

Article

Emissions from Solid Fuel Cook Stoves in the Himalayan Region

Jin Dang ^{1,*}, Chaoliu Li ², Jihua Li ³, Andy Dang ^{4,5}, Qianggong Zhang ², Pengfei Chen ⁶, Shichang Kang ^{2,6}  and Derek Dunn-Rankin ¹ 

¹ Department of Mechanical and Aerospace Engineering, Henry Samueli School of Engineering, University of California, Irvine, CA 92697, USA; ddunnran@uci.edu

² Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing 100101, China; lichaoliu@itpcas.ac.cn (C.L.); Qianggong.zhang@itpcas.ac.cn (Q.Z.); shichang.kang@lzb.ac.cn (S.K.)

³ Qujing Center for Disease Control and Prevention, Yunnan 655011, China; ynj_cn@sina.com

⁴ Program in Public Health, Susan and Henry Samueli College of Health Sciences, University of California, Irvine, CA 92697, USA; andyd@uci.edu

⁵ Department of Epidemiology, School of Medicine, University of California, Irvine, CA 92697, USA

⁶ State Key Laboratory of Cryosphere Science, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences, Lanzhou 730000, China; chenpengfeifeifei@126.com

* Correspondence: dangj1@uci.edu; Tel.: +1-949-385-1177

Received: 12 January 2019; Accepted: 12 March 2019; Published: 21 March 2019



Abstract: Solid fuel cooking stoves have been used as primary energy sources for residential cooking and heating activities throughout human history. It has been estimated that domestic combustion of solid fuels makes a considerable contribution to global greenhouse gas (GHG) and pollutant emissions. The majority of data collected from simulated tests in laboratories does not accurately reflect the performance of stoves in actual use. This study characterizes in-field emissions of fine particulate matter ($PM_{2.5}$), carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), and total non-methane hydrocarbons (TNMHC) from residential cooking events with various fuel and stove types from villages in two provinces in China (Tibet and Yunnan) in the Himalayan area. Emissions of $PM_{2.5}$ and gas-phase pollutant concentrations were measured directly and corresponding emission factors calculated using the carbon balance approach. Real-time monitoring of indoor $PM_{2.5}$, CO_2 , and CO concentrations was conducted simultaneously. Major factors responsible for emission variance among and between cooking stoves are discussed.

Keywords: solid fuel; cooking stove; field study

1. Introduction

Solid fuel cooking stoves continue to be used and relied upon in many parts of the world. There are more than two billion people using direct burning of solid fuel as their primary energy source [1,2], especially in developing countries where cooking stoves primarily burn biomass or coal. Furthermore, it has been estimated that worldwide domestic combustion of solid fuels from residential use and small-scale industry contribute approximately 34% of total black carbon (BC) emissions [3]. Biomass has been used directly as a fuel since the harnessing of fire by humans [4], and coal has been used since the second and third century of the Common Era [5]. Biomass fuels fall at the low end of the energy ladder and, consequently, require large volumes and mass relative to the energy delivered. As a result, they often produce a high level of combustion emissions. For household energy sources, the energy density ladder can be expressed as: dung < crop residues < wood < kerosene < gas < electricity [6]. Although switching to a higher energy ladder fuel or adopting new technology like gasification with co-generation provides a cleaner way to acquire energy [7], there are still large

populations that use biomass and coal directly as fuel for cooking and heating. Coal has a high energy density, but also contains substantial levels of dangerous compounds, including sulfur and heavy metals. The wide-spread use of solid fuel due to human activity results not only in significant emission contribution to the atmosphere, but also negatively affects indoor air quality and public health.

Unlike other well-studied categories of combustion emission sources such as diesel engines [8–10], the emission inventory for the residential and small-scale industry sector is rarely investigated. In particular, depending on the type of fuel, emissions from solid fuel cooking stoves have a complicated make-up which includes well-mixed greenhouse gases (WMGHG) like carbon dioxide and methane, pollutants such as carbon monoxide, sulfur dioxide (mostly when coal is used as the fuel source), hydrocarbons, and particulate matter (PM), as well as small concentrations of volatile organic compounds [2]. The potential radiative forcing from these complex emissions is still unclear, especially for particulate matter [11]. This study aims to measure cooking stove emissions while they are in use to permit more accurate characterization of the potential local and global climate impact from domestic solid fuel combustion.

Studies of domestic solid fuel combustion emission have been underway for many years, but due to the limitation of technology deployment, the experimental study of biomass combustion emissions started only in the late 20th century [6,12–15]. With the help of statistical models, historical emissions data are available for major species including methane, carbon monoxide, nitrogen oxides, total and specialized non-methane volatile organic compounds (NMVOCs), ammonia, organic carbon, black carbon, and sulfur dioxide [16]. Unfortunately, the complexity and dispersivity of the emission sources means that the model study does not provide estimates with high accuracy and precision. In particular, several studies indicated that models consistently underestimate the carbon monoxide [17–21] and black carbon contributions resulting from biomass cooking stoves' use [22].

Controlled laboratory measurements of solid fuel cooking stoves have been made by many groups [23–26]. The widely-used testing protocol includes the water boiling test (WBT) and kitchen performance test (KPT). However, it has not been well-demonstrated that current testing protocols represent the actual everyday cooking and heating activities in homes, and there is still a lack of a confirmed explanation regarding the difference between laboratory and in-field measurements [27]. The actual emissions from household cooking stoves depend on several variables including: stove type, fuel type, food type, and the behavior of the cooks cooking the food. Laboratory experiments with uniformity and repeatability are not similar to everyday cooking and heating activities and, therefore, may not reflect the in-home conditions, nor the unavoidable variation of resident stove activities. This situation leads to highly uncertain in-field data [28].

Compared with laboratory experiment studies, in-field measurement has significant challenges in producing high-quality field data. For example, since most of the residents who use solid-fuel cooking stoves as their primary energy source live in rural areas, it is often hard to access these in-field sites. Furthermore, rural areas that rely on solid-fuel often have limited, or even no electrical power supply, which greatly restricts the measurement capabilities [29]. Moreover, taking measurements in homes is not as straightforward as doing so in a laboratory since the in-field environment is generally in an active family location. Local coordination plays an important role in this process.

2. Methodology

Emission factors of carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), and fine particles ($PM_{2.5}$) were measured during normal daily activities using the carbon balance approach. Real-time monitoring of indoor $PM_{2.5}$, CO_2 , and CO concentrations was conducted simultaneously.

2.1. In-Field Sampling

Globally, the emission to the environment from household cooking stoves' activity largely depends on the regional population density. The Himalaya Mountains, as one of the largest fresh water resources in the world, have a population of more than two billion people living on the rivers that originate

from this source. The high population density in the nearby area leads to a significant contribution of emissions from domestic residential combustion. Hence, the field sites for this study were selected to be in two provinces of China (Tibet Autonomous Region and Yunnan Province) that are closest to the Himalaya Mountain range.

In general, each field campaign lasted for approximately two months with about 6 weeks of measurement time and 1 or 2 weeks for pre- and post- preparation. The basis for the selection of households included the ability to measure a variety of region-specific primary and secondary stove and fuel types. Depending on the given sites, there were also constraints and considerations taken into account regarding household selection. The campaigns took place during summer of 2012 and 2013 in Tibet and Yunnan, respectively. Thirty eight household samples were collected in the Tibet measurement and 40 for Yunnan. Figure 1 shows the geographical location of the field sites.

