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POP and PAH Contamination in the Southern Slopes of Mt. Everest (Himalaya, Nepal): Long-range Atmospheric Transport, Glacier Shrinkage, or Local Impact of Tourism?

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ABSTRACT

Due to their physico-chemical properties, POPs and PAHs are subjected to long-range atmospheric transport (LRAT) and may be deposited in remote areas. In this study, the contamination with DDX, PCBs, PBDEs, and PAHs was investigated in sediments and soils collected on the southern slopes of Mt. Everest (Himalaya, Nepal) in two different sampling campaigns (2008 and 2012). The results showed a limited contamination with POPs and PAHs in both soil and sediment samples. Therefore, the southern slopes of Mt. Everest can be considered a remote area in almost pristine condition. The LRAT mechanism confirmed its primary role in the transfer of contaminants to remote regions, while the gradual melting of glaciers, due to global warming, and the subsequent release of contaminants was suggested to be a secondary source of pollution of the lake sediments. In addition, the increase of tourism in this area during the last decades might have influenced the present concentrations of PAHs in the sediments and soils.

KEYWORDS

Persistent Organic Pollutants; Polycyclic Aromatic Hydrocarbons; Himalaya; Long-range atmospheric transport; Global warming

1. INTRODUCTION

Persistent organic pollutants (POPs), including DDT, polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs), are chemicals resistant to degradation processes and having the ability to be transported through long distances in the environment. Due to this ability, POPs have been detected also in places where they had never been used (Barra et al., 2005; Zhang et al., 2008). Although prohibitions on the uses of several POPs have been implemented in developing nations, some of them are still in use in South and South-East Asian countries, such as India (Gupta et al., 2012; Sharma et al., 2012; Sharma et al., 2014), Pakistan (Syed et al., 2014), Bhutan, Nepal, and Philippines (Gupta et al., 2012). In particular, even though DDT has been banned for agricultural uses, the National Malarial Program (NAMP) used 3750 tons of DDT in India for malaria control in 2001 (Gupta, 2004). In contrast, polycyclic aromatic hydrocarbons (PAHs) are not strictly considered POPs because most of them are not bio-accumulated by living organisms and some of them are also natural compounds, originating from the diagenic process of organic matter biodegradation. However, PAHs are compounds resistant to degradation and possess toxic properties; some of them have been reported to be mutagenic, carcinogenic and teratogenic (IARC, 1983). Therefore, they have been commonly included in the European Union priority pollutant list. Many human activities in India and China have led to the formation of PAHs, such as vehicle emissions, fuel use, industrial processes, electric production, and waste incineration (Wang et al., 2006).

Due to their physico-chemical properties, all these substances are subjected to long-range atmospheric transport (LRAT) and are deposited in remote areas including polar and high-altitude mountain regions (Daly and Wania, 2005; Wania and Mackay, 1993). The accumulation of POPs in cold regions may be driven by the process of cold condensation, promoted by low temperatures, and by falling snow as an efficient scavenger (Lei and Wania, 2004). Thus, high-altitude mountain regions, as well as the Poles, have received growing interest as receptor regions for long-range transported airborne contaminants (Tanabe et al., 1983; Hisato et al., 1994; Bidleman et al., 1987). Recently, the interest in quantifying POP levels in high mountain regions has increased, because contaminant transport and distribution in mountain areas could help in understanding the mechanisms operating on a larger scale and the influence of various environmental parameters (climate, altitude, etc.). In addition, mountainous regions have been traditionally considered pristine environments and thus are suitable for studying the effects of remote pollution sources. In this

context, soils and lake sediments have an important role as sink and reservoir for POPs. High elevation lakes, in particular, may capture primarily volatile POPs (e.g., tri-penta-chlorinated PCBs), whereas superficial soil may also be influenced by local pollutant sources (e.g., PAHs) (Guzzella et al., 2011; Belis et al., 2009). Moreover, lake sediments bear witness to climate change. In fact, due to global warming and the consequent shrinking of glaciers, glacial lakes located at the highest elevations have increased their surface area (Tartari et al., 2008; Salerno et al., 2012).

In this study, two different sampling campaigns (2008 and 2012) were undertaken on the southern slopes of Mt. Everest (Himalaya, Nepal) with the aim of investigating the presence and the sources of contamination with POPs and PAHs in this remote region. Both sediments and soils were sampled in 2008 at different altitudes to monitor DDT, its isomers and metabolites (hereafter indicated as DDx), PCBs, PBDEs and PAHs contamination, while in 2012, only soil samples were collected for the analysis of PAHs. Moreover, in 2008, a wide area was covered by the sampling plan, including the valleys of Imja, Thame and Gokyo. To the best of our knowledge, this is the first time these Southern Himalayan valleys were investigated for the presence of anthropogenic contaminants. In particular, the LRAT mechanism, global warming, and the recent increase of tourism in this area were all considered and evaluated as possible driving forces of the present contamination.

2. MATERIALS AND METHODS

2.1 Study area and the sampling stations

The current study focuses on the Mt. Everest region and, in particular, on the Sagarmatha National Park (SNP) and 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 central Himalayas (Fig. 1) (Amatya et al., 2010; Thakuri et al., 2014). The SNPBZ is the world's highest protected area (1148 km²), visited by over 30,000 tourists in 2008 (Salerno et al., 2013), and is extended from an elevation of 2845 to 8848 m a.s.l. (Mt. Everest). The region is located in the subtropical zone and is influenced by the monsoon system. Moreover, local circulation is dominated by a system of mountain and valley breezes (Salerno et al. 2015). Two different sampling campaigns were undertaken in

October 2008 and October 2012 in the SNPBZ (Fig. 1). In the first one, both sediments and soils were sampled. In particular, the sediment samples were collected from 8 lakes, located at different altitudes, in the areas of Khumbu and Imja valleys (Table 1). The 16 soil samples were taken from the same sites (at a distance of 10-100 m from the lakes), and additionally in three stations during the trekking ascent, avoiding areas with intense human activities, and in the Thame and Gokyo valleys (Table 2). In the second campaign, eight soils samples were collected exclusively in the Khumbu valley and in the same trekking ascent stations (Table 2).

All sediment samples were collected using a hand-held stainless steel scoop from the shore (Field Sampling Guidance Document n. 1215 U.S. EPA, California, 1999), collecting samples from the surface to about 3 cm of depth. This may represent a quite recent depositional period of about the last 10-40 years, according to the deposition rates suggested by Galassi et al. (1997), where a deposition rate of 15 years/cm and 3 years/cm was measured in lakes LCN 9 and LCN 10, respectively. Soil samples (sub-surface samples, organo-mineral A horizon) were collected at 5-10 cm depth, after root section removal. At each site, the soil samples were collected using a stainless steel spade and then sealed in clean plastic bags for transport. In October 2012, a specific soil survey supported the sampling campaign, and the main physical and chemical properties of the sampled soils are shown in Supplementary Material (SM), Table S1, and commented in section SM1.

2.2 Target analytes

The target analytes were: 17 PAHs, divided in Low Molecular Weight (LMW-PAHs) - 1-methylnaphthalene (1mNa), 2-methylnaphthalene (2mNa), acenaphthylene (Ayl), acenaphthene (Aen), fluorene (F), phenanthrene (Pn), anthracene (An), fluoranthene (Fl), pyrene (Py), and High Molecular Weight (HMW-PAHs) - benz[a]anthracene (BaA), chrysene (Ch), benzo[b+k]fluoranthene (BbkF), benzo[a]pyrene (BaP), perylene (Per), indeno[1,2,3-cd]pyrene (InP), dibenz[a,h]anthracene (DhA), and benzo[ghi]perylene (Bghi)), 14 PCBs (PCB-18, -28, -31, -44, -52, -101, -118, -149, -138, -153, -170, -180, -194, -209), 6 DDX (o,p'-

DDE, p,p'-DDE, o,p'-DDD, p,p'-DDD, o,p'-DDT, p,p'-DDT), and 8 PBDEs (BDE-28, -47, -99, -100, -153, -154, -183, -209). The chemical purchasing is reported in SM2.

2.3 Analysis of the soil samples

The chemical and physical analyses of the sampled soil were performed according to standard methods (Ministero delle Politiche Agricole e Forestali, 2000). All samples were air-dried and sieved to separate the fine earth (< 2 mm) from the coarse fraction. pH was determined potentiometrically in water extracts (1:2.5 w/w). The total C and N concentration was measured by dry combustion with an elemental analyzer (CE Instruments NA2100, Rodano, Italy). The clay and fine silt fraction were determined by the pipette method. Organic carbon content (OC) of the soils and sediments was determined on 0.5-1 g d.w. by back-titration after oxidation with potassium dichromate in the presence of sulfuric acid, following the method published by Walkley and Black (1934). The authors assessed that with this method, it is possible to oxidize the more active fraction of organic carbon content, mainly representing 77% of the total amount.

