

Manuscript Number:

Title: Chemical characterization of biomass fuel particulate deposits and ashes in households of Mt Everest region (NEPAL)

Article Type: Research Paper

Keywords: PM indoor, ash, remote area, PAHs, metals

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Abstract: During a sampling campaign, carried out during June 2012, inside some traditional households located in four villages (Phakding, Namche, Pangboche and Tukla) of Mt. Everest region in southern part of the central Himalaya (Nepal), particulate matter (PM) depositions and ashes have been collected. Moreover, outdoor PM depositions have also been analyzed. Chemical characterization of PM depositions and ashes for major ions, organic carbon, elemental carbon (EC), metal content and PAHs (Polycyclic Aromatic Hydrocarbons) allowed identifying, as major contributors to indoor PM, the following sources: biomass burning, cooking and chimney ashes. These sources significantly affect outdoor PM depositions: in-house biomass burning is the major source for outdoor EC and K+ as well as biomass burning and cooking activities are the major sources for Polycyclic Aromatic Hydrocarbons.

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To the Editor of Science of the Total Environment

Dear Editor,

coauthors and me would like to submit to your attention our manuscript entitled: *Chemical characterization of biomass fuel particulate deposits and ashes in households of MT Everest region (NEPAL)*.

In our study the chemical composition of indoor deposits and ashes generated by the combustion of different fuels in household stoves of four villages of a high elevated Central Himalayan region has been obtained. Indoor deposits and ashes compositions have been compared with corresponding outdoor deposits. Chemical characterization on the samples collected in this study suggested that biomass burning in house heating and cooking activities are relevant source affecting significantly particulate outdoor depositions. In fact we found that OC is the more abundant component of indoor particles; moreover OC average indoor concentration is more of double of the OC average outdoor one. Among analyzed cations, K^+ and Ca^{2+} were the more abundant ones in the indoor and ash samples. K^+ and Ca^{2+} average indoor and ash concentrations were more than three times of K^+ and Ca^{2+} average outdoor ones. Among compounds forming OC, phase particulate PAHs associated to cooking activities and biomass burning have relevant concentrations in indoor samples: average indoor concentration of chrysene is significantly higher than outdoor and ashes samples.

The field studies summarized in our manuscript are related to the atmosphere, anthroposphere and biosphere: in fact the manuscript concerns indoor and outdoor air quality. Moreover we found that poor air quality was due to anthropogenic activities such as: cooking activities and biomass burning, which effects can affect all the biosphere.

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Lecce 10/06/2016

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A handwritten signature in black ink, appearing to read 'Pierina Ielpo', written in a cursive style.

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Highlights:

OC, EC, PAHs, elements, anions and cations quantitatively analysed in particulate deposits and ashes collected in the Mt Everest region.

Indoor particulate deposits composition compared with outdoor particulate deposits one.

The OC average indoor concentration is more of double of the OC average outdoor one.

In indoor and ash samples PAH concentrations were higher than the outdoor ones, suggesting that the main source of PAHs in outdoor deposits is the biomass combustion.

Biomass burning is the largest contributor of K^+ to atmosphere particulate matter.

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1 **Chemical characterization of biomass fuel particulate deposits and ashes in**
2 **households of Mt Everest region (NEPAL)**

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30

31 **Abstract**

32 During a sampling campaign, carried out during June 2012, inside some traditional households located in four
33 villages (Phakding, Namche, Pangboche and Tukla) of Mt. Everest region in southern part of the central Himalaya
34 (Nepal), particulate matter (PM) depositions and ashes have been collected. Moreover, outdoor PM depositions have
35 also been analysed. Chemical characterization of PM depositions and ashes for major ions, organic carbon, elemental
36 carbon (EC), metal content and PAHs (Polycyclic Aromatic Hydrocarbons) allowed identifying, as major contributors to
37 indoor PM, the following sources: biomass burning, cooking and chimney ashes. These sources significantly affect
38 outdoor PM depositions: in-house biomass burning is the major source for outdoor EC and K⁺ as well as biomass
39 burning and cooking activities are the major sources for Polycyclic Aromatic Hydrocarbons.

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42 **Highlights:**

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44 Mt Everest region.

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48 PAHs in outdoor deposits is the biomass combustion.

49 Biomass burning is the largest contributor of K⁺ to atmosphere particulate matter.

50 **Keywords:** PM indoor, ash, remote area, PAHs, metals

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53 **1. Introduction**

54 Indoor air pollution is a major cause of ill health in developing countries. In some regions it is mostly due to the burning
55 of biomass fuel, particularly wood, dung, straw, and charcoal (“solid fuels”), used as a source of heat and light
56 (Rehfuess, 2006; Fullerton et al., 2008; Salerno et al., 2010b; Gurung and Bell, 2013). Because of incomplete
57 combustion of biomass fuels, indoor air concentrations of PM₁₀ can be up to 10000 µg/m³ during cooking (Rehfuess,
58 2006).

59 Globally, solid fuel use is estimated to cause 3.5 million premature deaths per year, around one million of which are
60 attributed to acute respiratory infections in young children (Stevens, 2009; Lim et al., 2012; Murray et al., 2012). There
61 is also strong evidence of linking of solid fuel use with chronic obstructive pulmonary disease (Kurmi et al., 2010),
62 pneumonia in children under 5 (Dherani et al., 2008), lung cancer (Kurmi et al., 2012), and tuberculosis (Sumpter and
63 Chandramohan, 2013). There is also weaker evidence for a link with low birthweight (Pope et al., 2010, Shah and
64 Balkhair, 2011), anemia and stunting (Rehfuess, 2006; Fullerton et al., 2008). Air pollution affects also cardiovascular
65 systems (Murray and Lopez, 1996; Nishtar, 2002; Lim et al., 2012; Yamamoto et al., 2014). Lack of research on
66 exposure to air pollution and human health burden, despite poor air quality, is a situation common in many countries.
67 Nepal is one of these (Shrestha and Shrestha, 2005). In the review of Gurung and Bell (2013), it’s been shown as Nepal
68 is observed to have research gaps that include understanding chronic effects of air pollution exposure, risk associated

69 with pollution mixture such as constituents of PM and vulnerability of sub-population. A recent study (Bruno et al,
70 2013) has shown as in rural Nepalese high altitude dwellers, using biomass fuels, absence of chimney in the stoves is
71 associated with selective impairment of conduit-artery endothelial function. It has been observed that replacing
72 traditional stoves with improved stoves in rural house-holds led to decrease in total suspended particles, carbon
73 monoxide, and PM_{2.5} (Reid et al.,1986; NPHO, 2008;.)

