detailed extraction and cleanup procedures for food and air samples can be found elsewhere (Wu et al., 2015). All samples were spiked with the surrogate standards before extraction. Gaseous, Tsampa flour and indoor dust samples were Soxhlet extracted for 48 h with a mixed solvent of hexane, dichloromethane and acetone (2:2:1 in volume). Particle samples were sonicated with 15 mL of the mixed solvent mentioned above. Butter tea samples were shaken with 20 mL of the mixed solvent. Each extract was concentrated and solvent-exchanged to hexane, and concentrated again to 0.1 mL. Before instrumental analysis, each extract was spiked with the internal standards. The final concentration of the surrogate and internal standards was 1.0 μg mL^{-1}. The correlation coefficient of calibration curve were > 0.995. Bio-Beads SX-3 was used to purify food samples for removing lipid before being concentrated to 1.0 mL.

A Shimadzu gas chromatograph–mass spectrometer (GCMS-2010 Plus) was used to analyze all samples. An HP-5MS capillary column (30 m × 0.25 mm with a 0.25 μm film thickness) was used for chromatographic separation. The column temperature was set initially at 60 °C (held for 1 min) and elevated to 250 °C at 20 °C min^{-1}, ramped to 280 °C at 5 °C min^{-1} (held for 2 min), and further increased to 300 °C at 20 °C min^{-1} (held for 15 min). All samples were injected (1 μL each) automatically in a temperature vaporizer programmed with an original temperature of 60 °C and elevated to 250 °C at 400 °C min^{-1}. Ultrahigh-purity helium was used as the carrier gas with a flow rate of 1 mL min^{-1}. The ion source temperature was 250 °C.

Urine samples were prepared following a previously reported procedure (Fan et al., 2012) with minor modifications. Briefly, 2 mL of urine sample was spiked with 10 μL of the surrogate standards (50 ng of 1-hydroxynaphthalene-d_7; 10 ng of 1-hydroxyphenanthrene-d_9 and 10 ng of 1-hydroxypyrene-d_9), and then 1 mL ammonium acetate buffer (1 M, pH = 6.5) and 50 μL β-glucuronidase (200 units mL^{-1}) for incubation at 37 °C overnight. Each hydrolyzed sample was subject to solid phase extraction with a C18 cartridge, which was preprocessed with 5 mL of methanol and water and 10 mL of monopotassium phosphate buffer (25.0 μmol L^{-1}). The flow rate was maintained at < 1 mL min^{-1}. The loaded cartridge was washed with 4 mL of monopotassium phosphate buffer (25.0 μmol L^{-1}) and 5 mL of water to remove matrix interferences. The cartridge was dried completely and eluted with 10 mL of methanol. The extract containing the target analytes was concentrated to 500 μL under a mild stream of nitrogen. The concentrated extract was filtered through a nylon filter (0.22 μm) and spiked with the internal standards before instrumental analysis.

All urine sample extracts were analyzed with a Shimadzu DGU-30A LC interfaced with an AB SCIEX TRIPLE QUAD™ 5500 MS system. An Agilent Zorbax Eclipse Plus C18 column (2.1 × 100 mm, 1.8 μm) was used for chromatographic separation. The mobile phases were 40% methanol in water (v/v; solvent A) and methanol (solvent B). The gradient elution program was as follows: 0–3 min, 5% solvent B; 3–5 min, 30% solvent B; 5–10 min, 30% solvent B; 10–15 min, 40% solvent B; 15–16 min, 80% solvent B; 16–20 min, 70% solvent B; 20–21 min, 5% solvent B. Each extract of 5 μL was injected by an autosampler into the chromatographic column at a temperature of 40 °C and a flow rate of 0.27 mL min^{-1}. Mass spectra were acquired in the multiple reaction-monitoring mode with an ion spray voltage of 4500 V