2.4 Sample preparation and instrumental analysis

The sample preparation and analysis of target compounds was performed as reported by Guzzella et al. (2011). Briefly, after lyophilisation, the sediments were sieved through 63 µm mesh, whereas soils were air-dried and sieved through 2 mm mesh to remove gravel, plant debris and other inert materials. For PAH analysis, a variable amount of sample (0.5-1 g d.w. of sediment and 30-50 g d.w. of soil) was spiked with 50 µL of the PAH internal standard (IS) and then extracted in a hot-Soxhlet apparatus (Buchi, Flawil, Switzerland) using a *n*-hexane/acetone mixture (3:1 v/v) for 25 cycles. The extract was concentrated to 1 mL under gentle nitrogen stream and cleaned-up on a glass column packed with 4 g of silica gel and alumina (2:1 w/w). The aliphatic fraction was discarded by elution with 3 mL of *n*-hexane, while the aromatic fraction was collected using 10 mL of *n*-hexane/dichloromethane (70:30 v/v). The final extract was then concentrated by nitrogen stream to 0.5 mL. For DDx, PCB and PBDE analysis, the same amount of sample was spiked with 50 µL of the DDx/PCB/PBDE IS and then extracted in the same way. The clean-up was performed using a

multilayer column (1.5 x 6.5 cm) packed (bottom to top) with 0.5 g of acidified silica gel (H₂SO₄: 30% w/w), 0.5 g of potassium silicate, 2 g of acidified silica gel and 1 g of Florisil[®]. The sample was eluted with 10 mL of *n*-hexane/DCM (1:1 v/v), and the final extract was concentrated by nitrogen stream to 100 µL. The instrumental analysis of all the target compounds was also undertaken as published by Guzzella et al. (2011). All GC details and conditions for the analysis of target compounds are fully reported in the SM3.

2.5 Quality Assurance (QA) and Quality Control (QC)

For PAH sediment and soil analysis, the method performance was evaluated using certified sediment purchased by IAEA (International Atomic Energy Agency): Reference Material IAEA-383 “Organochlorine Compounds Petroleum Hydrocarbons and Sterols in Sediment Sample”. All measured values ($n = 3$) were within the range of expected concentrations ($\pm 30\%$), whereas the relative standard deviation (RSD) ranged from 15% to 31% (the results for all the compounds are given in SM4, Table S2). Because of the possibility of laboratory contamination, especially due to dust deposition, a procedural blank was performed every batch of samples, and some 2-4 carbon ring PAHs (e.g., 1-, 2-methylnaphthalene, pyrene, phenanthrene, fluorene and acenaphthene) were found in the blank extracts, albeit at low concentrations. For these compounds, the blank concentrations were subtracted from the values found in the samples. For PBDE sediment and soil analysis, the method performance was evaluated using the sediment BROCC-2 CRM purchased from RIVO (Netherlands Institute of Fisheries Research). RSD of measured values ($n = 3$) ranged from 4% (BDE-49) to 25% (BDE-66) (whole results are given in Table S3). For DDX and PCBs, the method performance was evaluated using 1939a SRM river sediment, purchased from NIST (National Institute of Standard and Technology, Gaithersburg, Maryland). All measured values ($n = 3$) were within the range of expected concentration levels ($\pm 30\%$) except for PCB-170 (+45%) and PCB-194 (+57%), and the RSD ranged from 9% to 25% (whole results are given in Table S4). Blank concentration levels were below LOD levels for all analyzed POPs. Using a signal-to-noise ratio of 3:1, the limits of detection (LODs) were estimated in both the sediments and the soils as 0.1 and 0.01 ng/g d.w, for PAHs, and DDX/PCBs/PBDEs, respectively.

2.6 Data analysis

All maps were elaborated on ArcGis platform. The correlations among the results were tested with Spearman's Correlation Coefficients, using SPSS Statistics software, and Principal Component Analysis (PCA) was performed using STATISTICA Six Sigma ver. 8. Because of the different fraction of soils and sediments considered (<2 mm and <50 μm , respectively), the concentrations normalized on organic carbon content were used to represent the contamination due to PAHs and POPs in the samples, to ensure that their estimates were comparable. In addition, when calculating the averages, the values below the limit of detection were set to zero, to avoid overestimating the sample concentrations.

3. RESULTS AND DISCUSSION

3.1 Contaminant concentration and the distribution pattern of the sediments

The concentrations of DDx, PCBs, PBDEs, and PAHs normalized on organic carbon content measured in the sediments (< 50 μm) collected in 2008 are reported in Table S5. Regarding DDx contamination, only the metabolites p,p'-DDD and p,p'-DDE were measured at concentrations greater than the detection limit, ranging from 1.3 ng/g OC (in LCN 53_sed) to 45 ng/g OC (in LCN 9_sed). These concentrations are extremely low and can be considered as background concentrations, as previously suggested by Guzzella et al. (Guzzella et al., 2011). They are also comparable to concentrations measured in sediment samples collected from the Tibetan Plateau (Cheng et al., 2014; Wu et al., 2014). In addition, the mean percentage contribution of p,p'-DDE (56%) and p,p'-DDD (44%), with respect to the total DDx contamination, indicates that the parental compound p,p'-DDT was generally absent. This may be because p,p'-DDT had been degraded by biotic and abiotic reactions which occurred both in sediments and in the weathered and aged soils. Total PCB contamination ranged from 3.9 to 84.3 ng/g OC in LCN 6_sed and LCN 9_sed, respectively, with a mean value of 20.2 ± 26.3 ng/g OC. This concentration level is consistent with other studies conducted on sediments collected worldwide in other pristine areas and in remote and high elevation regions (Zhang et al., 2014; Usenko et al., 2007; Guzzella et al., 2011; Jiao et al., 2009; Pozo et al., 2007). It is known that while the lighter congeners (tri- to penta-CBs) tend to volatilize and be transported in the gas

phase from polluted regions to remote areas through the atmosphere, the heavier congeners (hexa- to deca-CBs) tend to be transported on atmospheric particulate and to be deposited near the pollution sources (Li et al., 2012). Confirming this theory, the average PCB profile in the SNP sediments was dominated by the lighter chlorinated congeners (73%), while the heavier ones accounted for the remaining 27%. Similar PCB profiles were determined previously by Guzzella et al. (2011) and in other remote areas (Li et al., 2012; Ruiz-Fernandez et al., 2014; Choi et al., 2008).

In addition to the LRAT model, the presence of PCBs in the sediments of the SNP lakes could also be related to the glacier hypothesis. Based on this hypothesis, glacial ice may contain significant amounts of chemicals deposited in earlier times that have been stored in the deeper layers of the ice during the last decades (Bogdal et al., 2009; Schmid et al., 2011; Ali et al., 2014; Kusky, 2009). Accelerated glacier melting may result in a release of chemicals from this kind of reservoir. To check if the considered lakes might have been affected by the delayed release of environmental contaminants, whose emissions were high in the past, the PCB range concentrations in the various sediments was graphically represented (Fig. 2). The highest contamination was observed in LCN 9 (84.3 ng/g d.w. OC); this could be because LCN 9 is a *first order* lake, with a glacier of 0.17 km² located within its basin (Table 1). By contrast, LCN 10 (located downstream lake LCN 9) showed a PCB concentration of about 10 times lower (9.1 ng/g OC). This huge difference (between lakes located quite close to each other and belonging to the same hydrological pathway) could be because lake LCN 9 is directly fed by melt water coming from the glacier located in its basin, whereas LCN 10 is a *second order* lake, fed primarily by the relevant upstream lake and by the small portion of its sub-basin which does not contain glaciers. Based on this observation, we can speculate that PCBs are also released by the shrinkage of the glacier, and then stored in the relevant first order lake. The other considered lakes presented concentrations of PCBs depending on the glacier extension in their basin. The least contaminated sediments were LCN 6_sed (3.9 ng/g OC), with 0.07 km² of glacier coverage, and LCN 53 (8.4 ng/g OC) with no glaciers located in its basin. Generally, a relationship between basins with more glaciers and the most polluted lake sediments was observed, with the only exception being LCN_31. This lake, in fact, presented a low PCBs concentration (7.2 ng/g OC) and could be defined as pro-glacial (a lake whose surface is in direct contact with a large glacier front - 2.16 km²) (Salerno et al., 2012). This different behavior can be explained by considering that this particular glacier (Duwo glacier) was observed, in the last fifty years, to have an opposite response to

climate change with respect to all of the other glaciers located in the region. Instead of retreating, its surface area remains constant (Tab.2) (surge phenomenon) (Thakuri et al., 2014). Therefore, LCN 31 does not have clear chemical features depending on the glacier release.