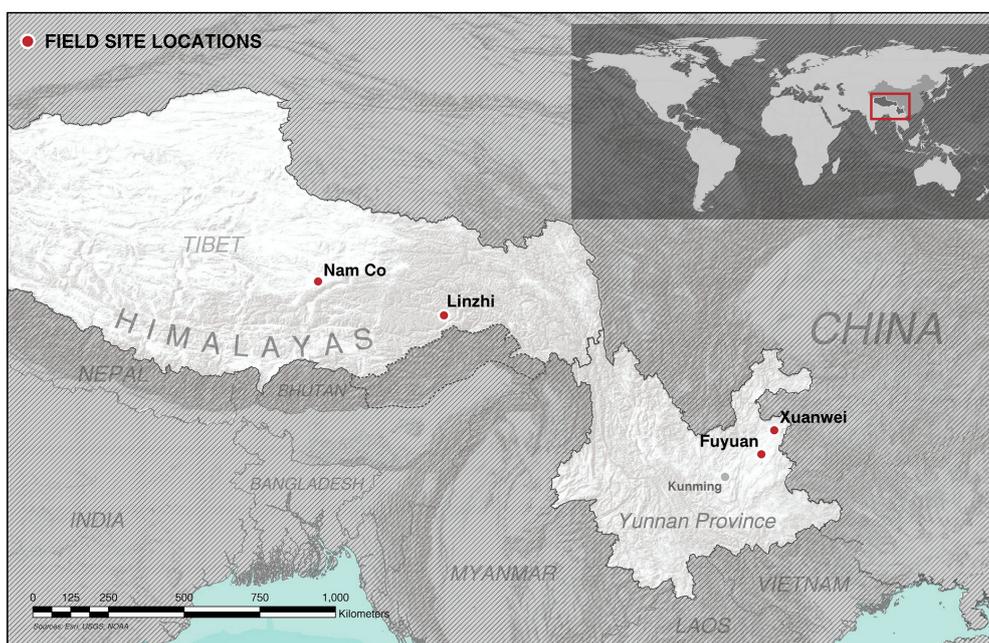


Figure 1. The geographical location of our field sites.

2.1.1. Tibet, China

The research in Tibet, China, was coordinated through the Institute of Tibetan Plateau Research, Chinese Academy of Sciences. Due to the limitation of the local environment, two regions, Nam Co and Linzhi, were chosen as the sites in which to conduct the in-field measurements. Figure 2 shows the household, stove, and fuel used in Tibet.

Nam Co has an extremely high elevation (approximately 4700 m). Due to the scarcity of plant and animal sources as biomass, local residents rely on yak dung as their primary fuel. Most of the local residents lead a traditional nomadic life, with a Tibetan tent as their shelter. The tourist industries being developed in the Nam Co area absorbed some residents to join the tourism business. As a result of their more settled living style, these people built fixed or semi-fixed households instead of traditional tents. There are two types of stoves being used in the Nam Co area: a traditional open fire stove and an improved chimney stove. The open fire stove is mostly used by nomadic residents, as it has better portability. All the fixed and semi-fixed households are now using an improved chimney stove. There was limited availability for household selection. As the population is very sparse in the Nam Co area, measurements were taken in whatever household could be found.

The Linzhi area has a much lower elevation (approximately 3300 m) compared to Nam Co. Lower elevation brings richer plant and animal sources. As a result, wood becomes the primary fuel

for the Linzhi residents. The well-developed agriculture and tourist industry significantly improved the living condition of local residents. All the residents have well-built houses with well-designed chimney stoves (different from the ones in Nam Co). As there is a uniformity of households, stoves, and fuel type, the selection of participating households in the Linzhi area was mostly based on regional considerations. All the measured households in the Linzhi area were located in the two villages that are close to the Linzhi Research Station, Chinese Academy of Sciences.



Figure 2. The household, stove, and fuel in Tibet (**top left:** Tibetan tent in Nam Co; **top middle:** chimney stove; **top right:** household in Linzhi; **bottom left:** open fire stove in Nam Co; **bottom middle:** household in Nam Co; **bottom right:** yak dung).

The sampling system contained two sampling trains: one for cooking stove emission and the other for background monitoring. Starting with the probe, the emission sample traveled through a length (depending on the stove, typically around 2 m) of conductive tubing where it entered the sample train. The particle loss through 3 meters of this tubing was measured at about 2% by the University of Illinois Urbana-Champaign (UIUC) field sampling team [30]. A dilution pump was utilized to avoid instrument saturation and also to reflect natural dilution of chimney emissions. A cyclone separator cut off particles larger than 2.5 μm at a fixed flow rate (1.5 L/min). The first branch of the train collected elemental carbon (EC), organic carbon (OC), and gas samples. The EC/OC sample were collected with a 47-mm quartz filter. The gas sample was collected with a 200-L Kynar bag. The sampling duration generally started from breakfast cooking to the end of dinner cooking. However, Tibetan residences do not hold a regular cooking schedule as people from most of the other places due to their nomadic tradition. On the other hand, being constrained to this logistical limitation, in this case, the sampling event started around mid-morning and lasted approximately 7 h. The flow rate was constrained at 0.2 L/min with an SKC pocket sampling pump to collect all the gas with the 200-L bag. The second branch included one 37-mm PTFE filter and one 47-mm quartz filter to collect PM samples and EC/OC in the gas phase. The filtered gas traveled through a TSI Q-Trak 7575 CO/CO₂ monitor and Dräger PAC 7000 monitor with a SO₂ sensor to acquire real-time CO, CO₂, and SO₂ information. The flow rate for the second branch was initially set at about 0.2 L/min. The third branch was for a DustTrak real-time aerosol monitor. The flow rate was set at 1.1 L/min to satisfy the 1.5-L/min requirement of the cyclone.

2.1.2. Yunnan, China

The study in Yunnan province was in cooperation with the Center for Disease Control and Prevention (CDC) of Qujing City. Yunnan is a rich coal province with mild climate, which brings an abundance of forestry and agricultural resources (elevation: approximately 2000 m). As a result, residents in Yunnan province have various fuels to choose from: coal, wood, corn, pine needles, and agricultural residue. However, people mostly use wood, corn, pine needles, or other biomass fuel to start the fire and then use coal to keep the stove burning (Figure 3). Hence, coal is considered as the primary fuel in Yunnan province, and covering all of the mainstream coal types (e.g., gas fat, smoky, coking, and smokeless) was the main consideration while selecting participating households.

The stove usage situation in Yunnan province is quite different from the other sites. As Yunnan province is relatively developed compared with Tibet, there are well-constructed electricity grids in this area, which provide local residents reliable and affordable power sources. As a result, many residents in the village switched to electric stoves (induction cooktop) for their primary cooking. Solid fuel stoves are still widely used for heating during the cold weather season (especially for the elderly) and preparing food for animals (electric stoves are not large enough for this task).

The solid fuel cooking stoves used in Yunnan generally are one of three different types: high stove, low stove, and portable stove (Figure 3). The high stove is a chimney stove, which is mainly used for cooking (before the electric stove became popular); the current low stove has a similar design to the high stove, but it sits lower to the ground level, which makes it a good floor heater. The portable stove is popular in villages, especially among seniors, as it is good for heating and easy to carry around.