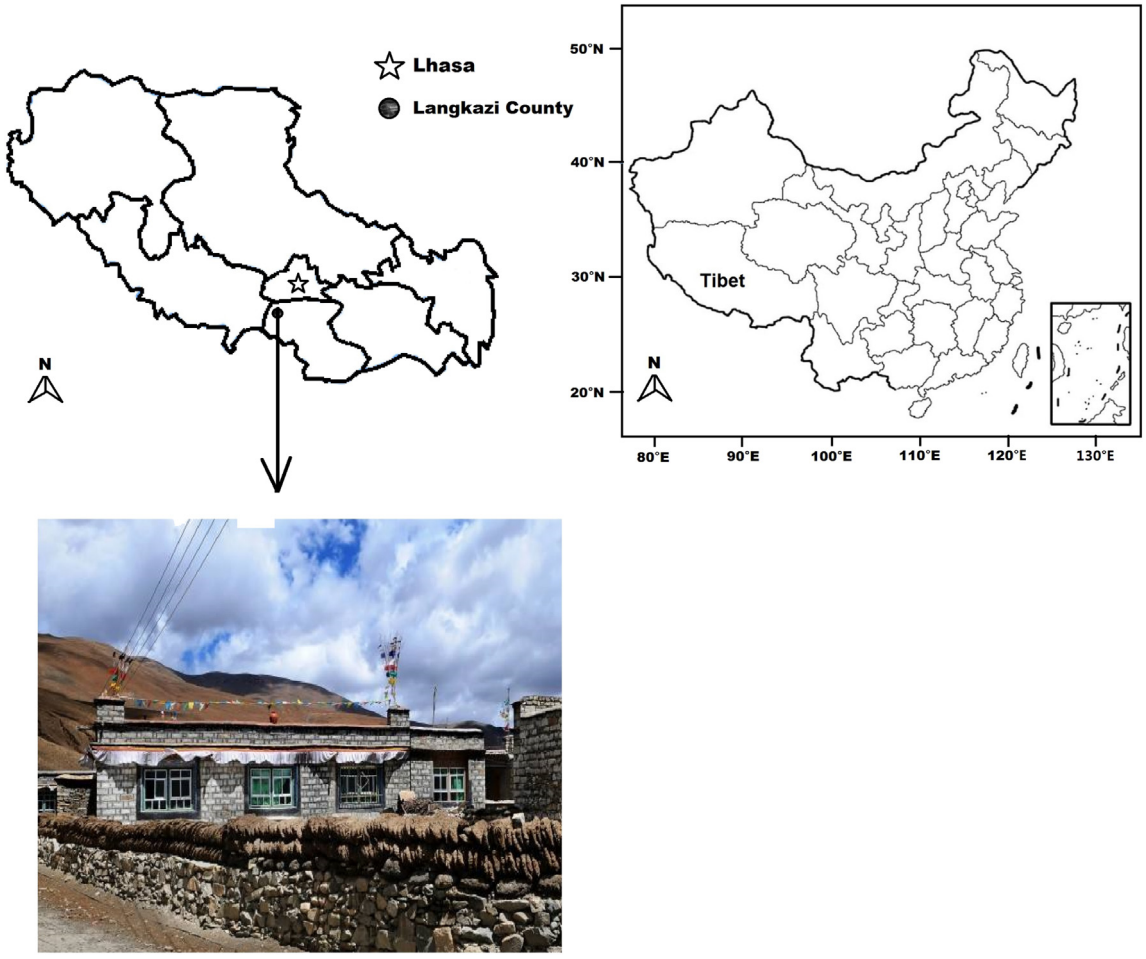

Fig. 1. A map showing the locality of the sampling sites in Langkazi County, Tibet Autonomous Region, China.

and an ion source temperature of 450 °C.

2.4. Quality assurance and quality control

In every batch of 20 samples, a single procedure blank sample was analyzed and the concentrations of detected target analytes were subtracted from those in the samples. The recoveries of the surrogate standards, naphthalene-d_8, acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12} and perylene-d_{12}, were 75 ± 16%, 91 ± 17%, 96 ± 13%, 111 ± 20%, 116 ± 28% and 92 ± 17% in suspended particulate samples, 42 ± 14%, 60 ± 19%, 75 ± 26%, 92 ± 25% and 67 ± 20% in gaseous samples, 59 ± 12%, 66 ± 9%, 87 ± 9%, 87 ± 17% and 50 ± 12% in indoor dust samples and 40 ± 10%, 56 ± 8%, 65 ± 10%, 66 ± 11% and 55 ± 21% in food samples. The recoveries of 1-OHN-d_7, 1-OHPhe-d_9 and 1-ONP-d_9, were 91 ± 16%, 66 ± 9% and 52 ± 14% in urine samples. The lowest calibration concentration divided by the actual sample volume was defined as the reporting limit for a target compound. The reporting limit was 0.15 ng m^{-3} for air samples, 1 ng m^{-2} for indoor dust samples, 0.03 ng g^{-1} for food samples and 0.02 ng mL^{-1} for urine samples. The blank concentrations of individual PAHs were 0.17–1.05 ng m^{-3} for air samples, 0.3–3.6 ng m^{-2} for indoor dust samples, 0.4–1.8 ng g^{-1} for food samples and below the reporting limit (0.02 ng mL^{-1}) for urine samples, which were all lower than the PAH concentrations in field samples. The concentrations of PAHs and OH-PAHs in all field samples were corrected by those in the corresponding procedural blanks, but not corrected for the surrogate standard recoveries.