A different effect was observed when considering the PBDE contamination in the sediment samples, where BDE-99 and BDE-209 were the only detected congeners. The total PBDE concentration ranged from <LOD to 103 ng/g OC (in LCN 31_sed), with average values of 41 ± 31.5 and 49 ± 51.7 ng/g OC in Khumbu and Imja valleys, respectively, in accordance with our previous study (Guzzella et al., 2011) and other remote lacustrine environments in Central Mexico (Ruiz-Fernandez et al., 2014) and Scotland (Russel et al., 2011). In the present investigation, the predominant congener was the fully brominated BDE-209, which contributed 95% of the total PBDE contamination. The presence of BDE-209 is in agreement with the composition of the Deca-BDE technical formulation (~97% of BDE-209), suggesting the local use of this commercial mixture. Deca-BDE has been banned in Europe in the electrical and electronic equipment production (European Court of Justice, 2008), and the major flame retardant producing companies have agreed with the market regulations on the production and use of Deca-BDE formulation (BSEF, 2012). For this reason, it will be interesting to investigate the future presence of PBDEs in such remote environments. The presence of BDE-99 (5%) in the sediments, a persistent PBDE congener, however, might confirm the hypothesis that remote regions act as “cold condensers”, being susceptible to the accumulation of lesser brominated congeners, transported through the atmosphere and then re-condensed on the surface (Jiao et al., 2009). The concentrations of individual DDx, PCB and PBDE compounds detected in the sediments did not correlate significantly with the elevation of sampling points. This could be due to the limited concentrations measured in the samples and the considered reduced altitudinal gradient (about 800 m) at which the lakes are located.

PAH contamination in the sediments collected from the SNP (Fig. 2) was on average ten times higher than those measured for POPs, ranging from 1020 ng/g OC (LCN 53_sed) to 7274 ng/g OC (LCN 6_sed), and the mean PAH concentrations reached values of 4323 ± 2796 ng/g OC in the Khumbu Valley and 2464 ± 1343 ng/g OC in the Imja Valley. Pn was the most abundant compound, accounting for 42% of the total PAHs considered, followed by Fl (15%), 1mNa (13%), and Py (10%), in accordance with our previous study

(Guzzella et al., 2011). In addition, the LMW-PAHs prevailed in the total sum of PAH (93%), with concentrations of up to 7000 ng/g OC, while the HMW-PAHs accounted only for the remaining 7%, with a concentration generally below 600 ng/g OC. In order to provide information on the input sources and transport pathways, PCA was applied (Fig. 3). The first two PCs represented 80% of the total variances of PAH concentrations in the sediments. Combined with the variable and case plots, the LMW-PAHs (2, 3, and 4 rings) were congregated into one group represented by the sediments collected in lake LCN 10 and LCN 6, whereas the HMW-PAH (5 and 6 rings) were the main compounds accumulated in sediments from LCN 24 lake. Elevation, considered a supplementary variable, was negatively correlated with 5 and 6 ring PAHs, mostly because these PAHs are transported by absorption on atmospheric particulates and their deposition is near the pollution sources. On the contrary, with increasing altitude, lakes are usually far from the pollution sources, commonly represented by densely populated villages. In this study, no relation with altitude was evident for the considered LMW-PAH compounds in sediments.

3.2 Contaminant concentration and distribution pattern in soil

The concentrations of DDX, PCBs, PBDEs, and PAHs normalized on organic carbon content measured in the soils (< 2 mm) collected in 2008, and those of PAHs detected in the soils sampled in 2012, are reported in Table S5. DDX concentrations in soil samples ranged from <LOD to 12.2 ng/g OC, detected at the GOK sampling station, with an average concentration of 2.6 ± 3.2 ng/g OC. Similar DDX concentrations were found in the SNP in 2007 (Guzzella et al., 2011), in soils collected from Southern and Western China, and in non-agricultural soils in other remote regions (Wang et al., 2007; Chen et al., 2008; Gai et al., 2014). The DDX contamination pattern in the soils is similar to the one observed in the sediments, with the predominance of p,p'-DDE (47%) and p,p'-DDD (41%). The presence of o,p'-DDE was also evidenced in three stations (LUK_soil, NAM_soil, and GOK_soil), suggesting the occurrence of biodegradation of o,p'-DDT in soils, which might be related to the usage of Dicofol in the surrounding agricultural territories. In fact, the use of this acaricide, widely used in India and China and containing high concentrations of DDT as technical impurities (o,p'-DDT accounted for about 77%), could have contributed to the current DDT profiles in soils (Ren et al., 2014; Wang et al., 2006; Rasenberg and Van de Plassche, 2003; Yang et al., 2013).

PCB and PBDE concentrations were very low in all considered soil samples, ranging from 0.3 to 7.0 ng/g OC (average value of 1.4 ± 1.6 ng/g OC), and from <LOD to 3.7 ng/g OC (average value of 0.9 ± 1.4 ng/g OC), respectively. Regarding PCBs, GOK_soil was the most polluted station, as for DDX, while the PBDE presence was very sporadic in all the considered area. The distribution of PCBs in soils was similar to the one observed in the sediment samples, but it must be pointed out that the PCB profile in soils is more stressed by the predominance of tri- penta- congeners (96%) with respect to the higher chlorinated congeners (4%). Such distribution of low-chlorinated PCBs has been observed also in surface soils collected from the Tibetan Plateau (Zheng et al., 2012; Wang et al., 2009). As expected, considering the very low concentrations measured for DDX, PBDEs and PCBs, the SNP soils can be classified as background levels for mid-latitude Northern Hemisphere, mainly determined by long-range atmospheric transport (LRAT) and therefore not directly influenced by local pollution sources (Wang et al., 2007; Wang et al., 2009; Sun et al., 2015; Yuan et al., 2015). The southern slope of the Himalayas can consequently be considered as a pristine area on a global scale (Zheng et al., 2012). Moreover, despite the wide altitudinal gradient considered for the soils (about 2700 m), the concentrations of individual DDX, PCBs and PBDEs detected in the soils did not correlate significantly with the elevation, most probably because of the extremely low levels of contamination.

Regarding PAHs, the soils sampled in 2008 and 2012 showed concentrations from 50 to 100 times higher than those detected for the other compounds, ranging from 42 to 677 ng/g OC in DOL_soil and LCN 9_soil, respectively. Jones et al. (1989) and Maliszewska-Kordybach (1996) proposed two classifications for evaluating the PAH contamination of an area based on the concentration measured in its soil. Based on both these classifications, the SNP soils could reasonably be considered low contaminated, far from direct industrial pollution sources (Wang et al., 2007). Such concentrations and similar conclusions were drawn from soils collected around the Mt Everest and from the Tibetan Plateau (Yang et al., 2013; Wang et al., 2007; He et al., 2015). In 2008, the highest PAH concentrations were observed in Namche, LCN 6, LCN 54 and Gokyo samples, while in 2012 in Lukla, Namche, LCN 9 and LCN 10, and, by considering the same stations in both years (Pharak zone and Khumbu valley, Tab. 1), the PAH contamination in 2012 was 1.5 times higher than in 2008. However, the difference between the two time periods was not statistically significant ($p > 0.05$). The documented increase of tourism activities in the last years, especially from 2008 to

2012 (Salerno et al, 2013; Salerno et al., 2010a), might be put in relation with this increase, even though this hypothesis cannot be confirmed. According to the local register compiled by the park authorities, in fact, the number of tourists in the SNP, and subsequently the number of human activities that could have released PAHs (house heating, energy plant, etc.), increased progressively from 30,599 units in 2008 to 36,212 units in 2012 (increase of 17%).