Figure 3. The stove and fuel in Yunnan (**top left:** high stove; **top middle:** portable stove; **top right:** low stove, **bottom left:** coal, **bottom right:** corn).

2.2. The Carbon Balance Method

The measurement and analysis relied on the carbon balance method [31], which is commonly used for biomass combustion emission studies [32]. This method calculates the emission factor based on the carbon processed during fuel consumption and the ratio between pollutants in the exhaust gas [2,29]. In order to achieve a representative measurement, the sample is taken after the plume is well-mixed [23]. A prior study indicated that emissions take less than 2.5 s to reach phase equilibrium [33]. In our setup for open-fire stoves, the sample probe was located about 1 m above the stove, which left 3–4 s for the plume to mix before reaching the probe. For the chimney stove measurements, as the length of the chimney was mostly more than 2 m long and the flow inside

the chimney was turbulent ($Re > 4000$) [23], we assumed the emissions were well-mixed within the chimneys, and we collected our sample from the chimney outlet.

2.3. Post-Measurement Analysis

The post-measurement analysis included gravimetric analysis for the $PM_{2.5}$ sample (PTFE filter) and gas chromatography analysis for the gas sample. The analysis of the EC/OC (quartz filter analysis) was conducted by collaborators in the UIUC group using a Sunset Laboratory OC/EC analyzer (thermal optical transmittance method) [34].

2.3.1. Gravimetric Analysis

Gravimetric analysis was applied to the PTFE filters collected from field measurements. Before heading to the field, the PTFE filters were weighed and sealed (pre-weights). A post-weight was conducted after the measurement in the field with the PM sample collected on the filter. With the recorded flow information and the weight difference between pre- and post-analysis, the $PM_{2.5}$ emission was calculated.

2.3.2. Gas Chromatography Analysis

Gas chromatography analysis was applied to the collected gas sample to investigate the concentration for interesting gas species, which include carbon dioxide, carbon monoxide, methane, and total hydrocarbon (THC). The detection of carbon dioxide and carbon monoxide were achieved with a flame ionization detector (FID) plus nickel catalyst methanizer (SRI Instruments, USA). The THC analysis was accomplished using a blank column plus FID. This approach takes advantage of the fact that the FID responds only to hydrocarbons [12]. External standardization was selected to calibrate the GC analysis. Calibration gases with known concentrations of target gas species were used to generate the calibration curve.

3. Results

The finalized database included emission factors for: carbon dioxide, carbon monoxide, methane, and $PM_{2.5}$ for each sample from the field sites. The emission factors were calculated with the carbon balance method. Modified combustion efficiency (MCE) was defined as the ratio between carbon in the form of carbon dioxide to that in the form of carbon dioxide plus that in the form of carbon monoxide. MCE is commonly used as an approximation of nominal combustion efficiency. Since most of the carbon emissions are in form of CO_2 and CO , MCE provides a robust approximation to the normalized combustion efficiency (NCE) [12].

$$MCE \equiv \frac{[CO_2]}{[CO_2] + [CO]} \quad (1)$$

3.1. Tibet

Table 1 provides the summary of the Tibet measurement. Thirty eight valid samples were collected: 28 samples from Nam Co and 12 from Linzhi. There were two small-scale industry measurements conducted in Nam Co. Both were small restaurants with exactly the same type of stoves used in local households.

The sparse population and limited transportation capability restricted the sampling time in each household. Tibet testing time was limited to around 6 h. Figure 4 shows a examples of the real-time pollutant concentration in Nam Co and Linzhi. The emission pattern from Tibet did not show obvious cooking events. This difference was attributed to the lifestyle of Tibetan residents and the desire for space heating for the stove. Particularly, in the Nam Co area, the nomadic life does not have a regular daily meal schedule (breakfast, lunch, snack, and dinner). As shown in Figure 4, the daytime stove activity in Tibet did not give obvious patterns representing regular cooking events. The cold weather

in these high altitude regions encourages local residents to use cooking stoves for heating purposes, as well. The statistical summary for the Tibet measurements is shown in Table 2.

Table 1. Tibet household summary.

Location	Residence Type	Stove Type	Fuel Type	Measurement	# of Samples
Nam Co	Tent	Open fire	Yak dung	1-Day	4
	Tent	Chimney	Yak dung	1-Day	4
	Prefab house	Chimney	Yak dung	1-Day	12
	Stone house	Chimney	Yak dung	1-Day	6
	SSI	Chimney	Yak dung	1-Day	2
Linzhi	Garret	Chimney	Wood	3-Day	4
	Garret	Chimney	Wood	1-Day	8
Total					38

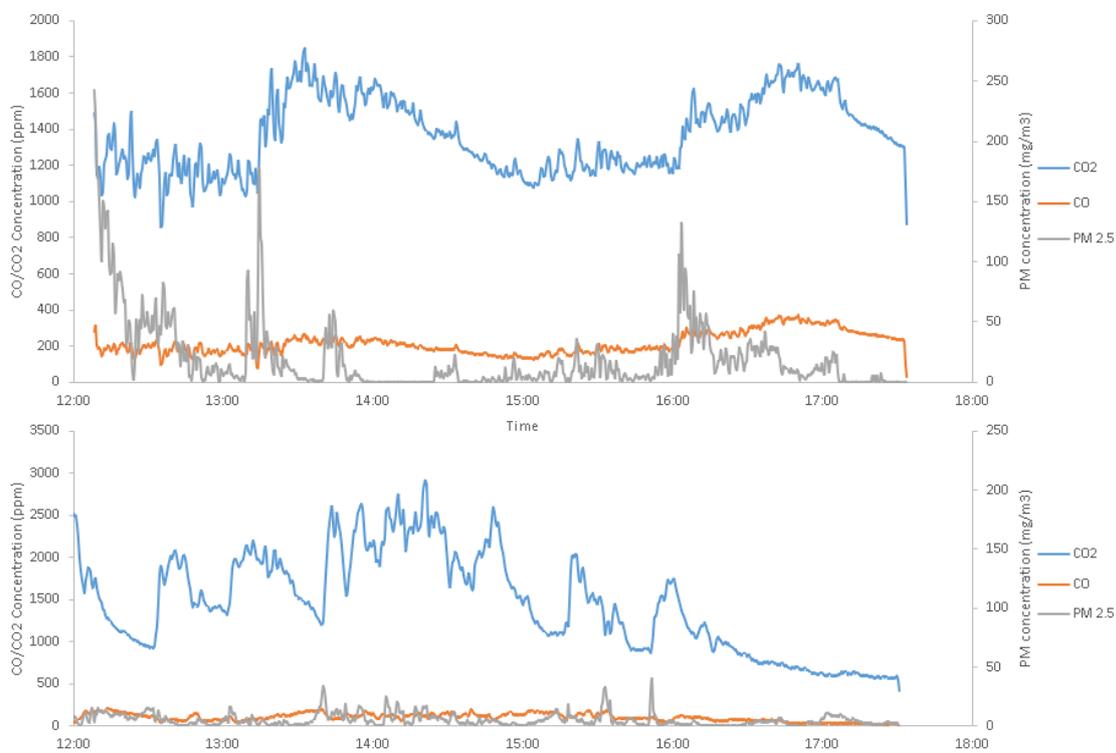


Figure 4. Typical real-time emission in Tibet, top: Nam Co; bottom: Linzhi.