2.5. Data analysis

Daily intakes of PAHs via inhalation (DI_{ip} represents for particle-bound PAHs; DI_{ig} represents for gaseous PAHs), dietary intake (DI_{food}), dermal contact (DI_{dp} represents for particle-bound PAHs and DI_{dg} represents for gaseous PAHs) and cancer risk (Risk) were estimated by (Shi and Zhao, 2014; USEPA; Weschler and Nazaroff, 2014).

$$DI_{ip} = \sum (C_{ip} \times TEF_{PAH}) \times IR_{in} \times t_{event} \quad (1)$$

$$DI_{ig} = \sum (C_{g} \times TEF_{PAH}) \times IR_{in} \times t_{event} \quad (2)$$

$$DI_{food} = \sum (C_{food} \times TEF_{PAH}) \times IR_{food} \quad (3)$$

$$DI_{dp} = \sum (C_{dp} \times TEF_{PAH}) \times k_{pp} \times SA \times f_{sa} \times t_{event} \quad (4)$$

$$DI_{dg} = \sum (C_{dg} \times TEF_{PAH}) \times k_{pg} \times SA \times f_{sa} \times t_{event} \quad (5)$$

$$Risk = \sum \frac{DIi \times CSFi \times ED \times EF}{BW \times AT} \quad (6)$$

where TEF_{PAH} is the toxicity equivalency factor of PAH based on benzo[*a*]pyrene; the benzo[*a*]pyrene equivalent (BaP_{eq}) concentration of an individual PAH compound equals the PAH concentration multiplying with its corresponding TEF; C_{ip} is the BaP_{eq} concentration of particle-bound PAHs (ng m^{-3}) via inhalation; C_{dp} is the BaP_{eq} concentration of particle-bound PAHs (ng m^{-3}) via dermal contact; C_g is the the BaP_{eq} concentration of gaseous PAHs (ng m^{-3}); C_{food} is the BaP_{eq} concentration of PAHs in Tsampa flour (ng g^{-1}); IR is the rate of inhalation (IR_{in}, m^3 h^{-1}) or daily dietary intake (IR_{food}, g d^{-1}); t_{event} is the indoor

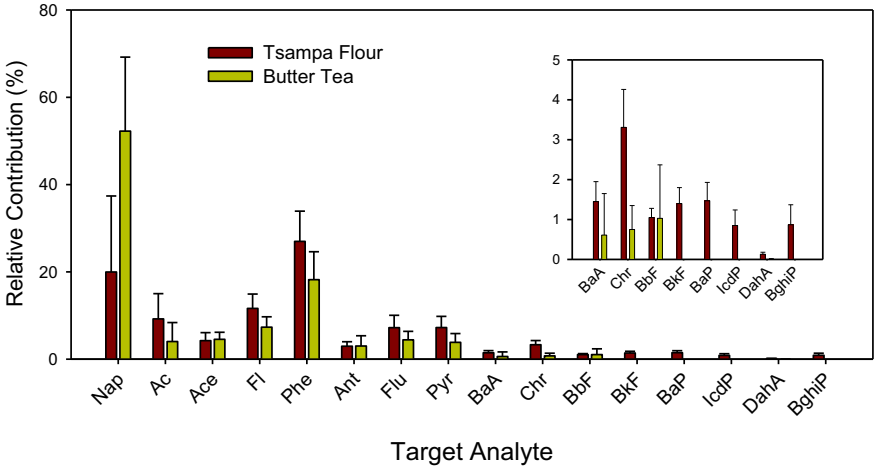

Fig. 2. Mean relative contributions of PAHs in Tsampa flour and butter tea samples from five households (n = 10). Error bars represent standard errors (three replicates for each sample).

exposure time duration in a day (h d^{-1}), which was taken as 16 h in the present study; f_{sa} is the exposed dermal fraction; SA is the skin surface area (m^2); k_{p_p} is the transdermal permeability coefficient of particle-bound compounds (m h^{-1}); k_{p_g} is the transdermal permeability coefficient of gaseous compounds (m h^{-1}); CSF_i is the cancer slope factor of BaP for exposure route i (kg d mg^{-1}) and is age-dependent as specified by average body weight (Chen and Liao, 2006); ED is the lifetime exposure duration (yr); EF is the exposure frequency, i.e., the number of days with exposure per year (d yr^{-1}); BW is the average body weight (kg) and AT is the average time for carcinogenic effects (d). Detailed values for the above-mentioned parameters are presented in Table 2. Some studies indicated that human lung, skin and bladder cancers have been associated with exposure to PAHs depending on the exposure scenarios (Hoseini et al., 2016). Consequently the use of different CSFs depending on different exposure routes is reasonable (Chen and Liao, 2006).

In the present study, Monte Carlo simulation was conducted in Crystal Ball software (Version 2000.2, Decisioneering, Denver, CO, USA), each with 10,000 iterations to generate 2.5 and 97.5 percentiles at a 95% confidence interval (CI) for all calculations presented.