Similar to the sediments, the contribution of LMW-PAHs to the total sum of PAHs prevailed on HMW-PAH (73% and 27%, respectively) in both years, and Pn was again the most abundant compound detected in the soils (Table S5), with an average percentage of 24% on the total amount of PAHs, followed by 1mNa (15%). Such a pattern, observed also in other studies conducted on soils collected from remote regions (Yang et al., 2013; Wang et al., 2007; Guzzella et al., 2011), showed a departure in behavior considering the soils collected in the Pharak zone and in the Khumbu valley in the two-year period. While in 2008 the distribution pattern showed LMW-PAHs > HMW-PAHs (66% and 34%), and Pn > 1mNa (24% and 13%), in 2012 the contribution of HMW-PAH to the total sum of PAHs increased up to 48%, due to the presence of perylene, the most abundant PAH (26%). However, such relevant percentage of Per, in relation to the other PAHs, can be indicative of a diagenetic rather than anthropogenic origin. Not considering this compound in the calculation of the PAH profile, the distribution pattern of PAHs in soils in 2012 resulted the same as in 2008, with LMW-PAHs > HMW-PAHs (66% and 34%), and Pn > Py (21% and 15%). At any rate, the PCA was not to point out significant differences in PAH soil contamination, neither due to the specific molecular weight, nor to the sampling stations. Even in this case, the concentration of PAHs in the soils did not correlate significantly with elevation.

3.3 Evaluation of PAH sources in sediments and soils from the SNP

PAHs may originate directly from both natural (e.g., biodegradation of humic substances and fires) and anthropogenic (e.g., vehicle emissions, incomplete combustion of fossil fuels, industrial processes, waste incineration) sources (Yunker et al., 2002). According to the formation mechanism, they can derive from pyrogenic (combustion processes) and/or petrogenic (release of uncombusted petroleum products) sources.

HMW-PAHs are believed to originate from anthropogenic combustion/pyrogenic sources, whereas petroleum-derived residues contain a relatively high abundance of LMW-PAHs (Wang et al., 2006; Mai et al., 2003). In addition, the ratio LMW/HMW-PAHs is generally used to distinguish PAH input sources in specific regions (Wang et al., 2006). Ratios <1 suggest the pyrogenic sources of PAHs (likely from vehicular exhausts) because, after combustion, fossil fuels contain a higher concentration of HMW-PAHs than LMW-PAHs (Zhang et al., 2004; Nozar et al., 2014). On the contrary, ratios >1 indicate PAHs originating from low temperature combustion and petroleum-derived residues (Mai et al., 2003). To investigate the sources of contamination of PAHs in the SNP, the ratios LMW/HMW-PAHs were calculated for the sediments and soils considered (Table S6). In the sediments, the ratio ranged from 4.7 to 36.0, comparable with values determined by Wang et al. (2006) in pine needles (6.5-24.3) collected along the Northern slope of the central Himalayas in China. Such results could reasonably indicate that petroleum and low-temperature combustion were the major contributions of PAH sources, because kerosene is the most common energy source (33%) for power and heating generation in the SNP, followed by fuel wood (30%), dung (19%), and liquefied petroleum gas (7.5%) (Salerno et al. (2010b)). In the SNP, energy is mainly used for human activities (i.e. cooking, boiling water, heating for homes, and lighting). Due to the scarcity of forestry wood and the increase of tourists, the use of commercial fuels (especially kerosene) has recently increased. In guesthouse buildings, like tourist lodges and hotels, kerosene is the main fuel used for heating and cooking. Therefore, its incomplete combustion in the SNP region might explain the emission of 2-, 3-, and 4-ring PAHs, leading to the significant contribution of these compounds in the sediment analyzed. At any rate, the LMW/HMW-PAHs ratio >1 could be also affected by the LRAT process, which supported the transport of LMW-PAHs in the gas phase, accumulating at high elevation region (Wang et al., 2007).

In soils, the ratios LMW/HMW-PAHs ranged from 0.3 to 31.8 in 2008 (as in the sediments) and from 0.5 to 5.4 in 2012 (not considering Per in the calculation, due to its diagenetic origin). This could suggest that in recent years the sources of PAHs in soils has become both petrogenic and pyrogenic. In fact, the ratio was >1 only in 50% of the soils collected in 2012, suggesting the important role of high temperature combustion sources. This ratio is typical of pyrogenic contamination, likely caused by vehicular exhausts and emissions

from solid high temperature combustion (coal). Namche (ratios of 0.3 in 2008, and 0.5 in 2012), in fact, is the main commercial base and tourist hub in the Khumbu region (Fig. 4). A stop at Namche village is necessary for trekkers who want to climb the 8000 m mountain peaks of the Khumbu valley, allowing a physiological body adaptation to the high altitude climate conditions. Another PAH local source of contamination could be the Syangboche Airport (the closest airstrip to Mount Everest and to Everest base camp) that is mostly used by tourists who plan to have a stop at Namche Bazaar. The airport is located on a hill near the village (at 3750 m) and therefore it can contribute to the HMW-PAH contamination of the soils.

4. CONCLUSIONS

In this study, a wide sampling area was considered, and, however the conditions faced during the collection of the samples were prohibitive (mainly caused by such high elevations), more than thirty samples were collected for the analysis. Based on the results obtained, most of them close or below the detection limits, it was possible to confirm that this area is characterized by a very limited contamination due to POPs, and, as expected, the southern slope of the Himalayas can generally be considered a pristine area on a global scale. The detectable presence of POPs in this remote region was therefore ascribed mainly to the long-range atmospheric transport mechanism, while the gradual melting of glaciers was suggested to be a secondary source of pollution of the lake sediments, strictly related to the LRAT tendency of the compounds (especially PCBs). Also considering the PAH contamination, the SNP could be classified as a low contaminated site, far from direct industrial pollution sources. However, considering the documented increase of human activities characterizing the SNP in the last decades and the prevalence of LMW-PAH with respect to HMW-PAH (likely caused by energy source for power and heating generation), the local impact of tourism can be considered as a possible source of contamination, which might become even more relevant in the near future.

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Table 1. Geographical and hydro-morphological features of the sediment sampling sites. The Coordinates are expressed in Modified UTM projection – Datum Everest 1830 (Salerno et al., 2012).

Name	Code	Longitude	Latitude	Elevation	OC	Valley	Lake surface	Basin surface	Sub-basin surface	Glacier surface in sub-basin	Glacier surface area variation	Lake order
		m	m	m (a.s.l.)	%		km ²	km ²	km ²	km ²	%	
Lake 24	LCN 24_sed	479769	3088514	4605	1.0	Khumbu	0.54	22.58	16.22	1.14	-21	2
Lake Inferior	LCN 10_sed	481656	3093053	5145	1.1	Khumbu	0.02	0.66	0.32	0.00	-	2
Lake Superior	LCN 9_sed	481154	3093343	5255	0.2	Khumbu	0.009	0.98	0.98	0.17	-27	1
Lake Kalapattar 6	LCN 6_sed	482684	3096815	5440	0.5	Khumbu	0.008	0.25	0.25	0.07	-36	1
Lake Kalapattar 5	LCN 5_sed	482394	3096744	5410	0.7	Khumbu	0.013	0.35	0.35	0.19	-30	1
Lake 31	LCN 31_sed	484600	3093892	4780	1.0	Imja	0.06	5.40	5.40	2.16	-3	1
Lake 53	LCN 53_sed	490511	3085873	5100	1.6	Imja	0.01	27.40	0.40	0.00	-	2
Lake 161	LCN 161_sed	490872	3086107	5090	1.4	Imja	0.71	27.00	27.00	25.65	-9	1

Table 2. Geographical coordinates, elevation, and OC content of the soil sampling sites. The Coordinates are expressed in Modified UTM projection – Datum Everest 1830 (Salerno et al., 2012).

Sample sites			Field campaigns							
Name	Code	Valley	2008				2012			
			Longitude m	Latitude m	Elevation m (a.s.l.)	OC %	Longitude m	Latitude m	Elevation m (a.s.l.)	OC %
Lukla	LUK_soil	Pharak	472705	3063487	2730	4.3	472521	3063625	2660	6.3
Namche	NAM_soil	Pharak	471912	3073523	3625	4.2	471868	3075335	3570	6.7
Periche	PER_soil	Khumbu	482152	3085827	4265	3.7	482157	3085606	4260	6.5
Lake 24	LCN 24_soil	Khumbu	479769	3088514	4605	3.0				
Pyramid	PYR_soil	Khumbu					481636	3092685	5050	4.5
Lake Inferior	LCN 10_soil	Khumbu	481656	3093053	5145	4.8	481648	3093068	5100	4.1
Lake Superior	LCN 9_soil	Khumbu	481154	3093343	5255	3.5	481217	3093360	5265	2.8
Lake Kalapattar 7	LCN 7a_soil	Khumbu					482290	3096428	5245	2.6
Lake Kalapattar 7	LCN 7b_soil	Khumbu					482383	3096269	5250	1.3
Lake Kalapattar 5	LCN 5_soil	Khumbu	482394	3096744	5410	1.1				
Lake Kalapattar 6	LCN 6_soil	Khumbu	482684	3096815	5440	0.4				
Lake 32	LCN 32_soil	Imja	484398	3094114	4750	5.7				
Lake 54	LCN 54_soil	Imja	491482	3085716	5140	1.6				
Dole	DOL_soil	Gokyo	473385	3082730	4150	7.6				
Gokyo	GOK_soil	Gokyo	469424	3094007	4850	0.9				
Lanmouche	LAN_soil	Thame	464852	3081102	3960	3.4				
Dig Cho	DIG_soil	Thame	460315	3082818	4345	1.7				
Chhule	CHH_soil	Thame	462907	3088314	4385	5.1				
Melun	MEL_soil	Thame	463618	3094218	5005	3.2				