Table 2. Statistical summary for the Tibet measurement.

	Sample Amount	MCE (%)	EF CO ₂ (g/kg Fuel)	EF CO (g/kg Fuel)	EF PM _{2.5} (g/kg Fuel)	EF CH ₄ (g/kg Fuel)
Dung, open fire stove in tent	3	73.0 ± 7.5	1298.8 ± 148.3	303.1 ± 80.8	18.5 ± 10.2	32.4 ± 7
Dung, chimney stove in tent	2	90.2 ± 7.1	1590.4 ± 176.3	107.1 ± 76.4	26 ± 26	5.8 ± 1.6
Dung, chimney stove in house	15	91.4 ± 1.8	1632.4 ± 38.1	96 ± 20	14.7 ± 4.1	22 ± 4.6
Wood, chimney stove in house	15	84.0 ± 3.5	1282.9 ± 64.4	150.7 ± 31.5	18.6 ± 3.8	9.8 ± 1.3
All dung	22	88.6 ± 2.1	1579 ± 41.5	127.7 ± 23.2	16.3 ± 3.6	24.1 ± 3.8
All household	35	86.6 ± 2.0	1451.6 ± 44.9	137.8 ± 19.6	17.3 ± 2.7	16.7 ± 2.5
All SSI	2	89.1 ± 4.6	1587.5 ± 97.5	122.6 ± 51.5	16.1 ± 7.7	46.1 ± 5.7
Overall	37	86.7 ± 1.9	1459 ± 42.9	137 ± 18.6	17.2 ± 2.6	18.3 ± 2.6

3.2. Yunnan

Table 3 shows the summary of the Yunnan measurements. The Yunnan measurement set contained 40 valid samples from two counties with 10 villages. The household types in Yunnan are uniform, while stove type varies.

Table 3. Yunnan household summary.

Location	Residence Type	Stove Type	Fuel Type	Measurement	# of Samples
Fuyuan	House	High stove	Coal	1-Day	13
	House	High stove	Coal	3-Day	2
	House	Portable stove	Coal	1-Day	5
	House	Portable stove	Coal	3-Day	1
	House	Low stove	Coal	1-Day	1
Xuanwei	House	High stove	Coal	1-Day	7
	House	High stove	Coal	3-Day	1
	House	Portable stove	Coal	1-Day	8
	House	Portable stove	Coal	3-Day	1
	House	Low stove	Coal	1-Day	1
Total					40

Figure 5 shows an example of the pollutant concentration throughout the day. The two major peak emission event groups indicated the traditional breakfast and lunch event in a Chinese village. The dinner cooking event, which usually happens at approximately 18:00, could not be monitored due to local collaborator unavailability. The smaller peak at around 15:00 was normally caused by water boiling or a snack event. As corn is often used as the stove starter, the moisture content in it generated a large amount of smoke at the starting stage of each cooking event; this process showed a strong $PM_{2.5}$ peak concentration at the beginning of every cooking event, while the concentration of CO and CO_2 did not have the same behavior.

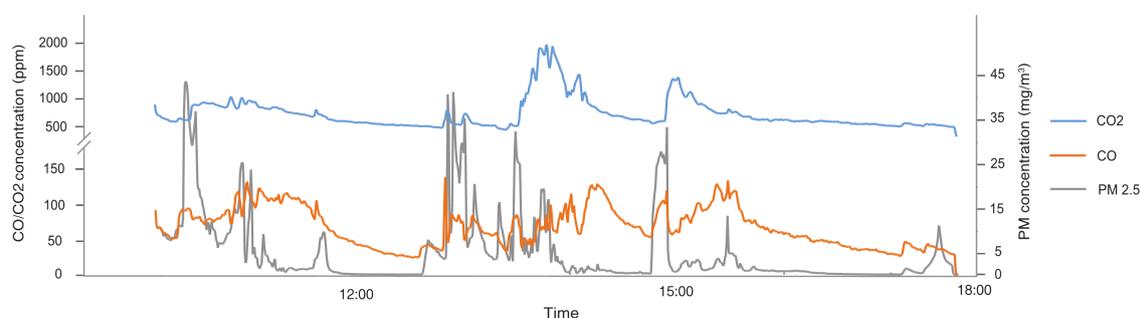


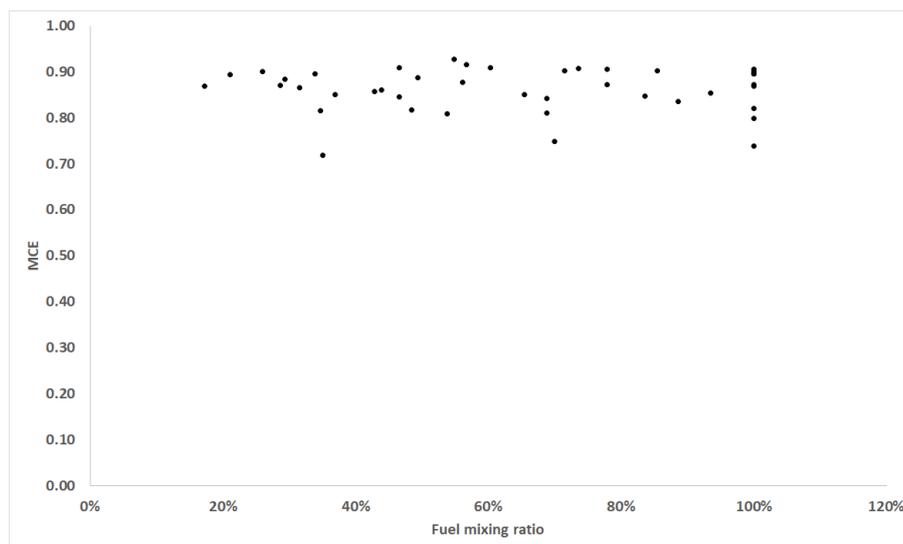
Figure 5. Typical real-time emission pattern for CO_2 , CO, and $PM_{2.5}$ in Yunnan.

Table 4 is the summary for the Yunnan dataset. As previously mentioned, mixed fuel usage is very popular in Yunnan. The native definition of the emission factor determined that it was very sensitive to fuel consumption [15]. Thus, it is important to separate when using agricultural residue as a lighter only and using coal as the major energy source and using both agricultural residue and coal as the energy source. Based on observations in the field, the threshold value of 2 kg of agricultural residue consumption was selected as the criteria of whether a household was using agricultural residue as a lighter only. In the case of more than 2 kg of agricultural residue consumed, the emission factors were calculated based on the weight of total fuel (coal plus agricultural residue). In the other case, only coal consumption was considered in the emission factor calculation.

Table 4. Statistical summary for the Yunnan measurement.