74 In addition to the health effects, particulate matter resulting from incomplete combustion of biomass and fossil fuel
75 containing high level of black carbon (BC), affects climate forcing. In South Asia, BC emission from residential
76 biofuels cooking is the largest source of atmospheric BC concentrations (Venkataraman et al., 2005; Bond et al., 2007;
77 Gustafsson et al., 2009). The high levels of biomass BC emissions can significantly affect climate forcing from local to
78 global scales (Forster et al., 2007; Ramanathan and Carmichael, 2008). Furthermore BC is estimated to contribute to the
79 disruption of the monsoon in South Asia (Ramanathan et al., 2001, 2005; Lau et al., 2008) as well as East Asia (Menon
80 et al., 2002) and heating of the elevated regions of the Himalayan-Tibetan region (Ramanathan et al., 2007; Flanner et
81 al., 2009; Menon et al., 2010). Although the importance of biomass burning in the Himalayan region is well recognized,
82 precise data are lacking and only recently efforts have been made to assess the contribution of this source to aerosol
83 (Vadrevu et al., 2012). Other studies carried out at Askole, Pakistan Northern Areas, highlighted as domestic
84 combustion from the nearby village of Askole could represent a possible source of short lasting pollution events with
85 high aerosol concentration (Putero et al., 2014).

86 The aim of the present study is a preliminary chemical characterization of particulate matter (PM) generated indoor
87 from the combustion of different fuels used in traditional stoves and assessing how this source can affects outdoor PM
88 composition. For this purpose, a sampling campaign has been carried out during June 2012 in order to collect PM
89 depositions and ashes inside traditional homes of the southern slopes of Mt Everest (Nepal). Currently, no information
90 about chemical characterization of PM indoor exists in the high elevated Central Himalayan region where the
91 remoteness and the harsh conditions of the region have complicated and obstructed monitoring and sampling.

92 **2. Region of investigation**

93 The current study is focused on the Mt. Everest region, and in particular on the Sagarmatha National Park (SNP) and
94 the Buffer Zone (BZ) (27.75° to 28.11° N; 85.98° to 86.51° E) that lies in eastern Nepal, in the southern part of the
95 central Himalaya (Fig. 1) (Thakuri et al., 2014). The SNPBZ is the world's highest protected area, visited by over
96 30,000 tourists in 2008 (Salerno et al., 2010a; Salerno et al., 2013). The park area (1148 km²), extending from an
97 elevation of 2845 to 8848 m a.s.l. (i.e., Mt. Everest), covers the upper Dudh Koshi Basin (Manfredi et al., 2010; Amatya
98 et al., 2010), presents a broad range of bioclimatic conditions (UNEP and WCMC, 2008) and its climate is
99 characterized by the monsoon system, with a prevailing direction S-N and SW-NE (e.g., Salerno et al., 2015).

100 It is worth noting that in this area campaigns based on the use of conventional PM sampling systems are made difficult
101 by the extreme conditions (difficulties of transport and samplers management, lack of electricity etc.)

102 During June 2012, as already followed in four reference villages lying in a remote valley located in the northeast India
103 (Deka and Hoque, 2015) particulate matter depositions (powder depositions) and ashes were collected inside traditional
104 lodges in four reference villages (Phakding, Namche, Pangboche and Tukla) located in the Khumbu Valley along an

105 altitudinal gradient ranging from 2610 to 4600 m a.s.l. (Fig. 1). Figure 1 describes in details sampling locations, code
 106 and features of each sample including the fuel used.

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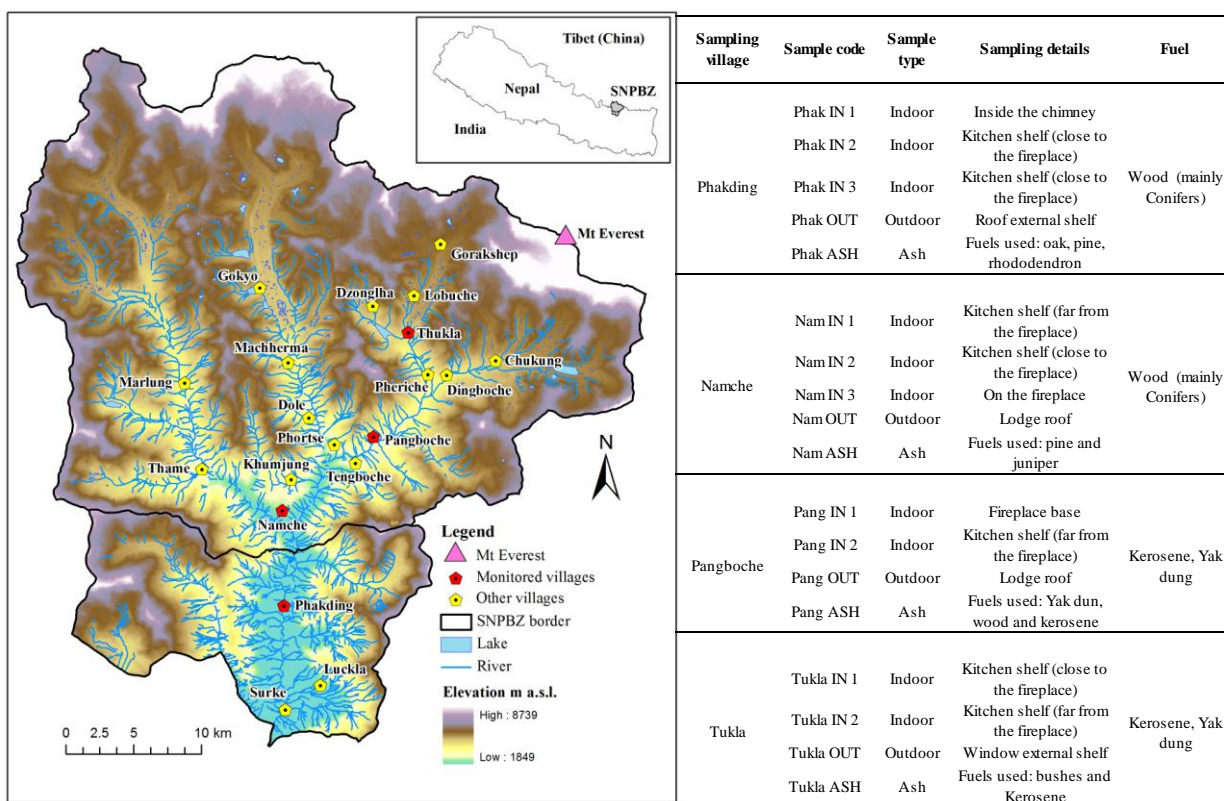
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119 **Fig. 1.** a) On the left, the map of the Sagarmatha National Park and Buffer Zone (SNPBZ) with topographic information
 120 and the four monitored villages. On the right, details on sampling sites.