3. Results and discussion

3.1. Potential sources of PAHs in indoor environment

Yak dung was the most common cooking fuel in all households and it generated a large quantity of cooking fumes which polluted each indoor microenvironment. During cooking time, the concentrations (arithmetic mean ± SD) of PAHs (Table S2) were slightly different in bedrooms (particulate: 30 ± 6.3 ng m^{-3}; gaseous: 2750 ± 2400 ng m^{-3}), living rooms (particulate: 11 ± 3.6 ng m^{-3}; gaseous: 3300 ± 660 ng m^{-3}) and kitchens (particulate: 33 ± 31 ng m^{-3}; gaseous: 4420 ± 2280 ng m^{-3}). These values were much greater than those during non-cooking periods (Tables S2 and S3). Besides, concentrations of benzo[*a*]pyrene were 4.1 ± 4.1, 0.5 ± 1.0 and 5.1 ± 7.0 ng m^{-3} in bedrooms, living rooms and kitchens, respectively, during cooking time (Tables S2 and S3), compared to the national ambient air quality standards of 2.5 ng m^{-3} in China (MOH, 2012). Apparently the concentrations of PAHs were rather variable, because they varied with the amount of yak dung used in each household. It nevertheless indicates that cooking fumes should be a great health concern for the local residents staying indoors for a long time every day. High molecular-weight (4–6 rings) PAHs contributed to 53–70% of the total particulate PAHs, similar to the finding of a previous study on indoor air pollution in Tibet (66%) (Gu, 2009). The results clearly showed that biomass fuel combustion was a main source of PAHs in cooking fumes. Combustion fumes from cooking deteriorate indoor air quality in summer, which is aggravated by poor ventilation typically encountered in remote rural areas due to unfavorable climatic conditions. Poor ventilation also allows combustion-derived particles to stay indoors for long duration and to infiltrate living spaces easily (Dasgupta et al., 2009; Wan et al., 2011).

Due to poor ventilation, large amounts of indoor dust originate from combustion-derived particles. Indoor dust can be used as an indicator of environmental pollution, as PAHs are more likely to accumulate in dust particles (Yang et al., 2015). In dust samples from different indoor locations, the concentrations of PAHs ranged from 80 to 4660 ng m^{-2} and high molecular-weight PAHs constituted 28–73% of total PAHs (Table S4). Specifically the relative abundances of high molecular-weight PAHs in blankets (32–52%) and bedrooms (28–63%) were generally lower than those in living rooms (54–73%) and kitchens (54–66%). Because living rooms and kitchens are usually the places for housing stoves, the concentrations of PAHs were higher in blankets than in other locations. Because the distance from the stove was different for each sampling site, the relative abundances of high molecular-weight PAHs may have varied, reflecting such difference. In addition, it was difficult to ensure the sampling conditions were similar, resulting in the wide variability of the results. A previous survey in a residential complex surrounded with contaminated soil indicated that the daily intake of dust was 0.56 mg for adults and 50–100 mg for children assuming residents spent 16 h day^{-1} indoors (Hawley, 1985). The Tibetans generally spend 18 ± 5 h at home in rural regions because of the cold high altitude climate. Even in some pasturing regions, they may stay indoors all day during cold weather (Gu, 2009). Thus indoor dust is a potential health risk to the Tibetans, particularly children, residing in remote regions.

The local residents in Langkazi County commonly consumed Tsampa flour and butter tea as staple foods every day. The profiles of PAHs in the samples of Tsampa flour and butter tea show slightly different patterns (Fig. 2). For example, although low to moderate molecular-weight PAHs, i.e., from naphthalene to pyrene, dominated both Tsampa flour and butter tea, the most abundant compound was phenanthrene for Tsampa flour and naphthalene for butter tea. In addition, high molecular-weight PAHs were rarely detected in both food types. These results indicated that the occurrence of PAHs in foods was controlled by the cooking type (e.g., fried, grilled, or roasted, etc.), temperature and duration and fuel, as well as the fat content in foods (Gungormus et al., 2014).

The concentrations of PAHs and benzo[*a*]pyrene were 80 ± 30 ng g^{-1} and 1.1 ± 0.4 ng g^{-1}, respectively, in Tsampa flour

Table 1
Daily exposure (benzo[*a*]pyrene equivalent dosage; ng/d) through inhalation, dermal contact and Tsampa flour consumption by adults.[a]

	DI_{ip}[b]	DI_{dp}[c] f_{sa}[g]	DI_{ig}[d]	DI_{dg}[e] f_{sa}[g]	DI_{food}[f]
Bedroom	7.1 (0.9–15)[h]	0.01 (< 0.01–0.04)	8.6 (1.2–21)	5.6 (0.5–16)	348 (110–764)
Living room	0.9 (0.05–2.5)	< 0.01	11 (4.1–20)	7.4 (0.5–22)	348 (110–764)
Kitchen	9.4 (0.7–23)	0.02 (< 0.01–0.05)	12 (2.3–27)	7.8 (0.3–23)	348 (110–764)

[a] The daily exposure duration is 16 h.
[b] DI_{ip} is the daily inhalation exposure to particle-bound PAHs.
[c] DI_{dp} is the daily dermal exposure to particle-bound PAHs.
[d] DI_{ig} is the daily inhalation exposure to atmospheric PAHs.
[e] DI_{dg} is the daily dermal exposure to atmospheric PAHs.
[f] DI_{food} is the daily dietary intake of Tsampa flour, which was assumed to be 200 ± 100 g (Gao et al., 2017).
[g] f_{sa} is the fraction of dermal adsorption, and 25% was used as the fraction of dermal uncovered by thick clothes normally in the present study.
[h] A (B–C) represents a mean and 95% confidence interval.