	Sample Amount	MCE (%)	EF CO ₂ (g/kg Fuel)	EF CO (g/kg Fuel)	EF PM _{2.5} (g/kg Fuel)	EF CH ₄ (g/kg Fuel)
High stove in Fuyuan	15	82.7 ± 2.7	1528.366 ± 174.458	194.982 ± 30.674	16.585 ± 3.49	83.584 ± 17.85
Portable stove in Fuyuan	6	87.1 ± 3.1	1973.451 ± 217.36	199.102 ± 60.626	12.052 ± 2.311	106.308 ± 24.3
Low stove in Fuyuan	1	85.2	1379.019	152.8224	32.09369	89.74726
High stove in Xuanwei	9	85.8 ± 1.4	1573.745 ± 251.195	172.03 ± 34.978	21.757 ± 8.609	61.563 ± 11.245
Portable stove in Xuanwei	11	88.2 ± 0.7	1457.356 ± 269.818	125.194 ± 25.393	13.323 ± 4.251	88.588 ± 20.922
Low stove in Xuanwei	1	89.7	2437.381	178.345	2.360746	54.69838
Overall Fuyuan	22	84.0 ± 2.1	1642.964 ± 137.47	194.189 ± 25.892	16.054 ± 2.58	90.061 ± 13.71
Overall Xuanwei	21	87.2 ± 0.7	1553.905 ± 178.872	147.798 ± 20.169	16.415 ± 4.332	75.392 ± 12.083
Overall high stove	24	83.8 ± 1.8	1545.383 ± 140.819	186.375 ± 22.867	18.525 ± 3.817	75.326 ± 11.943
Overall portable stove	17	87.8 ± 1.1	1639.507 ± 196.098	151.279 ± 27.279	12.874 ± 2.814	94.842 ± 15.704
Overall low stove	2	87.4 ± 2.3	1908.2 ± 529.181	165.584 ± 12.761	17.227 ± 14.866	72.223 ± 17.524
Overall	43	85.6 ± 1.1	1599.47 ± 111.006	171.533 ± 16.7	16.23 ± 2.463	82.897 ± 9.128

In order to explore the effect from mixing agricultural residue (mostly corn cob) together with coal, the correlation of modified combustion efficiency with different fuel mixing factors is plotted in Figure 6. The mixing ratio here is defined as the fraction of coal consumption (kg) in the total fuel consumption (kg). Modified combustion efficiency, which is essentially a comparison of how much carbon is emitted in CO₂ and CO form, was used as the indicator because it is estimated independently of fuel information. The result in Figure 6 shows that MCE distributed evenly through the whole fuel mixing ratio range, which implies that the mixing ratio did not have a significant effect on the stove performance.

**Figure 6.** MCE at different fuel mixing ratios.

4. Discussion

4.1. Efficiency and Emissions

Figure 7 compares the modified combustion efficiency for wood burning cooking stoves between this study (with uncertainties) and several previous works, including both the water boiling test and the measurement of actual stove usage. The MCE measured from actual stove use in the field was consistently lower when compared with those measured from standard water boiling tests. A comparison for the CO emission factor (Figure 8) gave similar results. The measurements on actual stove use in homes gave significantly higher CO emission and larger uncertainty than the WBTs.

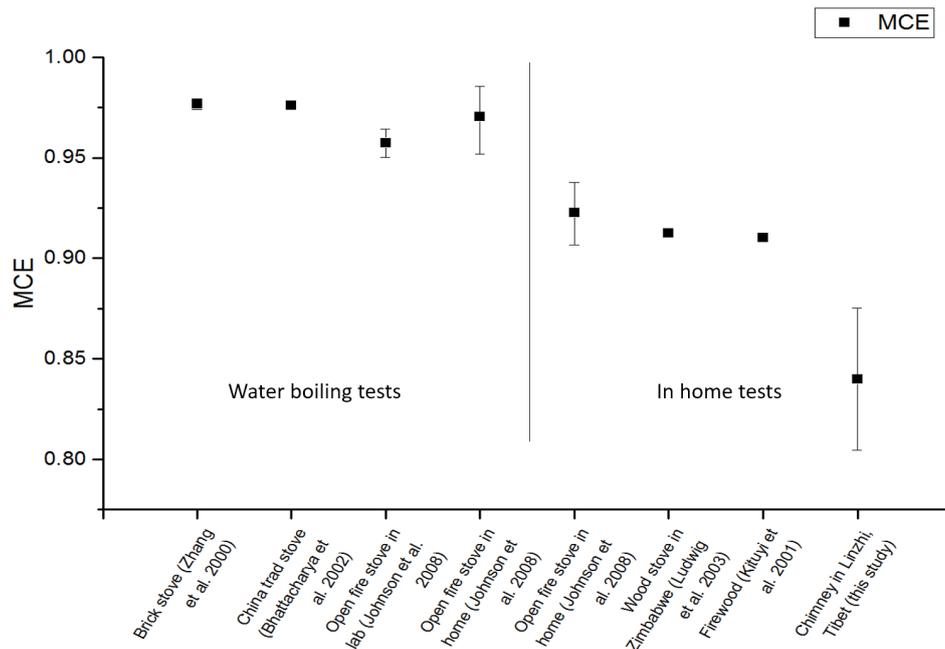


Figure 7. The MCE comparison.

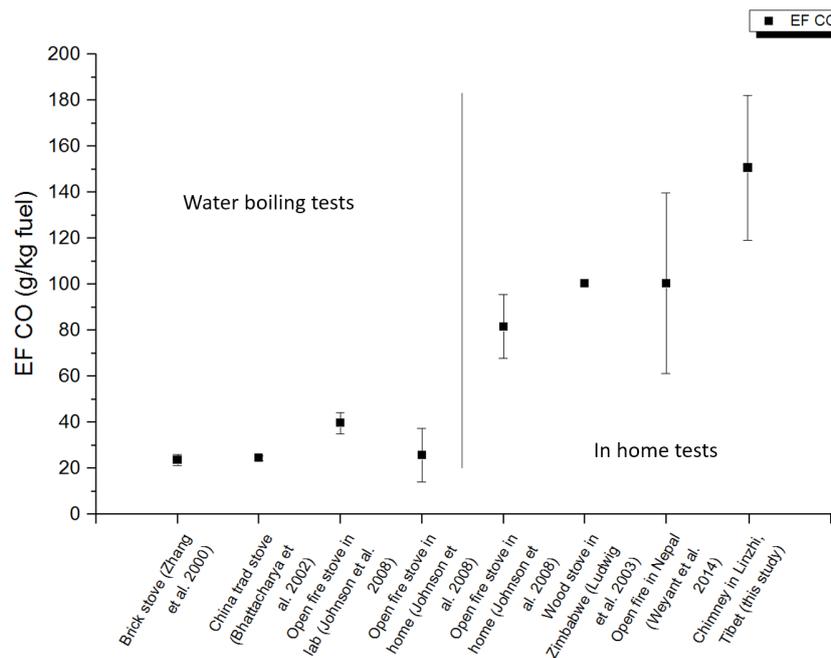


Figure 8. The CO emission factor comparison.

Figure 9 compares the emission parameters from different regions with different fuels within this study. The MCE results from all regions with all different fuels showed a similar average value.

The CO_2 emission factor is a good indicator for carbon emission, as most emissions come out in the form of CO_2 . As it has been well-addressed, fuel makes a big difference in the carbon emission. One major reason for this is the carbon density in fuel, which directly affects the emission factor of carbon compounds. For example, trunk woods normally have a carbon content of about 50%. The carbon content for coal can be over 90% [35]. Comparing the CO_2 emission factor for the wood stoves (Tibet) and coal stoves (Yunnan), the carbon emission from coal stoves was significantly higher (Figure 9).