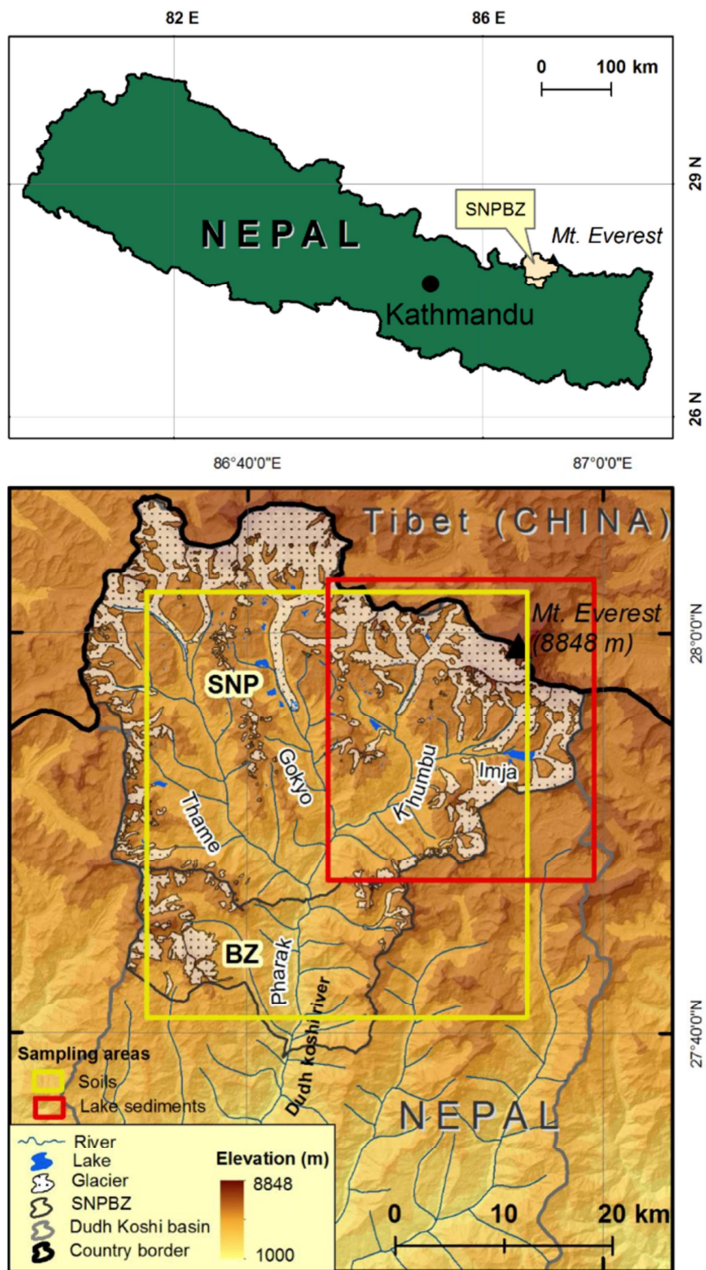


Fig. 1. Study area and sampling areas for soils and lake sediments.

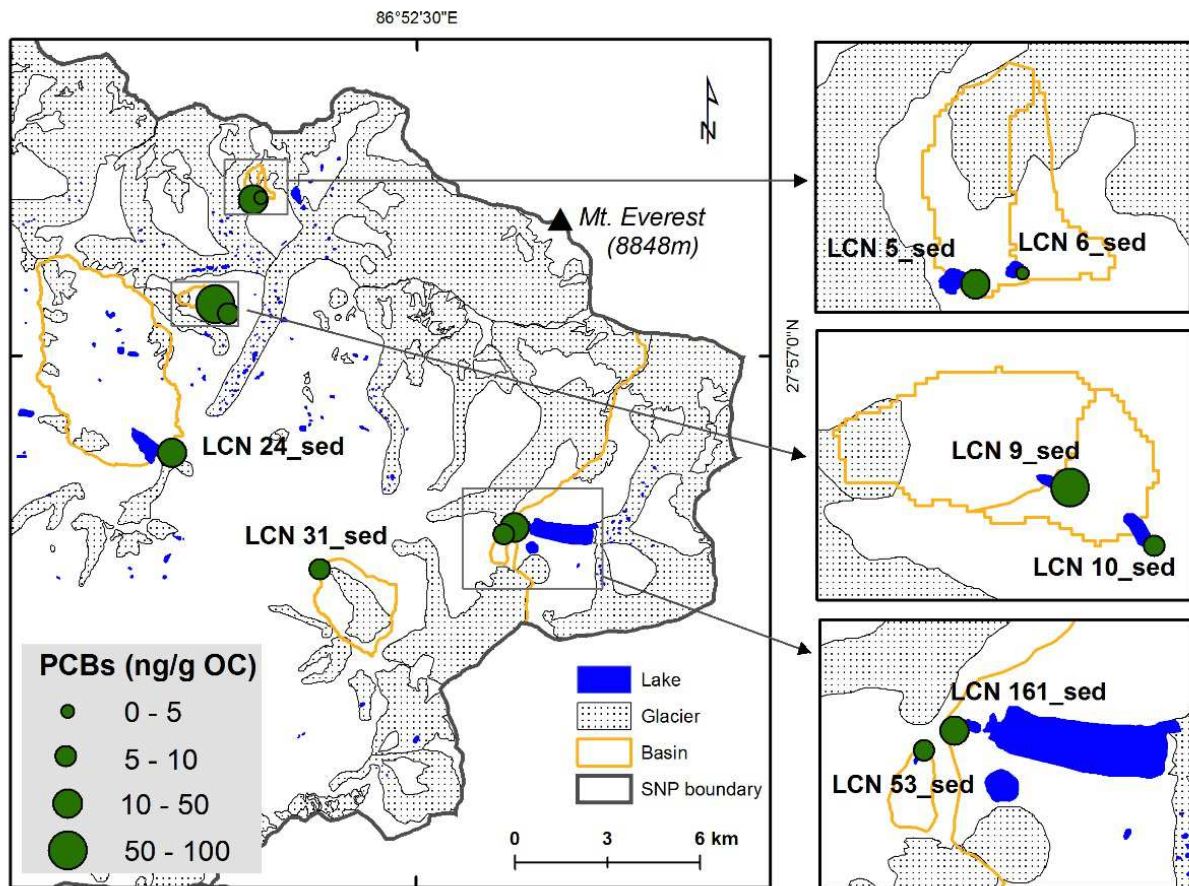


Fig. 2. PCB concentration range in the sediment samples from the SNP lakes.