samples (Table S5). It was noteworthy that the concentrations of benzo[*a*]pyrene were similar to those in barbecued food (0.9–1.1 ng g^{-1}) (Zhang et al., 2014). Thus Tsampa flour may have adverse health impacts on local residents ingesting the food daily. Consequently the health risk from Tsampa flour was as a main research focus in food samples. As for butter tea samples, the concentrations of PAHs were 4.0 ± 3.0 μg L^{-1} (Table S5), far less than those in sixteen commercial brands of milk (220–610 μg L^{-1}) (Chung et al., 2010), but comparable to those in other five commercial brands of milk (mean: 7.9 μg L^{-1}) (Yu et al., 2011). The carcinogenic PAH compounds, such as benzo[*b*]fluoranthene (0.02 ± 0.04 μg L^{-1}), were detected in individual butter tea samples. As one of the most favorite beverages, butter tea is the daily necessity for local residents, with an estimated daily intake of 2270 mL in Lhasa (Yang et al., 2014). Therefore drinking butter tea frequently also poses potential health hazards to local residents.

3.2. Daily exposure pathways

Daily intake doses of PAHs through inhalation, dermal contact and diet for the local residents are presented in Table 1. Dietary intake of PAHs through ingesting Tsampa flour in the amount of 200 ± 100 g was 110–760 ng d^{-1} of BaP_{eq}, which was far more than inhalation (1.2–50 ng d^{-1} of BaP_{eq}) and dermal contact (0.5–23 ng d^{-1} of BaP_{eq}) exposures to PAHs. Obviously diet was the main source of human exposure to PAHs. Different food categories contributed to different levels of PAH exposure. For example, the BaP_{eq} concentration of dietary exposure was 170 ng d^{-1} and 570 ng d^{-1} for adults consuming mainly rice, fruits and vegetables in Korea (Yoon et al., 2007) and wheat, pork and fish in Taiwan (Xia et al., 2010), respectively. Besides Tsampa flour, the Tibetans also consumed other foods, such as meat and rice. So the daily intake of PAHs through diet was likely underestimated, which merits further investigation. Daily exposure to gaseous (1.5–50 ng d^{-1} of BaP_{eq}) and particulate (0.05–23 ng d^{-1} of BaP_{eq}) PAHs through inhalation and dermal contact contributed to the total daily intake equally, i.e., exposure to gaseous pollutants should also be considered in assessing human health risk. The BaP_{eq} concentrations of gaseous PAHs through dermal contact (0.3–23 ng d^{-1}) were comparable to those of gaseous PAHs from inhalation (1.2–27 ng d^{-1}). It was noteworthy that the BaP_{eq} concentrations through dermal contact with indoor cooking fumes were similar to those with barbecue fumes (0.03–13 ng day^{-1}) (Wu et al., 2015). Consequently, when the residents spend a long time for cooking in summer, dermal contact with fumes from biomass fuels burning cannot be overlooked.

3.3. Association between OH-PAHs and daily intake of PAHs

The concentrations of Σ_{13}OH-PAH ranged from 0.8 to 19 μg L^{-1} in urine samples, with a mean value of 6.0 μg L^{-1}. Sample collection time was important; the concentrations of Σ_{13}OH-PAH were in the ranges of 1.1 − 19 μg L^{-1} and 0.8 − 12 μg L^{-1} in urine samples collected at 7:00–8:00 and at 18:00–19:00, respectively. In different age groups, the concentrations of Σ_{13}OH-PAH was higher in adults (mean: 6.0 μg L^{-1}) than in children (mean: 3.5 μg L^{-1}). These results reflect the variability of metabolism activities with time and age.

The concentrations of OH-PAHs in urine were used to assess the daily intake of PAHs. To estimate internal exposure of local residents to PAHs, the following equation (Guo et al., 2013) was used to estimate the daily intakes of naphthalene, fluorene, phenanthrene and pyrene:

$$DI_1 = C_1 \times V \times \frac{1}{f} \times \frac{M_1}{M_2} \quad (7)$$

$$DI_2 = C_2 \times IR \quad (8)$$

where DI_1 is the daily intake of PAHs (μg d^{-1}) assessed by urine; C_1 is the concentration of urinary OH-PAH (μg L^{-1}); V is the human daily excretion volume of urine (L d^{-1}) and set as 2.0 L d^{-1} (Guo et al., 2013); f is the ratio of OH-PAH excreted in urine relative to total exposure dose, which was 100%, 60%, 11% and 6.8% for naphthalene, fluorene, phenanthrene and pyrene, respectively (Li et al., 2012c); M_1 and M_2 are the molecular weights (g mol^{-1}) of the parent PAH and its metabolite; DI_2 is the daily intake of PAHs (μg d^{-1}) that is not based on BaP_{eq} concentration of PAHs in food; C_2 is the concentration of PAHs in different samples types; IR is the rate of inhalation (IR_{in}, m^3 h^{-1}) or daily dietary intake of Tsampa flour (g d^{-1}) and butter tea (mL d^{-1}).