CO and $PM_{2.5}$ are the major products of incomplete combustion, and they are closely related to indoor air quality and residents' health. One challenge for CO and $PM_{2.5}$ measurements is the associated uncertainties, especially for $PM_{2.5}$. Dung is generally considered as a more "dirty" fuel. However, the yak dung stove in Nam Co, Tibet, gave lower CO and $PM_{2.5}$ than the wood stoves in Linzhi, Tibet. As previously discussed, the stoves in Nam Co, Tibet, due to the harsh environment (high altitude, cold), are partially used as heating stoves. This difference on "how the stove is used" may explain this unusual behavior. Among all the field sites in this study, Yunnan coal stoves had the highest CO and $PM_{2.5}$ emission factors. According to the local CDC, the field sites in Yunnan Province, Fuyuan and Xuanwei Counties, all have a high occurrence of lung cancer.

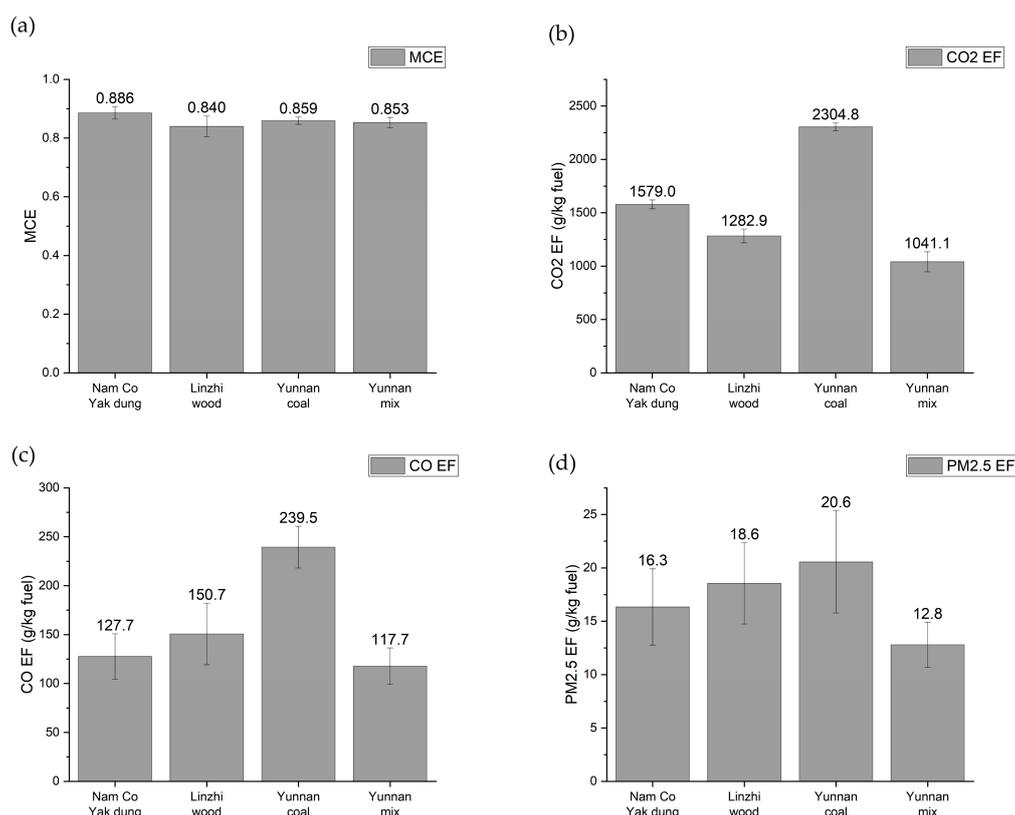


Figure 9. Comparison of emission parameters between sites and fuels: (a) Modified combustion efficiency (MCE); (b) Emission factor for CO_2 ; (c) Emission factor for CO ; (d) Emission factor for $PM_{2.5}$.

4.2. Carbon Particulate Emission

EC/OC particulate measurements are inherently noisier than gas measurements and total particulate measurements because they are often less uniformly mixed at the sampling zone (the complete mixing into the atmosphere occurs over a longer time). Sample numbers of EC and OC were also much smaller than for the other species. Nevertheless, the EC/OC data are a critical

component of the uncertainty in climate forcing, so it is important to include even the limited validated data obtained. The summary of EC and OC measurement is shown in Table 5.

Table 5. Summary of elemental carbon and organic carbon results.

Location	Fuel	Sample Amount	EF EC (g/kg Fuel)	EF OC (g/kg Fuel)
Tibet, China	Yak Dung	10	0.25 ± 0.05	15.41 ± 2.54
Tibet, China	Wood	2	0.11 ± 0.05	16.03 ± 14.48
Yunnan, China	Coal	16	1.46 ± 0.47	10.09 ± 2.71
Yunnan, China	Mix	18	0.51 ± 0.17	7.02 ± 1.26

Figure 10 shows a comparison of EC and OC emission factors through all field sites and fuels. The coal stoves in Yunnan, China, produced the highest elemental carbon emissions, even considering its high uncertainty, while other stoves burning agricultural-based fuel had a similar result. A plot of the EC and OC ratio for the different regions and fuel combinations is shown in Figure 11, as this ratio is usually of more interest.

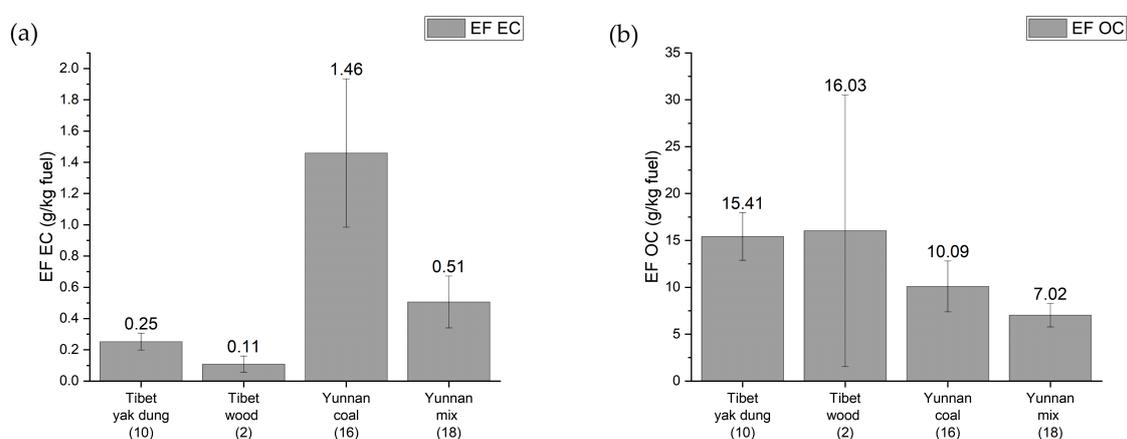


Figure 10. Comparison of carbon particulate emission factor between sites and fuels: (a) Elemental carbon; (b) Organic carbon.