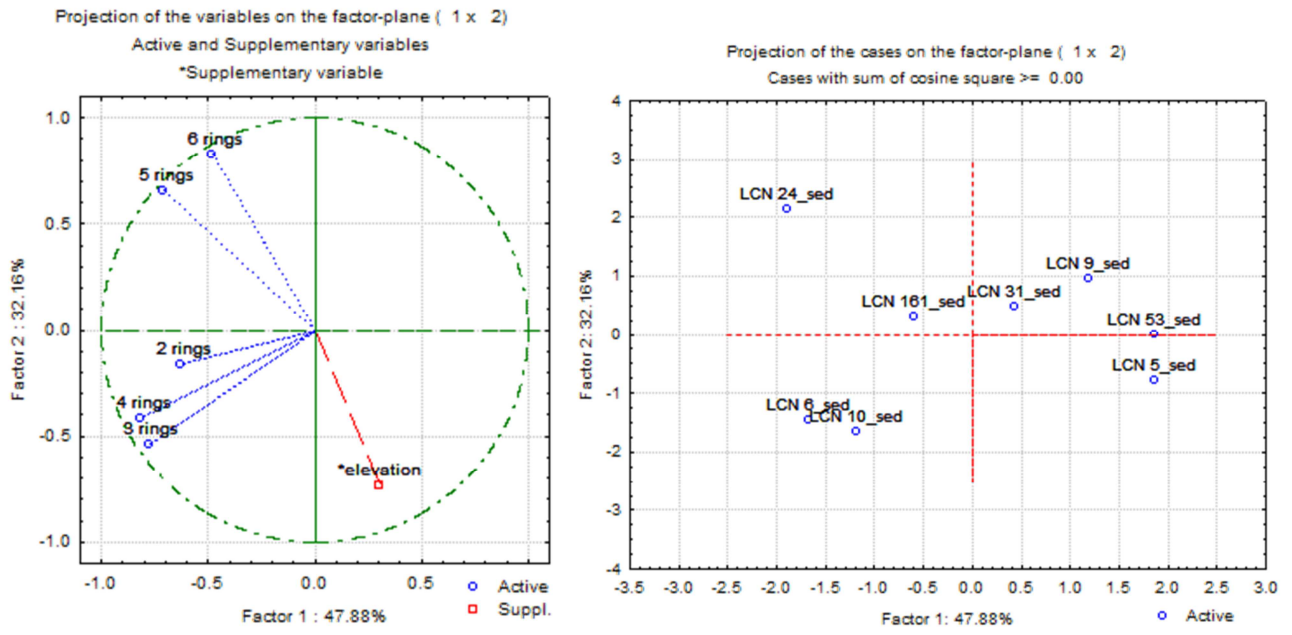


Fig. 3. PCA diagram showing the relationships between the variables (left) and the cases (right).

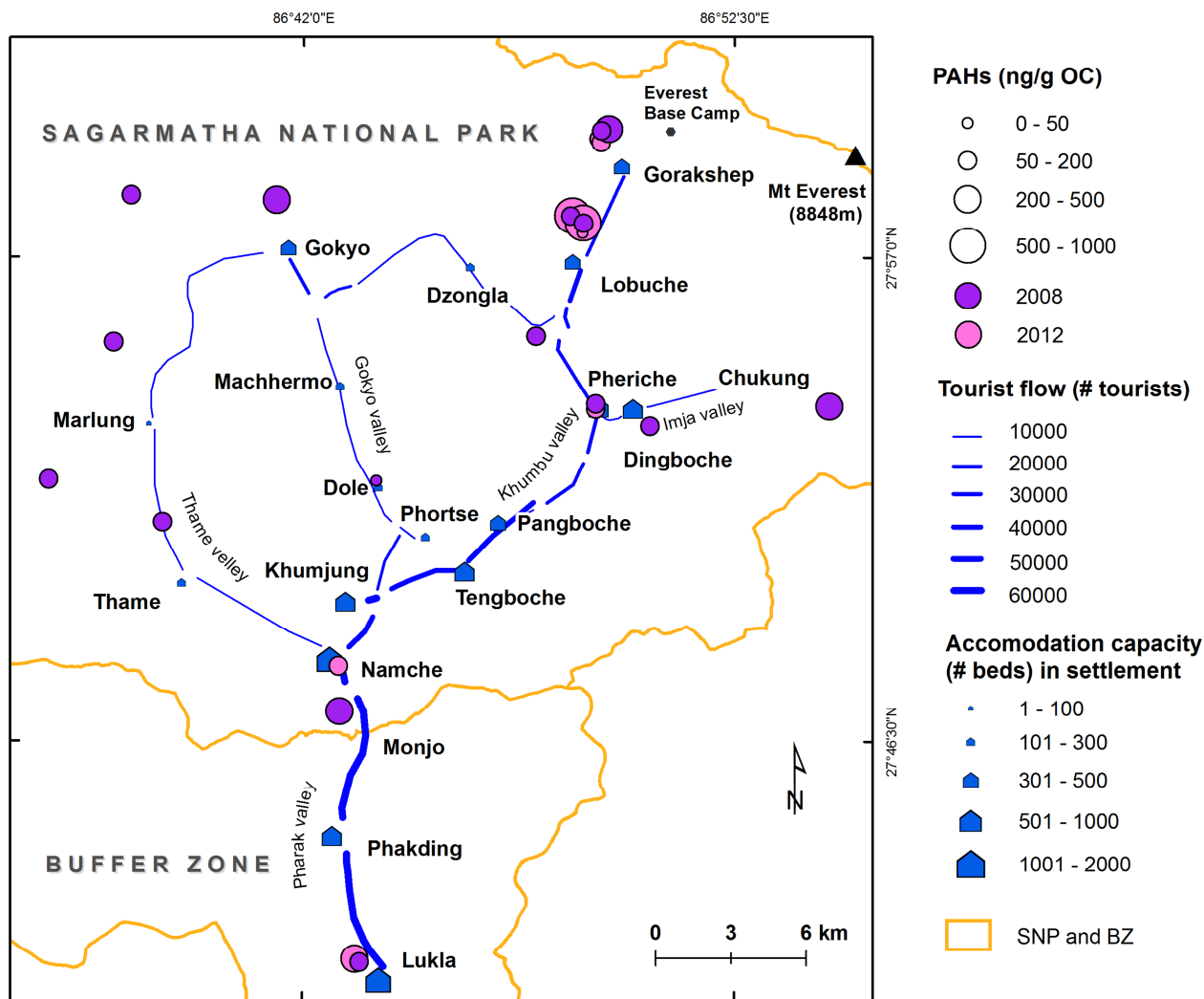


Fig. 4. Total PAHs (ng/g OC) in soil samples (2008 and 2012), touristic flow and accommodation capacity of settlements

SUPPLEMENTARY MATERIAL

POP and PAH Contamination in the Southern Slopes of Mt. Everest (Himalaya, Nepal): Long-range Atmospheric Transport, Glacier Shrinkage, or Local Impact of Tourism?

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SM1. Soil characteristics

The SNP is located in a complex transition zone between the High Himalayas and Tibet, characterized by diverse geological units. Bortolami et al. (1998) discussed the distribution of the geo-lithological characteristics of the territory. The soils in the high valleys is primarily Entisol; below 4000 m asl, Spodosols have developed in forested areas, which are mainly located in the north-facing slopes; the extensive grassland and shrub land areas, mainly in the southern slopes below 3750 m asl, have Inceptisol and Entisol (Nepal and Nepal, 2004).

The organic-mineral horizons considered in this study (A horizons for all soil, Ap horizon for Lukla) were strongly acidic (Table S1), with the exception of the samples collected in Lukla and Namche, which were moderately acidic. The soil OC (SOC) (Table S1) ranged from 1.3 % in LCN 7b_soil to 6.7 % in NAM_soil, and, using the Spearman correlation, was significantly correlated ($r=0.945$; $p<0.01$) with the TN. The SOC and TN content (in the samples collected in this study above ~4000 m asl) was comparable to values recorded at around 2800 m asl in the Alps (Freppaz et al., 2010) and of those above 5000 m asl in recently de-glaciated high-altitude soils of the Himalayas (Stres et al., 2013). The SOC and TN (Table S1) contents were inversely correlated with the elevation ($r=-0.749$; $p<0.05$ and $r=-0.833$; $p<0.05$, respectively). The C/N ratio, with the exception of Lukla, was relatively low and comparable to alpine sites with scarce vegetation like in the Rocky Mountains (Williams et al., 2011). The clay fraction, very low and comparable with values recorded in recently de-glaciated high-altitude soils of the Himalayas (Stres et al., 2013), were significantly inversely correlated with the C/N ratio ($r=-0.786$; $p<0.05$). Therefore, soils with the highest clay content seemed characterized by the highest mineralization. The SOC content was generally higher than the critical point of 1-2 %, value below which the principal dynamic for the absorption of contaminants in soils can also be mainly dominated by mineral content (Yuan et al., 2012). Effectively, in this study, the clay content and fine silt fraction did not show any significant correlation with respect to the considered contaminants.