For Tibetan adults, the median DI values estimated from the OH-PAH data by Eq. (7) were 3.9, 1.9, 12 and 3.3 μg day^{-1} for naphthalene, fluorene, phenanthrene and pyrene, respectively. On the other hand, the estimated median DIs of the four target PAHs by Eq. (8) were 5.4, 2.0, 4.7 and 1.1 μg day^{-1} from dietary intake of Tsampa flour and butter tea combined and were 4.1, 0.5, 0.4 and 0.04 μg day^{-1} through inhalation (gaseous and particulate phases combined). Apparently, the estimated DIs of naphthalene and fluorene through ingestion and inhalation were slightly greater than those estimated from OH-PAHs. As for phenanthrene and pyrene, food ingestion was the dominant source, but DIs through ingestion may have been underestimated as food items other than Tsampa flour and butter tea were not considered. A recent study indicated that the dietary pattern has evolved in Tibet, i.e., consumption of multiple food items has become popular (Dermience et al., 2017). Nevertheless the DIs estimated from urinary OH-PAHs and dietary intake and inhalation of PAHs are reasonably consistent, indicating the feasibility of assessing DIs of PAHs with external exposure routes only.

It should be noted that the estimated DIs through ingestion and inhalation may have been underestimated, because several uncertain exposure sources were present in the indoor settings. Besides, concentrations of OH-PAHs in urine may also be influenced by some factors, such as residents' lifestyle, environmental variables, etc. Therefore the DIs estimated from OH-PAHs should also have been subject to

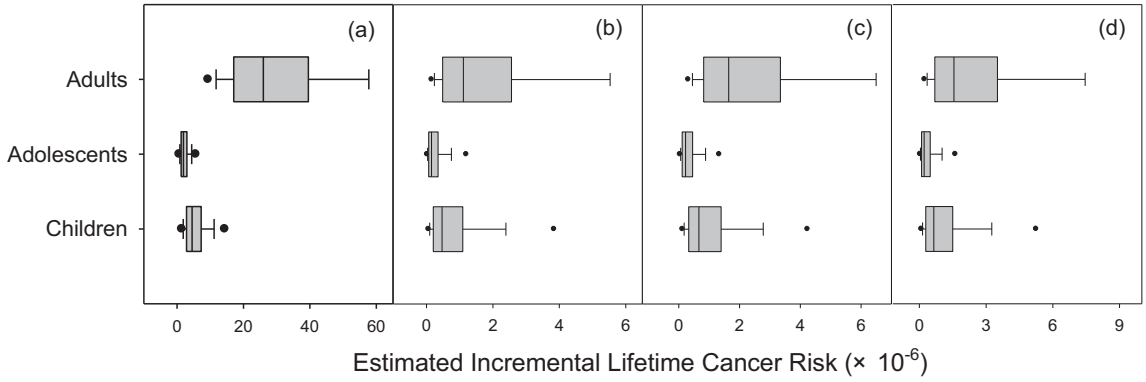

Fig. 3. Estimated incremental lifetime cancer risk for the local residents of different age groups from exposure to PAHs through: (a) dietary intake; (b) inhalation and dermal contact in bedroom; (c) inhalation and dermal contact in living room and (d) inhalation and dermal contact in kitchen. The vertical lines in the middle of the boxes represent median values. Detailed values are provided in Table S6.

certain variability, due to the small sample size. This will definitely needs further investigations.

3.4. Assessment of potential health risk

Potential incremental lifetime cancer risk (ILCR) of an exposure scenario was evaluated for the three age groups: childhood (1–11 years), adolescent (12–17 years) and adulthood (18–70 years). This exposure scenario was for residents who stayed at home 16 h a day and consumed Tsampa flour. Dietary intake contributed substantially to the ILCR, with the median values of 4.5×10^{-6} (95% CI: 1.4–14 $\times 10^{-6}$), 1.9×10^{-6} (95% CI: 0.67–5.4 $\times 10^{-6}$) and 26×10^{-6} (95% CI: 9.4–73 $\times 10^{-6}$) for children, adolescents and adults, respectively. The median ILCR value due to inhalation and dermal contact combined was 1.8×10^{-6} (95% CI: 0.06–5.3 $\times 10^{-6}$), 0.6×10^{-6} (95% CI: 0.02–1.6 $\times 10^{-6}$) and 4.3×10^{-6} (95% CI: 0.2–12 $\times 10^{-6}$) for children, adolescents and adults, respectively (Table S6). In this exposure scenario, dietary intake poses higher health risk than inhalation and dermal contact (Fig. 3).