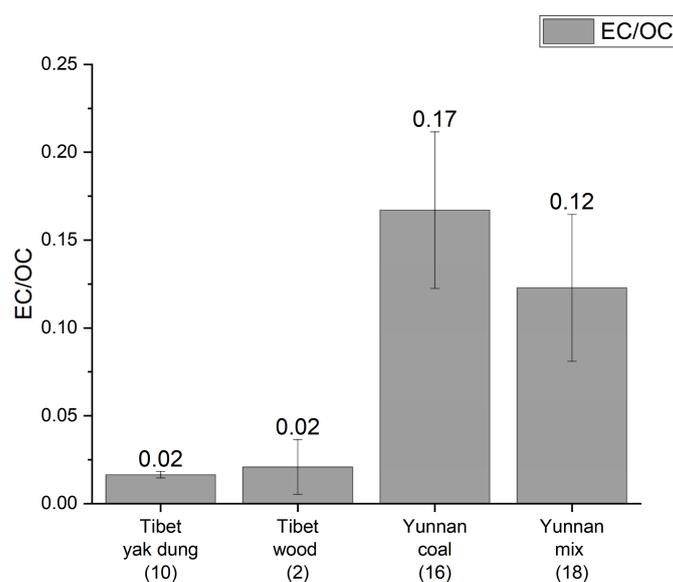


Figure 11. Comparison of the EC/OC ratios between sites and fuels.

Elemental carbon emission relates closely to the carbon content in the fuel: a higher carbon fuel such as coal produces more EC during the combustion process. From the perspective of combustion chemistry, the formation of soot, which is mostly elemental carbon, is largely related to the production of acetylene (C_2H_2). The agricultural-based fuels (wood, dung, agricultural residue, etc.) essentially consist of lingo-cellulosic materials, with hydrogen, carbon, and oxygen the dominate elements. In the combustion process, before soot (EC) is produced, the long carbohydrate polymers (larger C number) break up into shorter carbohydrates (smaller C number) until acetylene. For a stove that does not burn fuel completely (which is common for cooking stoves), the decomposition reactions for some molecules can stop before reaching the acetylene stage. In this case, organic carbon is emitted. Elemental carbon, on the other hand, means that reactions reach the key soot precursor acetylene, which represents more complete combustion as compared to high OC emission combustion.

5. Conclusions

A series of in-field emission measurements for solid fuel cooking stoves has been conducted. Emission factors for major compounds, including CO_2 , CO , $PM_{2.5}$, CH_4 , and elemental and organic carbon, with their statistical uncertainties were calculated using the carbon balance method. The acquired emission database fills in the inventory for residential cooking stoves' emission in the corresponding regions. These unique data from in-field measurements showed the real variability of cooking stove use and caution against simple methods to incorporate cooking stove emissions in global climate models. The results show clearly that in average, real-life cooking, stove use emits more than occurs under laboratory and controlled conditions.

The result from these field measurements show that fuel type is a critical variable in cooking stove performance. However, the effect of stove activity, such as the fluctuation during the combustion process, and the start and stop stages, should be considered in modeling and designing controlled laboratory experiments.

Author Contributions: Methodology, J.D.; formal analysis, J.D.; investigation, J.D., A.D., C.L., Q.Z., P.C., S.K., and J.L.; resources, J.D., A.D., C.L., Q.Z., P.C., S.K., and J.L.; data curation, J.D., A.D., and J.L.; writing, original draft preparation, J.D.; writing, review and editing, J.D., A.D., and D.D.-R.; visualization, J.D., and A.D.; supervision, D.D.-R.; project administration, A.D.

Funding: This research has been supported by a grant from the U.S. Environmental Protection Agency (EPA)'s Science to Achieve Results (STAR) through its Office of Research and Development in the research described here under Grant Number 83503601. It has not been subject to Agency review and therefore does not necessarily reflect the views of the U.S. EPA. The contents are solely the responsibility of the authors. No official endorsement should be inferred. The mention of trade names or commercial products in the publication does not constitute endorsement or recommendation for use.

Acknowledgments: We would like to acknowledge our collaborators from the University of Illinois at Urbana-Champaign, Tami Bond, Cheryl L. Weyant, and Ryan Thompson, and their research support staff for their significant contributions in the implementation, execution, and sample analyses. Many thanks to Rufus Edwards for his supervision and funding acquisition. Special acknowledgments to Kirk Smith for his original conceptualization and research design that laid the foundation and groundwork for this study. Additional acknowledgments to Qing Lan, Nathaniel Rothman, and Wei Hu for facilitating field measurements in Yunnan Province, China. We would also like to thank the following collaborators: the Institute of Tibetan Plateau Research, Chinese Academy of Sciences, and the Center for Disease Control and Prevention (CDC) of Qujing City for their assistance in the field. Special thanks to Allison Mok, Harman Chauhan, Vy Pham, Stephanie Fong, Kunaal Kapoor, and Scott Ondap for their contributions.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; nor in the decision to publish the results.