Table S1 - Main physical and chemical properties of the soils sampled in 2012

<i>Soils 2012</i>	pH	TN %	C/N	Clay %	Fine silt %
LUK_soil	5.6	0.32	19.5	0.1	8.8
NAM_soil	5.8	0.50	13.3	0.6	10.7
PER_soil	5.5	0.57	11.5	1.9	2.3
PYR_soil	5.4	0.28	16.1	0.7	4.3
LCN 10_soil	5.2	0.29	14.0	1.3	10.0
LCN 9_soil	5.4	0.19	15.1	0.3	5.6
LCN 7a_soil	5.5	0.20	13.0	1.5	8.9
LCN 7b_soil	5.5	0.10	12.7	0.9	5.0

SM2. Chemicals

Solvents *n*-hexane (*n*-hex), dichloromethane (DCM), isooctane, and sulfuric acid were pesticide-grade and all purchased by Sigma-Aldrich, Germany. Florisil[®] adsorbent for chromatography (100–200 mesh), silica gel (70–230 mesh), potassium silicate (US EPA 1614 method), and aluminum oxide for column chromatography were all supplied by Sigma-Aldrich, Germany. PAH recovery standards (acenaphthene-d₁₀, chrysene-d₁₂, naphthalene-d₈, perylene-d₁₂, and phenanthrene-d₁₀) and PAH standard solution (mix containing all PAH target analytes) were supplied by Sigma-Aldrich (Germany), while OC/PBDE recovery standards ([¹³C₁₂]CB-101 and 153; [¹³C₁₂]BDE-28, -47, -99, -153, -154, -183) were all purchased from Wellington Labs, Canada. Pesticide (custom mix containing *p,p'* and *o,p'*-DDT, -DDD and -DDE) and PCB (custom mix containing all the target congeners) standard solutions were supplied by o2si Smart Solutions (South Carolina, USA). PBDE standard solution (custom mix containing all the target analytes) was purchased by AccuStandard (Connecticut, USA).

SM3. Instrumental conditions

The instrumental analysis of PAHs was performed using selective ion monitoring GC-MS (with a minimum of two ions), using a FocusGC coupled with a DSQ mass spectrometer (ThermoElectron, Austin, Texas). Separation of compounds was achieved using a Rxi-5MS capillary column, 60 m x 0.25 mm i.d. x 0.25 µm film thickness (Restek, Bellefonte, USA). The OC and PBDE analysis were analyzed by GC-MS/MS using a Thermo Electron TraceGC 2000 coupled with a PolarisQ Ion Trap (ThermoElectron, Austin, Texas) mass spectrometer and equipped with a PTV injector and an AS 3000 autosampler. Separation of congeners was achieved using a Rxi-5MS capillary column, 60 m x 0.25 mm i.d. x 0.25 µm film thickness (Restek, Bellefonte, USA). BDE-209 measurements were taken using a TraceGC Ultra equipped with a cold on-column injector and an ECD-40 detector (ThermoElectron, Austin, Texas) using a Rtx-5 capillary column (15 m x 0.53 mm i.d. x 0.10 µm film thickness).

GC conditions:

PAHs. GC conditions were helium as the carrier gas at 1.1 mL/min; injection temperature of 280 °C and a split-less time of 120 s. The GC oven was programmed from 110 °C (held 1.5 min) to 220 °C (held 0.1 min) at 50 °C/min, at 12 °C/min to 290 °C (held 20 min) and at 20 °C/min to 310 °C (held 2 min). The GC transfer line temperature was 290 °C and the source temperature was 250 °C.

OCs and PBDEs. GC conditions were helium as the carrier gas at 1 mL/min; injection pressure of 100 kPa; transfer pressure of 200 kPa; initial injector temperature of 70 °C (held 1 min), then raised at 14 °C/min to 270 °C (held 1 min). The initial oven temperature was 80 °C, raised to 230 °C at 50 °C/min and to 270 °C at 14 °C/min (held 20 min).

BDE-209. Injections (0.5 µL) were performed using a TriPlus autosampler (Thermo Electron) and carried out in the following analytical conditions: carrier gas helium at 6.0 mL/min; starting temperature of 100 °C (held 0.5 min) after which it was ramped to 280 °C at 15 °C/min (held 8 min). Quantitative analysis was obtained by comparing results with the external standard.

SM4. QA/QC

Table S2 - Results of Reference Material IAEA-383 (ng/g dw)

	<i>Measured (n=3)</i>							Reference	
	<i>1</i>	<i>2</i>	<i>3</i>	Mean	SD ±	RSD %	Error %	Mean	Range ±
PAHs									
Na	96.3	112.7	135.6	114.8	19.7	17	19.6	96	52-110
Ayl	61.9	55.1	42.3	53.1	10.0	19	13.0	47	31-59
Aen	17.5	22.3	17.0	18.9	2.9	15	18.4	16	13-21
F	29.9	37.6	27.3	31.6	5.4	17	17.0	27	24-34
Pn	218.4	214.1	117.6	183.4	57.0	31	14.6	160	140-190
An	21.3	30.8	23.3	25.1	5.0	20	-16.3	30	25-34
Fl	214.7	256.4	315.2	262.1	50.5	19	-9.6	290	260-350
Py	171.7	216.5	235.2	207.8	32.7	16	-25.8	280	210-350
BaA	117.0	124.2	168.7	136.6	28.0	20	30.1	105	83-130
Ch	145.9	186.5	144.4	159.0	23.9	15	-6.5	170	120-220
BbkF	175.8	200.5	144.9	173.7	27.9	16	15.8	150	96-190
BkJF	62.4	83.6	54.4	66.8	15.1	23	-8.5	73	48-76
BaP	100.3	136.5	101.8	112.9	20.5	18	-5.9	120	77-140
Per	59.3	87.2	62.3	69.6	15.3	22	20.0	58	41-130
Bghi	148.6	158.3	116.3	141.1	22.0	16	28.3	110	69-230

Table S3 - Results of Candidate Reference Material BROC-2 (ng/g dw)

	<i>Measured (n=3)</i>							<i>Reference</i>
	<i>1</i>	<i>2</i>	<i>3</i>	Mean	SD ±	RSD %	Error %	Mean
PBDE congener								
BDE-28/33	0.41	0.44	0.53	0.46	0.06	14	-27	0.63
BDE-47	10.8	9.9	11.7	10.8	0.90	8	7	10.14
BDE-49	2.5	2.7	2.7	2.6	0.12	4	-4	2.75
BDE-66	0.16	0.22	0.27	0.22	0.06	25	-25	0.29
BDE-85	0.56	0.88	0.73	0.72	0.16	22	10	0.66

BDE-99	11.6	12.7	13.4	12.6	0.91	7	-12	14.2
BDE-100	2.50	2.60	2.80	2.63	0.15	6	-13	3.04
BDE-153	1.6	1.4	1.7	1.6	0.15	10	-19	1.93
BDE-154	1.6	1.5	1.7	1.6	0.10	6	-6	1.71
BDE-183	0.38	0.54	0.56	0.49	0.10	20	10	0.45
BDE-209	1156	1301	1277	1245	77.72	6	7	1163.7

Table S4 - Results of analysis of Reference Material 1939a (ng/g dw)

	<i>Measured (n=3)</i>							<i>Certified</i>	
	<i>1</i>	<i>2</i>	<i>3</i>	<i>Mean</i>	<i>SD ±</i>	<i>RSD %</i>	<i>Error %</i>	<i>Concentration</i>	<i>SD ±</i>
PCB congener									
44	836	656	685	726	97	13	-36	1131	74
52	3694	3280	3106	3360	302	9	-22	4320	130
118	442	349	380	391	47	12	-8	423	88
138	327	235	268	277	46	17	7	258,1	6,9
149	387	278	342	336	55	16	-21	427	47
153	245	234	201	227	23	10	-24	297	19
170	60	44	73	59	15	25	-45	107	17
180	136	105	126	122	16	13	-13	140,3	6,1
194	58	49	60	56	6	10	57	35,5	4,1
4,4'-DDD	6.0	4.6	5.5	5	0.7	13	-2	5,5	0.97
4,4'-DDT	2.1	1.8	2.3	2,1	0.3	12	-23	2,7	0.42

Table S5 - Concentrations of single target analytes (a: PCBs; b: PAHs; c: PBDEs; d: DDX) expressed in ng/g OC

a.	PCBs												
	PCB 18	PCB 28+31	PCB 52	PCB 44	PCB 101	PCB 149	PCB 118	PCB 153	PCB 138	PCB 180	PCB 170	PCB 194	PCB 209
<i>Sediment 2008</i>													
LCN 24_sed	8.21	<LOD	0.65	1.82	1.53	0.55	5.02	<LOD	<LOD	<LOD	<LOD	0.08	<LOD
LCN 10_sed	2.98	2.65	<LOD	0.53	1.50	1.02	0.27	<LOD	<LOD	<LOD	<LOD	0.14	<LOD
LCN 9_sed	15.46	11.65	5.84	8.28	5.84	5.38	7.55	4.23	8.85	5.14	4.98	<LOD	1.08
LCN 5_sed	1.69	<LOD	<LOD	0.28	2.39	4.70	2.83	3.90	2.27	<LOD	<LOD	<LOD	<LOD
LCN 6_sed	3.62	<LOD	0.30	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 31_sed	3.26	<LOD	0.49	0.14	1.22	0.26	1.71	<LOD	<LOD	<LOD	<LOD	0.17	<LOD
LCN 53_sed	3.72	2.02	0.43	1.46	<LOD	<LOD	0.81	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 161_sed	3.41	2.06	1.42	2.24	1.18	0.08	1.71	0.48	<LOD	<LOD	<LOD	<LOD	<LOD
<i>Soil 2008</i>													
LUK_soil	0.13	0.18	0.08	0.21	0.06	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NAM_soil	0.03	<LOD	0.11	0.10	0.04	0.05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PER_soil	0.16	0.13	0.07	0.10	<LOD	<LOD	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 24_soil	0.25	0.26	0.13	0.28	0.06	0.11	0.05	<LOD	<LOD	0.06	<LOD	<LOD	<LOD
LCN 10_soil	0.06	0.07	0.06	0.06	0.04	0.04	0.02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 9_soil	0.47	<LOD	0.11	0.15	0.13	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