121 As described by Salerno et al., 2010b, fuelwood from forests remains one of the major energy sources in SNPBZ,
 122 constituting 30% of all energy use. Kerosene is the most common energy source (33%), and dung (19%) and liquefied
 123 petroleum gas (LPG; 7.5%) are used less often. Energy is mainly used for activities such as cooking, boiling water,
 124 space heating, and lighting. Kitchens of most private houses in SNPBZ are equipped mainly with open fireplaces for
 125 cooking (and heating in winter), known as traditional cooking stoves. Due to the lack of suitable ventilation systems
 126 (chimney or other fume outlet), these facilities emit fumes directly into the kitchen area.

127 3. Materials and Methods

128 3.1 Sampling methods

129 It is well known that PM is generally collected using specific pumping low or high volume devices, but logistic aspects
 130 and lack of local electricity sources in this research activity did not allow to use these conventional samplers. As already
 131 followed in other remote or rural environments (Deka and Hoque, 2015), the sampling method used in the present study
 132 considered the collection of the PM powders without pumping. In particular, we collected indoor PM by means of a
 133 brush (for example in the case of the powder taken from the kitchen shelves) or using a spoon (as in the case of the ash).
 134 PM outdoor samples have been obtained collecting from several zones of the lodge roof (or window shelf) particulate
 135 deposits by a little spatula. For each sampling site, one integrated outdoor sample has been obtained and the chemical

136 concentrations shown are the results on three replicates analyses on it. Also in the case of ash samples, one integrated
137 ash sample per site has been obtained. For each village, a lodge equipped with traditional stoves has been selected. Each
138 reference site has been characterized sampling the fuel ash, the indoor depositions in two or three places, and the
139 outdoor deposition (Fig. 1). We considered the ashes and deposited dusts as chemical-physical composition assimilated
140 to the inhalable fraction of particulate matter one.

141

142 3.2 Chemical characterization

143 PM and ash samples have been characterized from the chemical point of view as concerns major ions (Na^+ , NH_4^+ , K^+ ,
144 Mg^{2+} , Ca^{2+} , Cl^- , NO_3^- , SO_4^{2-}), OC (Organic Carbon), EC (Elemental Carbon), elements (Li, Be, B, Al, V, Cr, Mn, Fe,
145 Co, Ni, Cu, Zn, As, Sb, Cs, Ba, U, Na, Mg, K, Ca, Rb, Sr) and PAHs (naphtalene, acenaphthene, fluorene,
146 phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b+j]fluoranthene,
147 benzo[k]fluoranthene, benzo[a]pyrene, dibenzo(a,h)anthracene, benzo[g,h,i]perylene and indeno[1,2,3-c,d]pyrene).

148 3.2.1 SEM-EDS analyses

149 The powder deposits were analyzed by SEM-EDS (electron microscopy coupled to an energy dispersive spectrometer)
150 in order to observe the surface morphology and to study the chemical composition. The instrument used was a Hitachi
151 TM1000 equipped with an energy dispersive X-ray spectrometer (Oxford Instruments Swift ED). The spectra were
152 acquired directly on a small portion of powder attached to the sample holder by adhesive tape.

153 3.2.2 Inorganic components analyses

154 An ICS-1000 Ion Chromatograph (Dionex) was used for the water-soluble inorganic constituents determination (NO_3^- ,
155 SO_4^{2-} , F^- , Cl^- , NO_2^- , Br^- , NH_4^+ , Na^+ , K^+ and Ca^{2+}). The samples, about 20 mg, were dissolved in 10 mL of MQ water.
156 The set-up of the extraction procedure is described in detail in Fermo et al. 2006a and Piazzalunga et al. 2013a. All
157 reagents were of analytical grade (Fluka, Milwaukee, WI, USA). Ultrapure water was produced by a Milli-Q system
158 (Millipore, Bedford, MA, USA). All the ions concentrations determined were higher then the LOD for this technique
159 (Piazzalunga et al. 2013a).

160 3.2.3 Carbonaceous fraction analysis

161 For the analysis of the carbonaceous fraction of the powder deposits two different methodologies have been used. Total
162 carbon (TC) content has been estimated by both Thermogravimetric analyzer coupled with FT-IR (TGA/FTIR) and a
163 Carbon, Nitrogen, Hydrogen (CHN) analyzer. TC is given by the sum of OC (organic carbon) and EC (elemental
164 carbon), apart from a small quantity of CC (carbonatic carbon).