It is interesting to note that ILCRs through inhalation and dermal contact for rural Tibetans were lower than those via inhalation exposure in Shanxi, China where the main domestic energy source was honeycomb briquette (Chen et al., 2015) and in India with dung cakes and firewood as the major energy types (Tiwari et al., 2014). The different ILCRs were likely to result from different combustion conditions. The estimated ILCRs from inhalation and dermal contact for adults in kitchens in the present study (median: 1.5×10^{-6}; 95% CI: 0.2–12 $\times 10^{-6}$) were comparable to those for inhalation and dermal exposure to barbecue fumes (median: 1.1×10^{-6}; 95% CI: 0.06–12 $\times 10^{-6}$) (Wu et al., 2015), further confirming cooking fumes as an important source of health risk for rural Tibetans.

Other models are also available for evaluating health risks of PAHs; hence the present study adopted another model (Gungormus et al., 2014; Hoseini et al., 2016) to cross-examine the potential health risks already assessed above:

$$d'CDI = \frac{C_{d'} \times IR_{d'} \times EF \times ED}{BW \times AT} \quad (9)$$

$$iCDI = \frac{C_i \times IR_i \times EF \times ED}{BW \times AT} \quad (10)$$

$$dCDI = \frac{C_d \times K_p s \times SA \times ED \times EF}{BW \times AT} \quad (11)$$

$$SA = 0.1173 \times BW^{0.6466} \quad (12)$$

$$ILCR = CDI \times \left\{ CSF \times \left(\frac{BW}{70} \right)^{\frac{1}{3}} \right\} \times cf \quad (13)$$

where d'CDI, iCDI and dCDI are chronic daily intake (CDI) via dietary, inhalation and dermal contact; $C_{d'}$ and C_i/C_d are the concentrations of BaP_{eq} in Tsampa flour and air (ng m^{-3}); IR is the rate of daily dietary intake ($IR_{d'}$, g d^{-1}) or inhalation (IR_i, m^3 d^{-1}); SA is the total body surface area (m^2) and the exposed dermal fraction is 25% of SA in the present study; K_ps is the permeability coefficient of gaseous or particle-bound compounds (m d^{-1}); CSF is the cancer slope factor of BaP for a specific exposure route (kg d mg^{-1}) and *cf* is the conversion factor (10^{-6}). Other parameters are already defined in Section 2.5 (Data analysis). Values of these parameters are provided in Table 2.

Dietary intake contributed substantially to the ILCR, with median values of 0.39×10^{-6} (95% CI: 0.14–1.1 $\times 10^{-6}$), 0.43×10^{-6} (95% CI: 0.15–1.2 $\times 10^{-6}$) and 7.6×10^{-6} (95% CI: 2.7–21 $\times 10^{-6}$) for children, adolescents and adults, respectively. The median ILCR value due to inhalation and dermal contact combined was 4.9×10^{-8} (95% CI: 0.55–36 $\times 10^{-8}$), 4.3×10^{-8} (95% CI: 0.44–32 $\times 10^{-8}$) and 0.44×10^{-6} (95% CI: 0.04–3.3 $\times 10^{-6}$) for children, adolescents and adults, respectively. Apparently, this model obtained lower potential health risks than the model represented by Eqs. (1)–(6), but still indicated certain levels of cancer risk rural Tibetans.

3.5. Global implications for health risk of remote rural groups

Residing in one of the cleanest regions in the world, the majority of Tibetans still follows traditional lifestyles, e.g., leading a nomadic life, using traditional Tibetan tents (Li et al., 2012a) and consuming traditional foods. The outdoor air of Tibet is considerably cleaner relative to urban China (Zhang et al., 2003; Zhang et al., 2008), but indoor air pollution is serious due to combustion of solid biomass fuels, notably yak dung (Carter et al., 2016; Hu et al., 2016). Health statistics show that the hospitalization rates of respiratory system diseases in Tibet are higher than the national average (MOH, 2009; The Health Department of Tibet Autonomous Region, 2006). Because of limited economic options and unaffordable fossil fuel prices (Xiao et al., 2015), biomass fuels are expected to remain for a long time as the dominant energy types in Tibet.

The above-mentioned scenario is not only for Tibetans, but also for residents in other remote rural regions with traditional lifestyles. For example, biomass fuels accounted for 90% of domestic energy consumption in Malawi (Jumbe and Angelsen, 2011). The use of low-quality biomass fuels was found to highly associate with neurologic, respiratory, cardiopulmonary and other symptoms (Das et al., 2017). Rural areas of Bhutan also accounted for 96% of the total firewood consumption in the country (Wangchuk et al., 2017). A previous study found that large-scale combustion of firewood may result in substantial burdens of respiratory diseases (Bruce et al., 2013). Lung cancer was also demonstrated to significantly relate to household air pollution (Iii

Table 2
Risk parameters used for estimating human exposure to polycyclic aromatic hydrocarbons.[a]