References

1. Bruce, N.; Rehfuess, E.; Mehta, S.; Hutton, G.; Smith, K. Indoor air pollution. In *Disease Control Priorities in Developing Countries*, 2nd ed.; Jamison, D.T., Breman, J.G., Measham, A.R., Alleyne, G., Claeson, M., Evans, D.B., Jha, P., Mills, A., Musgrove, P., Eds.; World Bank: Washington, DC, USA, 2006.
2. Jetter, J.J.; Kariher, P. Solid-fuel household cooking stoves: Characterization of performance and emissions. *Biomass Bioenergy* **2009**, *33*, 294–305. [[CrossRef](#)]
3. Bond, T.C.; Bhardwaj, E.; Dong, R.; Jogani, R.; Jung, S.; Roden, C.; Streets, D.G.; Trautmann, N.M. Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000. *Glob. Biogeochem. Cycles* **2007**, *21*, GB2018. [[CrossRef](#)]
4. Turns, S.R. *An Introduction to Combustion, Concepts and Applications*; McGraw Hill: New York, NY, USA, 2012.
5. DOE—Fossil Energy: A Brief History of Coal Use in the United States; Department of Energy: Washington, DC, USA, 2019.
6. Smith, K.R.; Apte, M.G.; Yuqing, M.; Wongsekiarttirat, W.; Kulkarni, A. Air pollution and the energy ladder in asian cities. *Energy* **1994**, *19*, 587–600. [[CrossRef](#)]
7. Ahrenfeldt, J.; Thomsen, T.P.; Henriksen, U.; Clausen, L.R. Biomass gasification cogeneration—A review of state of the art technology and near future perspectives. *Appl. Therm. Eng.* **2013**, *50*, 1407–1417. [[CrossRef](#)]
8. Subramanian, R.; Winijkul, E.; Bond, T.C.; Thiansathit, W.; Oanh, N.T.K.; Paw-Armart, I.; Duleep, K. Climate-relevant properties of diesel particulate emissions: results from a piggyback study in Bangkok, Thailand. *Environ. Sci. Technol.* **2009**, *43*, 4213–4218. [[CrossRef](#)] [[PubMed](#)]
9. Maricq, M.M.; Podsiadlik, D.H.; Chase, R.E. Examination of the size-resolved and transient nature of motor vehicle particle emissions. *Environ. Sci. Technol.* **1999**, *33*, 1618–1626. [[CrossRef](#)]
10. Yan, F.; Winijkul, E.; Jung, S.; Bond, T.C.; Streets, D.G. Global emission projections of particulate matter (PM): I. Exhaust emissions from on-road vehicles. *Atmos. Environ.* **2011**, *45*, 4830–4844. [[CrossRef](#)]
11. Bond, T.C.; Doherty, S.J.; Fahey, D.W.; Forster, P.M.; Berntsen, T.; DeAngelo, B.J.; Flanner, M.G.; Ghan, S.; Kärcher, B.; Koch, D.; et al. Bounding the role of black carbon in the climate system: A scientific assessment. *J. Geophys. Res. Atmos.* **2013**, *118*, 5380–5552. [[CrossRef](#)]
12. Johnson, M.; Edwards, R.; Alatorre Frenk, C.; Masera, O. In-field greenhouse gas emissions from cookstoves in rural Mexican households. *Atmos. Environ.* **2008**, *42*, 1206–1222. [[CrossRef](#)]
13. Zhang, J.; Smith, K.R. Hydrocarbon emissions and health risks from cookstoves in developing countries. *J. Expo. Anal. Environ. Epidemiol.* **1995**, *6*, 147–161.
14. Zhang, J.; Smith, K.R.; Uma, R.; Ma, Y.; Kishore, V.V.N.; Lata, K.; Khalil, M.A.K.; Rasmussen, R.A.; Thorneloe, S.T. Carbon monoxide from cookstoves in developing countries: 1. Emission factors. *Chemosph. Glob. Chang. Sci.* **1999**, *1*, 353–366. [[CrossRef](#)]
15. Zhang, J.; Smith, K.R. Emissions of carbonyl compounds from various cookstoves in China. *Environ. Sci. Technol.* **1999**, *33*, 2311–2320. [[CrossRef](#)]
16. Lamarque, J.F.; Bond, T.C.; Eyring, V.; Granier, C.; Heil, A.; Klimont, Z.; Lee, D.; Liousse, C.; Mieville, A.; Owen, B.; et al. Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: Methodology and application. *Atmos. Chem. Phys.* **2010**, *10*, 7017–7039. [[CrossRef](#)]
17. Bergamaschi, P.; Hein, R.; Heimann, M.; Crutzen, P.J. Inverse modeling of the global CO cycle: 1. Inversion of CO mixing ratios. *J. Geophys. Res. Atmos.* **2000**, *105*, 1909–1927. [[CrossRef](#)]
18. Kasibhatla, P.; Arellano, A.; Logan, J.A.; Palmer, P.I.; Novelli, P. Top-down estimate of a large source of atmospheric carbon monoxide associated with fuel combustion in Asia. *Geophys. Res. Lett.* **2002**, *29*, 6. [[CrossRef](#)]
19. Gros, V.; Williams, J.; Lawrence, M.; Von Kuhlmann, R.; Van Aardenne, J.; Atlas, E.; Chuck, A.; Edwards, D.; Stroud, V.; Krol, M. Tracing the origin and ages of interlaced atmospheric pollution events over the tropical Atlantic Ocean with in situ measurements, satellites, trajectories, emission inventories, and global models. *J. Geophys. Res. Atmos.* **2004**, *109*. [[CrossRef](#)]
20. Streets, D.G.; Zhang, Q.; Wang, L.; He, K.; Hao, J.; Wu, Y.; Tang, Y.; Carmichael, G.R. Revisiting China's CO emissions after the transport and chemical evolution over the Pacific (TRACE-P) mission: Synthesis of inventories, atmospheric modeling, and observations. *J. Geophys. Res. Atmos.* **2006**, *111*. [[CrossRef](#)]

21. Venkataraman, C.; Habib, G.; Kadamba, D.; Shrivastava, M.; Leon, J.F.; Crouzille, B.; Boucher, O.; Streets, D. Emissions from open biomass burning in India: Integrating the inventory approach with high-resolution Moderate Resolution Imaging Spectroradiometer (MODIS) active-fire and land cover data. *Glob. Biogeochem. Cycles* **2006**, *20*. [[CrossRef](#)]
22. Koch, D.; Bond, T.C.; Streets, D.; Unger, N.; Van der Werf, G.R. Global impacts of aerosols from particular source regions and sectors. *J. Geophys. Res. Atmos.* **2007**, *112*. [[CrossRef](#)]
23. Roden, C.A.; Bond, T.C.; Conway, S.; Osorto Pinel, A.B.; MacCarty, N.; Still, D. Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves. *Atmos. Environ.* **2009**, *43*, 1170–1181. [[CrossRef](#)]
24. Zhang, J.; Smith, K.R.; Ma, Y.; Ye, S.; Jiang, F.; Qi, W.; Liu, P.; Khalil, M.A.K.; Rasmussen, R.A.; Thorneloe, S.A. Greenhouse gases and other airborne pollutants from household stoves in China: A database for emission factors. *Atmos. Environ.* **2000**, *34*, 4537–4549. [[CrossRef](#)]
25. Smith, K.R.; Khalil, M.A.K.; Rasmussen, R.A.; Thorneloe, S.A.; Manegdeg, F.; Apte, M. Greenhouse gases from biomass and fossil fuel stoves in developing countries: A Manila pilot study. *Chemosphere* **1993**, *26*, 479–505. [[CrossRef](#)]
26. Bhattacharya, S.C.; Albina, D.O.; Abdul Salam, P. Emission factors of wood and charcoal-fired cookstoves. *Biomass Bioenergy* **2002**, *23*, 453–469. [[CrossRef](#)]
27. Taylor, R.P. The Uses of Laboratory Testing of Biomass Cookstoves and the Shortcomings of the Dominant U.S. Protocol. Ph.D. Thesis, Iowa State University, Ames, IA, USA, 2009.
28. Bond, T.C.; Streets, D.G.; Yarber, K.F.; Nelson, S.M.; Woo, J.H.; Klimont, Z. A technology-based global inventory of black and organic carbon emissions from combustion. *J. Geophys. Res. Atmos.* **2004**, *109*, D14203. [[CrossRef](#)]
29. Roden, C.A.; Bond, T.C.; Conway, S.; Pinel, A.B.O. Emission factors and real-time optical properties of particles emitted from traditional wood burning cookstoves. *Environ. Sci. Technol.* **2006**, *40*, 6750–6757. [[CrossRef](#)] [[PubMed](#)]
30. Bond, T.C.; Anderson, T.L.; Campbell, D. Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols. *Aerosol Sci. Technol.* **1999**, *30*, 582–600. [[CrossRef](#)]
31. Crutzen, P.J.; Heidt, L.E.; Krasnec, J.P.; Pollock, W.H.; Seiler, W. Biomass burning as a source of atmospheric gases CO, H₂, N₂O, NO, CH₃Cl and COS. *Nature* **1979**, *282*, 253–256. [[CrossRef](#)]
32. Ward, D.E.; Hao, W.M.; Susott, R.A.; Babbitt, R.E.; Shea, R.W.; Kauffman, J.B.; Justice, C.O. Effect of fuel composition on combustion efficiency and emission factors for African savanna ecosystems. *J. Geophys. Res. Atmos.* **1996**, *101*, 23569–23576. [[CrossRef](#)]
33. Lipsky, E.M.; Robinson, A.L. Design and evaluation of a portable dilution sampling system for measuring fine particle emissions. *Aerosol Sci. Technol.* **2005**, *39*, 542–553. [[CrossRef](#)]
34. *NIOSH Manual of Analytical Methods (NMAM)*, 4th ed.; Third Supplement; NIOSH: Washington, DC, USA, 2018. [[CrossRef](#)]
35. Gaur, S.; Reed, T.B. *Thermal Data for Natural and Synthetic Fuels*; CRC Press: Boca Raton, FL, USA, 1998.



© 2019 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).