165 The quantification of OC and EC has been carried by TGA/FTIR as described in Fermo et al., 2006b. The
166 concentrations determined were higher then LOD (Piazzalunga et al. 2013).

167 CHN analyses were carried out by a CHNS/O Perkin Elmer 2400 Series II elemental analyzer using an accessory for the
168 analysis of solids.

169 3.2.4 Polycyclic Aromatic Hydrocarbons analysis

170 PAHs (polycyclic aromatic hydrocarbons) analyses have been performed by high performance liquid chromatography
171 with fluorimetric detection (HPLC-FLD) according to ISO 16362 (2005), which is specific for the determination of
172 PAHs by HPLC.

173 For the analysis about 30 mg of powder were treated. An ultrasonic bath extraction for 15 min was performed using 10
174 mL of dichloromethane (x3 times) and a solution of 6-methylchrysene 40 µg/L and 1-metilntracene as internal
175 standard. After this step the solution was reduced to known volume (1 mL) and filtered by PTFE 0.45 µm filter. Before
176 the step of evaporation 50 µL of dimethyl sulfoxide (DMSO) were added in order to retain in solution the lighter
177 hydrocarbons (DMSO has a higher boiling temperature). The column used for the analyses was Vydac 201 TP 52 (25
178 cm x 4,6 mm internal diameter). The concentrations reported were higher then LOD for this techniques.

179 *3.2.5 Elemental analysis by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS)*

180 Aliquots of about 60 mg of powder or ash were dissolved by acid attack, through a digestion procedure based on a
181 'controlled microwave' technique by PEEKEM mod. EXCEL 2000, using an acid solution (Fluka trace selected for
182 trace analysis reagents), of HNO₃:HCl:HF:H₂O in a 3:1:05:3 (v/v/v/v) ratio. Table SI1 (reported as Supplementary
183 Information (SI)) shows the digestion conditions including four steps (A, B, C and D). The operating conditions are
184 reported in Table SI2. Twenty-three chemical elements were quantified by ICP-MS. The concentrations reported were
185 higher then LOD for this techniques. Furthermore the recovery percentages for some analytes were determined
186 analyzing the certified reference material 'Road dust BCR-CRM_CW7'. In most of the cases (Co, Ni, Zn, As, Ba) the
187 recovery was higher than 90%.

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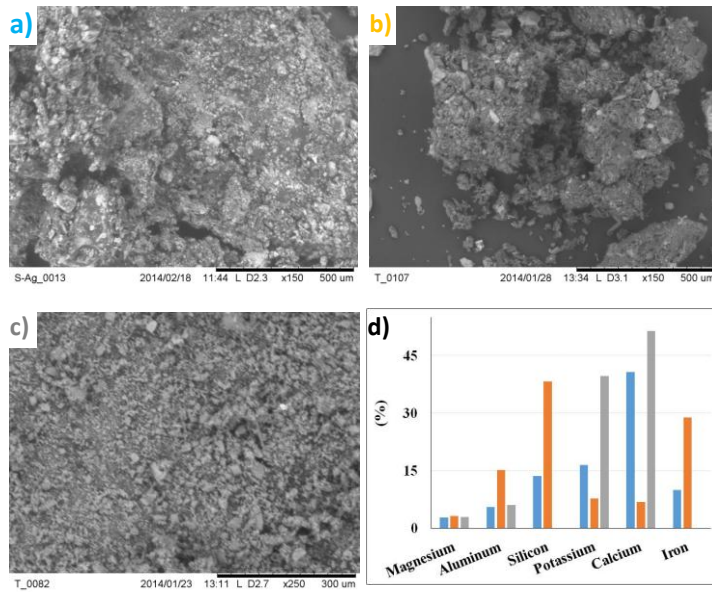
189 **4. Results and discussion**

190 *4.1 Morphology and chemical composition of particles*

191 In order to acquire information on the morphology, dimension and average bulk composition of the three kinds of
192 samples (indoor, outdoor and ash) SEM-EDS analyses have been performed. In Figure 2 the SEM images for three
193 samples collected at Phakding are reported. Comparing the images acquired on the indoor and outdoor samples with
194 those on the wood ashes it has been observed that indoor and outdoor powders are generally formed by larger particles
195 (Fig. 2a and Fig. 2b) with respects to what is observable for the wood ash (Fig. 2c). By EDS analyses (see Fig. 2d), it is
196 possible to put in evidence the major differences among the three kinds of samples. In particular it is evident how the
197 outdoor samples show a higher Si and Al content due to soil resuspension, while Ca is higher in the ash and the indoor
198 powder confirming the contribution of wood ash resuspension to the indoor samples; in fact, Ca and K are generally
199 present in high concentrations in wood ashes (Campbell, 1990; Misra et al., 1993; Nordin, 1994; Salam et al., 2013;
200 Deka and Hoque, 2015).

201 In the case of Pangboche from the comparison among indoor, outdoor and ash samples (see Fig. SI1a, Fig. SI1b and
202 Fig. SI1c) it is even more evident how the dimensions of the particles are lower for the ash (Fig. SI1c) and some of
203 these particles are distinguishable in the indoor samples (Fig. SI1a) where larger particles are predominant. From the
204 analyses carried out on the single particles the presence of heavy metals such as, for example, titanium, has been
205 disclosed (insert of Fig. SI1a).