Parameter		Children (1–11 years old)	Adolescent (12–17 years old)	Adult
IR_{food}		LN (50, 25)	LN (100, 50)	LN (200, 100)[b]
IR_{in}		LN (1.1, 0.07)[c]	LN (1.4, 0.04)	LN (1.5, 0.05)
SA		LN (0.95, 0.24)[c]	LN (1.59, 0.05)	LN (1.75, 0.21)
BW		LN (19.0, 6.28)[c]	LN (47.3, 7.30)	LN (60.8, 3.97)
f_{sa}		25%[d]	25%	25%
t_{event}	Cooking time	U (0.5, 1)[e]	U (0.5, 1)	U (0.5, 1)
	Non-cooking time	U (15, 15.5)	U (15, 15.5)	U (15, 15.5)
k_{p_p} (for BaP)		LN (0.007, 0.014)[f]	LN (0.007, 0.014)	LN (0.007, 0.014)
k_{p_g} (for BaP)		LN (2.25, 3.17)[g]	LN (2.25, 3.17)	LN (2.25, 3.17)
ED		11[c]	6	53
EF		365	365	365
AT		25,550[c]	25,550	25,550
CSF for ingestion		LN (7.3, 1.56)[h,i]		
CSF for inhalation		LN (3.9, 1.8)[j,k]		
CSF for dermal contact		37.4[j], 30.5[i], 25[l]		
C_{ip}	Cooking time	LN (4.7, 4.7) for bedroom; LN (0.6, 1.1) for living room; LN (6.3, 8.3) for kitchen[m]		
	Non-cooking time	LN (0.03, 0.01) for bedroom; LN (0.03, 0.02) for living room; LN (0.03, 0.01) for kitchen[m]		
C_g	Cooking time	LN (3.1, 2.7) for bedroom; LN (3.7, 0.9) for living room; LN (4.7, 2.6) for kitchen[m]		
	Non-cooking time	LN (0.2, 0.2) for bedroom; LN (03, 0.2) for living room; LN (0.2, 0.3) for kitchen[m]		
C_{dp}	Cooking time	LN (4.7, 4.7) for bedroom; LN (0.6, 1.1) for living room; LN (6.3, 8.3) for kitchen[m]		
	Non-cooking time	LN (0.03, 0.01) for bedroom; LN (0.03, 0.02) for living room; LN (0.03, 0.01) for kitchen[m]		
C_{food}		LN (1.7, 0.6)[m]		

[a] IR is the rate of dietary intake (IR_{food}; g d^{-1}) or inhalation (IR_{in}; m^3 h^{-1}); SA is the skin surface area (m^2); BW is the average body weight (kg); f_{sa} is the exposed dermal fraction; t_{event} is the duration time for a day with exposure (h d^{-1}); k_{p_p} and k_{p_g} are the transdermal permeability coefficients (m h^{-1}) of particle-bound and gaseous compounds, respectively; ED and EF refer to the lifetime exposure duration (yr) and exposure frequency (d yr^{-1}), respectively; AT is the average time for carcinogenic effects (d); CSF_i is the cancer slope factor of BaP for exposure route i (kg d mg^{-1}) and is age-dependent as specified by average body weight; C_{ip} and C_{dp} represent the sum of BaP_{eq} concentrations of particle-bound PAHs (ng m^{-3}); C_g is the BaP_{eq} concentrations of gaseous PAHs (ng m^{-3}); C_{food} is the BaP_{eq} concentration of PAHs in Tsampa flour (ng g^{-1}); LN (A, B) is the lognormal distribution with A as the mean value and B as the standard deviation. U (A, B) is the uniform distribution with the minimum of A and the maximum of B.

[b] Adopted from Gao et al. (2017).

[c] Adopted from the International Commission on Radiological Protection Publication 66 (1994).

[d] "25%" is the percent of dermal area (neck, head, and/or ante brachium) that can be in touch with cooking fumes.

[e] Estimated from the local schedules.

[f] Adopted from the United States Environmental Protection Agency (1992).

[g] Adopted from Weschler and Nazaroff (2012). Because the temperatures (19–24 °C) in the present study were different from that (32 °C) in Weschler et al.'s study, half of the cited value (4.5) was used.

[h] Adopted from Chen and Liao (2006).

[i] Adopted from Office of Environmental Health Hazard Assessment (2017).

[j] Adopted from the United States Environmental Protection Agency (USEPA) (2011).

[k] Adopted acquired from Chen et al. (2012).

[l] Adopted from Knafla et al. (2006).

[m] From the present study.

et al., 2011). Therefore the pristine lifestyles still led by remote rural populations result in PAH exposures should be adequately evaluated.

4. Conclusions

The present study indicated that rural remote Tibetans were subject to meaningful health risk despite their pristine lifestyle and good outdoor air quality. Although dietary ingestion remained the most prominent source of health risk, indoor exposure to biomass fuel-combusted PAHs through inhalation and dermal contact combined could pose higher incremental lifetime cancer risk than 10^{-6}, a commonly accepted no-effect threshold (USEPA, 2013). The findings from the present study have raised concerns about the potential health risk to rural remote residents despite their pristine lifestyles, which merits adequate investigations in the near future.

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Appendix A. Supplementary data

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