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217 **Fig. 2.** SEM images for a) Phakding indoor sample (Phak IN 1), b) Phakding outdoor sample (Phak OUT), c) Phakding
218 wood ash sample (Phak ASH), d) percentage composition analyzed by SEM-EDS for Phak IN (blue), Phak OUT
219 (orange), Phak ASH (grey)

220 4.2 Organic and Elemental Carbon

221 Regarding OC and EC analytical determination, it is worth noting that the analysis of the two constituents in PM
222 samples is commonly carried out by a dedicated methodology, namely TOT (thermal optical transmittance) (Chow et
223 al., 1993, Piazzalunga et al., 2013a). However this technique requires that particulate matter is collected on a quartz
224 fiber filter, and so it cannot be here applied being our samples in form of powder deposits. Therefore an alternative
225 method, namely TGA-FTIR (Fermo et al., 2006b), has been employed.

226 Average indoor OC concentration (Table 1) shows higher value than outdoor and ashes one. Comparing OC values
227 found in our study with those obtained by Deka and Hoque, 2015 for biomass fuel smoke particles (BSFP) and by Alves
228 et al. analyzing smoke emission from biomass burning (Alves et al., 2010), we have found similar values. The
229 variability of EC values could be due to the variety of fuels employed in the stoves. In particular, average EC
230 concentration for the ashes is higher than indoor and outdoor ones and is similar to what obtained by Deka and Hoque,
231 2015. It is worth noting that EC is almost equivalent to BC (black carbon) that is generally determined by optical
232 methods (Massabò et al., 2015). As mentioned in the introduction BC in the Himalayan area has a high impact on both
233 climate change and environment. For these reasons, EC quantification is of great interest.

234 **Table 1.** Mass percentage of indoor, outdoor and ashes average values of organic carbon (OC), elemental carbon (EC)
235 and total carbon (TC), expressed in mass percentage, obtained by TGA-FT IR; the ranges of variation are shown in the
236 brackets. Together with TC, also hydrogen (H) and total nitrogen (TN) obtained by CHN analyzer are also reported.

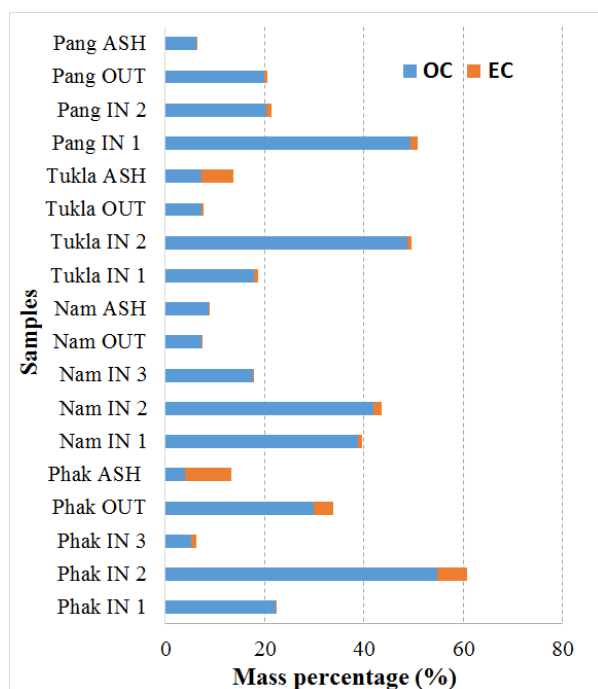
Analyzer	Parameters	Sample type		
		Indoor	Outdoor	Ashes
TGA/FTIR	organic carbon (OC)	46.8 ± 6.4 (39-55)	19.0 ± 11.3 (7-30)	5.1 ± 1.6 (4-6)
	elemental carbon (EC)	2.0 ± 2.2 (0.6-6)	1.7 ± 2.0 (0.5-4)	4.8 ± 6.5 (0.2-9)
	total carbon (TC)	48.8 ± 8.1 (40-61)	20.7 ± 13.1 (8-34)	9.9 ± 4.9 (6-13)
CHN	total carbon (TC)	43.5 ± 13.3	19.9 ± 11.8	4.50 ± 5.90
	hydrogen (H)	5.6 ± 2.2	2.5 ± 1.4	0.45 ± 0.07
	total nitrogen (TN)	1.11 ± 0.70	0.97 ± 0.7	0.10 ± 0.02

237

238 In order to validate the data obtained by TGA-FTIR, CHN analyses have been also performed in order to compare TC
 239 values (total carbon values, TC=OC + EC). In fact, while by TGA-FTIR the carbon chemical speciation is obtainable in
 240 terms of OC and EC, CHN allows quantifying total carbon only. The correlation between the two techniques, shown in
 241 Figure SI2, is quite fair.

242 Figure 3 shows the mass percentages for OC and EC for all collected samples. It is possible to note that ash samples
 243 show lower OC than indoor ones suggesting that there is an accumulation of ashes in the powder indoor deposits.
 244 Sample Phak IN 2 shows the higher EC mass percentage among indoor samples. In this case, the fuel is the same as that
 245 used at Namche (see Fig. 1) so the different ash composition could be due to different burning conditions and/or to the
 246 employment of a different stove. It is worth noting that the highest EC percentage is showed by the corresponding ash.
 247 In spite of the variability among the samples belonging to the same category (indoor, outdoor and ash), biomass burning
 248 in house heating and cooking activities is the major source for outdoor EC: it is to be taken in mind, in fact, the total
 249 absence in the investigated area of other sources, such as vehicular traffic. Referring to the literature Rehman et al.
 250 (2011) evaluating the amount of BC concentrations during cooking hours found that, in the Indo-Gangetic-Plains region
 251 of India, cooking based on the use of solid biomass fuels was a major source of ambient BC with indoor BC values
 252 during the peak hours much higher than the outdoor ones. This confirms our hypothesis on the high contribution of
 253 heating and cooking activities to EC emissions.

254



263

264 **Fig. 3.** Mass percentage of OC and EC detected for all samples.

265 In Table 2 some characteristic carbon mass ratios have been reported. Ratios between chemical species present in PM
 266 emissions are generally used to discriminate the sources of ambient particulate matter (Novakov et al., 2000, Saud et al.,
 267 2013). In particular high OC/EC ratios indicate the contribution of biomass burning (Saarikoski et al., 2008, Ram et al.,
 268 2010). In Table 2 the ratios coming from the literature and calculated for emissions from burning of the most
 269 widespread biomass fuel used for the cooking, are also reported. High value of the ratios OC/EC, as those determined in
 270 our study, are typical of biomass burning combustion, even if our values are likely higher with respect to those reported
 271 by Deka and Hoque, 2015. Nevertheless according to Deka and Hoque, 2015 the moisture content of biomass fuel
 272 influences OC/EC ratio in emitted particles and generally OC/EC ratio for different biomass fuel vary to a greater
 273 extend, which greatly depend on the fuel type and burning conditions. This may justify the great variability among the
 274 OC/EC values reported in Table 2. Furthermore it is worth to note that the highest values observed in the present study
 275 (namely 95 and 128) are in accordance with what reported by Alves (Alves et al., 2010) examining emissions from
 276 biomass burning and attributed to condensation process of large hydrocarbons. To our knowledge in the literature there
 277 are no reference values for OC/EC ratio from biomass burning source that relate to the Mt Everest region considered in
 278 our study. Also the other ratios reported in Table 2 show higher values than literature ones, in particular concerning
 279 K^+/EC ratio. Alves et al., 2010 and Saud et al., 2013 attribute significant amounts of K^+ to emissions from different
 280 types of biomass burning. The highest values determined in our case are due to lower EC. On the contrary the two ratios
 281 Cl^-/EC and SO_4^{2-}/EC are in agreement with what found by Deka and Hoque, 2015. It is important to underline that no
 282 information about chemical characterization for indoor PM deposits and chemical parameters ratios are present for the
 283 Mt Everest region considered in our study.

284 **Table 2.** Indoor OC/EC, Cl⁻/EC, SO₄²⁻/EC and K⁺/EC mass ratios. The standard deviations are reported in brackets.
 285

Sample code	Mass Ratios			
	OC/EC	Cl ⁻ /EC	SO ₄ ²⁻ /EC	K ⁺ /EC
Nam IN 1	50	0.2	0.2	3.8
Nam IN 2	27	0.1	0.4	4.3
Nam IN 3	95	7.2	13.3	143
Pang IN 1	36	1.4	0.1	2.4
Pang IN 2	22	1.6	1.6	4.8
Phak IN 1	128	7.1	8.9	130
Phak IN 2	9	-	-	0.3
Phak IN 3	5	0.5	1.7	45
Tukla-IN 1	20	3.6	0.8	14
Tukla-IN 2	75	1.6	0.8	2
Indoor average	47 (40)	2.3 (2.7)	2.8 (4.5)	35 (55)
Agricultural residue *	3.77 (0.39)	1.72 (0.15)	0.24 (0.03)	1.19 (0.18)
Fuel wood*	2.82 (0.21)	1.14 (0.12)	0.24 (0.05)	0.43 (0.04)
Dung cake*	16.20 (1.65)	1.40 (0.16)	0.24 (0.02)	0.29 (0.04)
CDMBF-D**	6.83	0.2	0.54	0.88
CDMBF-W**	4.36	0.56	0.77	0.66
Mediterranean*** shrubland		0.24		0.48

*Saud et al 2013; ** Deka et al 2015 (CDMBF-D = Cow dung and mixed biomass fuel in dry; CDMBF-W = Cow dung and mixed biomass fuel in wet season); ***Alves et al 2010

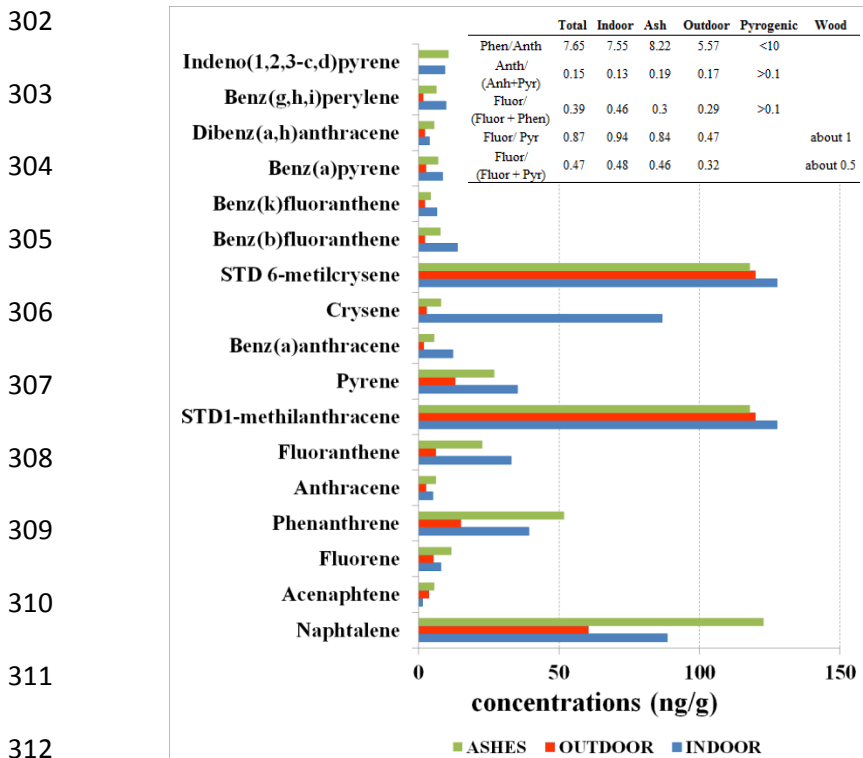
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288 4.3 Polycyclic Aromatic Hydrocarbons

289 It is well known that wood burning is a source of PAH (IARC 2010; Piazzalunga et al., 2011; Piazzalunga et al., 2013b)
 290 and this class of substances is of great interest to be analyzed because of their adverse effects on human health (IARC,
 291 2010). Observing Figure 4 it is interesting to note how the average indoor and ash concentrations for the 15 analysed
 292 PAHs, are very similar and higher than the outdoor values suggesting that biomass combustion is a source PAHs in the
 293 outdoor environment. A recent study carried out by Rajput et al. (2013) has shown that in the Northeastern Himalaya,
 294 an area not far from the SNPBZ, the average concentrations of PAHs sum in the atmospheric particulate matter was
 295 13.5 ng/m^3 with an B(a)P average value of 1.82 ng/m^3 for air mass TYPE I and 16.5 ng/m^3 with an B(a)P average
 296 concentration of 2.84 ng/m^3 for air mass TYPE II respectively, while the European Community has established a B(a)P
 297 limit value of 1 ng/m^3 that must not be exceeded.

298 It is worth to notice that in our study the sampling campaign has been carried out during June, so lower PAH
 299 concentrations are expected with respect to those that would be registered in wintertime. In another study (Chen et al.,
 300 2014), carried out on the Chinese side of the Tibetan Plateau, a more distant area, the PAH sum average concentrations
 301 ranged between 0.06 and 2.53 ng/m^3 , with the higher values during the autumn and winter seasons.



313 **Fig. 4.** Indoor, outdoor and ash PAHs average concentrations; in the inset: principal PAHs diagnostic ratio as source
 314 markers (average values).

315 Since in our case it was not possible to perform an active sampling of the aerosol particulate matter, as a consequence it
 316 was not possible to make a direct comparison with the PAHs concentrations reported in these literature studies.
 317 Nevertheless an estimation could be tried: considering that in our case PAH concentrations in indoor and ash samples
 318 are more than 3 times the outdoor values, on the base of the data reported for PAHs outdoor concentration in the same
 319 area, we can expect indoor concentrations, which far exceed the limits set for the protection of human health.

320 As regards PAHs profile (Fig. 4) the average indoor concentration of chrysene is substantially higher than the ash and
321 outdoors ones; it is worth to notice that high emissions of chrysene are attributable to the cooking source (IARC, 2012).
322 In the inset in Figure 4 principal diagnostic ratios for determined PAHs are showed. Comparing our values with some
323 reference values reported in the literature (Rajput et al., 2013) a pyrogenic source can be assumed for PAH in the
324 particulate phase; moreover fluoranthene/pyrene and fluoranthene/fluoranthene+pyrene ratios indicate wood as
325 prevalent source in indoor deposits and ashes, while in the outdoor samples the ratios are little lower than 1 for
326 fluoranthene/pyrene and lower than 0.5 for fluoranthene/fluoranthene+pyrene indicating probably the contribution of
327 other combustion sources. It is to be notice, in fact, that in the Namche village there is a dump where the wastes are
328 burnt.

329 *4.5 Inorganic constituents and trace elements*

330 As concerns the inorganic constituents Ca^{2+} and K^+ (Figure SI4a) are the more abundant cations in the indoor samples
331 in accordance with what found by other authors analyzing biomass fuel smoke particles (Deka and Hoque, 2015). In
332 several works K^+ and Ca^{2+} are considered as major contributions to biomass burning particulate or deposits (Salam et al,
333 2013, Deka and Hoque, 2015) and K^+ is often used as marker of wood (biomass) smoke. $\text{K}^+_{\text{indoor}}/\text{K}^+_{\text{outdoor}}$ ratio found in
334 our study was 4.4 suggesting that in house biomass burning is the largest contributor of potassium atmospheric
335 concentration. Moreover $\text{Ca}^{2+}_{\text{indoor}}/\text{Ca}^{2+}_{\text{outdoor}}$ ratio was bigger than 1 (namely 2.9). Even though Ca does not have
336 harmful effect, it plays a significant role on rain, water as well as soil chemistry for the ionic balance.

337 For the anions in the case of biomass fuel smoke particles the two main observed species are generally chloride and
338 sulphate (Deka and Hoque, 2015). In our samples sulphate average concentrations (Figure SI4a) have lower average
339 value in the outdoor samples with respect to the indoor and ash one suggesting an indoor relevant source for this specie,
340 according to elemental emissions by biomass burning (Nordin, 1994). Indoor, outdoor and ash elements average
341 concentrations are shown in Table SI3.

342 Chromium, cobalt, nickel, arsenic, copper, zinc and barium are among of the more toxic elements analyzed. In fact Cr
343 and As are included in U.S. Environmental Protection Agency (USEPA) and IARC (IARC) as ‘known’ or ‘probable’
344 human carcinogens. In particular reactive oxygen species (ROS) production and oxidative stress play a key role in the
345 toxicity and carcinogenicity of trace elements such as As, Cd, Cr, Pb and Hg (Tchounwou et al., 2012). For these
346 elements average indoor concentrations are slight higher or higher than outdoor ones (Fig. SI5), except for arsenic and
347 barium, suggesting an indoor sources for most of them. Cr concentration was higher with respect to the other
348 anthropogenic elements such as Ni, Co and As confirming the results of Deka and Hoque 2015 showing that rural
349 kitchens are source of carcinogenic elements. Finally Zinc measures confirm that this metal is also emitted from
350 biomass burning and wood combustion (Nordin, 1994; Yamasoe et al., 2000, Deka and Hoque, 2015).

351 From the literature, it is known that calculation of enrichment factors (EF) values helps to determine whether a certain
352 element has additional or anthropogenic sources other than its major natural sources. Since iron (Fe) has been used as a
353 reference element for an EF evaluation, assuming that the contribution of its anthropogenic sources to the atmosphere is
354 negligible (Yaroshevsky, 2006), the EF calculation formula has been applied in our study as follows (1):

$$355 \quad EF = \frac{\left(\frac{X}{Fe}\right)_{air}}{\left(\frac{X}{Fe}\right)_{crust}} \quad \text{Eq. 1}$$

356 where X is the element's concentration in our samples. If the EF value approaches unity, then crustal sources are
 357 predominant. In general, an EF > 5 indicates that a large fraction of the element can be attributed to non-crustal or
 358 anthropogenic sources (Wu et al., 2007).

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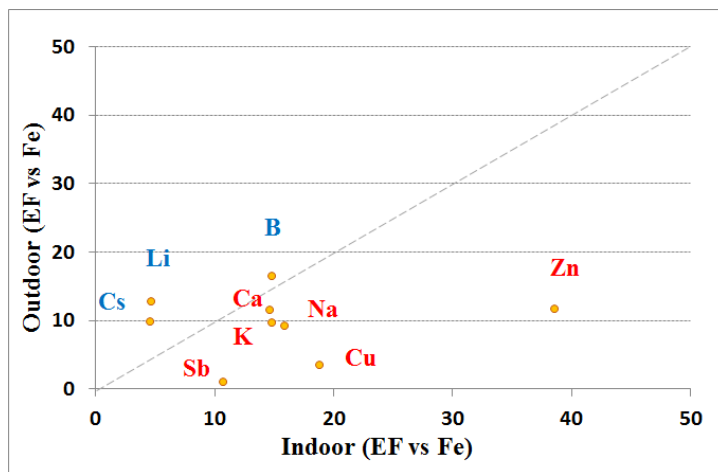
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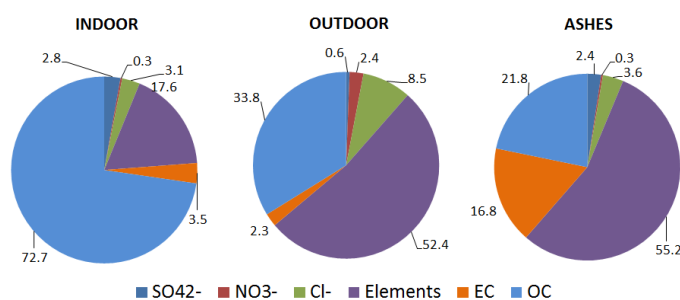
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367 **Fig. 5.** Enrichment factor (EF) bigger than 5 in the outdoor and indoor collected samples.

368 In Figure 5, where only EF bigger than 5 are shown, it is possible to note that K, Ca, Na, Zn and Cu show an EF much
 369 bigger than 5 for outdoor and indoor samples with outdoor values lower than indoor ones. This suggests for these
 370 elements a mixed origin, natural and anthropogenic. Moreover because EF for indoor samples is bigger than outdoor,
 371 for the indoor samples the anthropogenic sources are more relevant. The application of the enrichment factor approach
 372 confirms the importance of in-house biomass burning on particulate deposits.

373 Taking into account all the chemical constituents analytically determined on the samples (ash, indoor and outdoor), it
 374 has been possible to reconstruct the mass percentage composition as reported in Figure 6.



375

376 **Fig. 6.** Average relative contribution of analyzed species: Chloride (Cl⁻), Nitrate (NO³⁻), Sulphate (SO₄²⁻), Organic
 377 Carbon (OC), Elemental Carbon (EC) and elements (sum of elements analyzed).

378 OC is by far the more abundant component of indoor powder deposits followed by sum of elements and EC; for the
 379 outdoor particles the sum of elements is the more than measured indoor, followed by OC and Cl⁻. As discussed above,
 380 for outdoor samples Si and Al show higher values than for indoor samples, suggesting a considerable contribution of the
 381 soil resuspension for outdoor deposits. In the case of ash particles, the sum of elements is the more abundant

382 contribution followed by OC and EC. It is interesting to note as for ash particles EC average concentration is similar to
383 OC ones.

384 **Conclusions**

385 In our study, the chemical composition of indoor deposits and ashes generated by the combustion of different fuels
386 employed in household stoves in four villages of a high elevated Central Himalayan region has been obtained. In this
387 area the very extreme conditions (as lack of local electricity sources) do not allow sampling campaigns with
388 conventional systems. Indoor deposits and ashes compositions have been compared with corresponding outdoor
389 deposits. Chemical characterization on the samples collected in this study suggested that biomass burning in house
390 heating and cooking activities is a relevant source affecting significantly particulate outdoor depositions. OC, the more
391 abundant component of indoor particles, with an average indoor concentration double of the OC outdoor, indicate the
392 importance of in-house burning as source of OC at local level. . Among analyzed cations, K^+ and Ca^{2+} were the more
393 abundant ones in the indoor and ash samples. $K^+_{indoor}/K^+_{outdoor}$ ratio found in our study is bigger than 4 confirming the
394 important contribution of wood burning since potassium is a specific marker for this source. Some toxic anthropogenic
395 elements such as Cr, Ni, Co and As, Cr showed higher concentration levels with biggest average concentration in ash
396 samples and almost similar between indoor and outdoor samples, confirming the importance of these emissions in
397 possible diseases in the domestic environment.

398 Among the organic compounds, phase particulate PAHs associated to cooking activities and biomass burning have
399 relevant concentrations in indoor samples: indoor average concentration of PAHs whit bigger molecular weight are
400 generally three times higher than average outdoor ones and average indoor concentration of chrysene is particularly
401 higher than outdoor and ashes ones. Considering these results and, on the base of the data reported for PAHs outdoor
402 concentration in the same area, we can expect indoor concentrations, as $ng \times m^{-3}$, which far exceed the limits set for the
403 protection of human health.

404 A regular exposure to such high levels of PAHs and toxic elements such as Cr and As has an impact on health of people
405 mostly women and children that spend in indoor environment more time. For these reasons, more efforts in the studying
406 and investigations on this kind of rural environments would be needed. Moreover an improving of stove's technology
407 (William et al, 2014) used for cooking, lightening and heating activities can reduce the human exposure risk.

408 **Acknowledgments**

409 Authors wish to thank to Dr Lucia Valsecchi for his technical support during chemical analyses of samples.

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