LCN 5_soil	-	-	-	-	-	-	-	-	-	-	-	-	-
LCN 6_soil	0.13	0.71	0.28	0.44	<LOD	0.23	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 32_soil	0.02	0.17	0.11	0.14	0.06	0.04	0.02	<LOD	<LOD	0.01	<LOD	<LOD	<LOD
LCN 54_soil	0.45	0.11	0.17	0.32	0.17	0.00	0.14	0.05	<LOD	0.12	0.02	<LOD	0.01
DOL_soil	0.41	0.07	0.01	0.04	<LOD	0.01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
GOK_soil	6.43	0.27	0.10	0.21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LAN_soil	0.86	0.20	0.05	0.13	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
DIG_soil	0.37	0.42	0.18	0.25	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
CHH_soil	0.91	<LOD	0.05	0.07	0.03	<LOD	0.04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
MEL_soil	0.97	<LOD	0.11	0.25	0.12	0.02	0.06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

b.	PAHs																
	1mNa	2mNa	Ayl	Aen	F	Pn	An	Fl	Py	BaA	Ch	BbkF	BaP	Per	InP	DhA	Bghi
<i>Sediment 2008</i>																	
LCN 24_sed	390.0	91.2	160.0	<LOD	110.0	1730.0	45.0	860.0	570.0	83.0	70.0	60.0	53.0	84.0	62.0	84.0	93.0
LCN 10_sed	313.4	170.8	56.3	91.6	201.0	3037.0	201.7	1441.1	1132.0	12.9	33.7	44.1	23.4	31.7	14.2	3.6	21.1
LCN 9_sed	73.3	41.2	36.6	22.1	39.7	584.3	9.3	146.4	94.7	<LOD	16.0	84.0	10.0	42.0	67.9	2.4	<LOD
LCN 5_sed	103.8	56.1	40.2	<LOD	37.5	824.8	<LOD	290.6	273.5	3.5	12.6	43.2	0.7	5.7	0.6	1.9	<LOD
LCN 6_sed	661.5	371.6	420.0	249.5	163.8	3487.2	<LOD	1147.6	556.6	9.0	18.5	151.2	5.2	13.0	12.0	7.0	<LOD
LCN 31_sed	139.7	77.5	43.5	22.5	61.0	1520.6	12.8	272.8	275.0	13.4	32.3	118.6	13.0	24.6	23.7	26.8	18.0
LCN 53_sed	70.9	28.3	27.3	16.9	14.8	498.2	14.8	134.8	88.9	<LOD	12.4	78.3	4.8	10.4	10.8	0.8	8.1
LCN 161_sed	1227.0	245.9	221.6	7.5	48.0	852.4	<LOD	488.6	271.9	50.7	31.0	74.5	10.5	83.1	22.6	12.0	28.3
<i>Soil 2008</i>																	
LUK_soil	14.0	11.6	14.0	11.6	11.6	53.5	<LOD	7.0	7.0	1.2	3.5	5.8	2.3	4.7	4.7	3.5	7.0
NAM_soil	4.8	2.9	7.1	2.9	4.8	40.5	<LOD	23.8	23.8	31.0	40.5	52.4	45.2	16.7	45.2	16.7	71.4
PER_soil	42.9	18.2	3.6	<LOD	8.4	21.6	<LOD	3.9	2.5	<LOD	1.5	1.5	<LOD	3.4	0.7	<LOD	<LOD
LCN 24_soil	7.3	0.7	10.0	<LOD	13.3	63.3	<LOD	10.0	10.0	2.7	3.3	3.3	<LOD	<LOD	11.7	<LOD	30.7
LCN 10_soil	25.9	12.2	<LOD	<LOD	3.1	8.4	<LOD	<LOD	1.3	1.2	3.2	0.3	<LOD	1.3	<LOD	<LOD	<LOD
LCN 9_soil	<LOD	0.6	0.6	<LOD	17.1	44.3	<LOD	6.9	2.9	2.3	2.0	3.4	<LOD	2.9	<LOD	<LOD	<LOD
LCN 5_soil	50.4	15.0	<LOD	<LOD	9.4	29.4	1.2	6.5	5.4	0.6	3.9	4.2	2.0	1.9	2.7	<LOD	3.9
LCN 6_soil	53.0	41.3	<LOD	8.8	17.3	86.0	<LOD	16.0	10.8	5.8	3.0	4.5	1.3	5.3	1.3	<LOD	1.5
LCN 32_soil	8.8	5.3	16.8	2.5	3.5	36.8	<LOD	5.3	3.5	0.9	1.1	2.1	0.5	5.3	0.9	<LOD	36.8

LCN 54_soil	27.5	21.3	18.8	0.0	18.8	125.0	<LOD	25.0	18.8	1.9	15.0	15.6	6.9	3.1	8.8	<LOD	13.1
DOL_soil	10.0	10.1	3.8	2.4	2.2	5.4	<LOD	1.0	1.1	0.2	1.1	2.0	1.0	<LOD	1.9	<LOD	<LOD
GOK_soil	119.4	57.3	9.7	<LOD	31.9	91.1	<LOD	11.2	4.3	0.9	3.9	2.6	2.9	<LOD	<LOD	<LOD	<LOD
LAN_soil	23.6	11.7	4.4	<LOD	5.3	13.7	0.6	2.1	1.3	0.2	0.6	2.0	<LOD	0.8	0.5	<LOD	<LOD
DIG_soil	31.1	21.4	5.8	5.1	7.9	28.5	<LOD	2.9	2.9	1.7	0.8	0.9	<LOD	2.8	1.5	<LOD	<LOD
CHH_soil	12.2	3.5	3.9	<LOD	6.3	2.7	<LOD	3.1	2.9	<LOD	1.4	2.7	<LOD	4.1	<LOD	<LOD	45.1
MEL_soil	6.9	<LOD	6.3	<LOD	13.1	59.4	<LOD	6.9	6.9	<LOD	1.6	7.8	<LOD	7.2	<LOD	<LOD	53.1

Soils 2012

LUK_soil	14.3	7.9	12.7	14.3	14.3	71.3	<LOD	6.3	30.1	3.2	7.9	11.1	<LOD	103.0	9.5	<LOD	<LOD
NAM_soil	8.9	3.0	4.5	<LOD	3.0	17.9	<LOD	6.0	11.9	4.5	16.4	32.7	<LOD	41.7	22.3	8.9	17.9
PER_soil	12.3	3.1	1.5	<LOD	6.1	5.1	<LOD	9.2	7.7	6.1	13.8	24.6	4.6	20.0	4.6	9.2	7.7
PYR_soil	<LOD	<LOD	<LOD	<LOD	<LOD	18.8	<LOD	<LOD	4.4	<LOD	<LOD	<LOD	<LOD	19.9	<LOD	<LOD	<LOD
LCN 10_soil	75.4	19.5	12.2	17.0	21.9	26.8	<LOD	51.1	116.8	12.2	18.2	104.6	2.4	73.0	<LOD	<LOD	<LOD
LCN 9_soil	49.5	9.2	22.3	30.7	22.6	81.3	<LOD	67.1	109.5	15.9	24.7	134.3	7.1	102.5	<LOD	<LOD	<LOD
LCN 7a_soil	<LOD	0.8	0.8	3.8	17.3	19.2	<LOD	3.8	2.3	3.1	3.8	1.5	2.7	69.2	<LOD	<LOD	<LOD
LCN 7b_soil	<LOD	<LOD	<LOD	<LOD	<LOD	76.7	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	116.3	<LOD	<LOD	<LOD

c.	PBDEs							
	PBDE 28	PBDE 47	PBDE 99	PBDE 100	PBDE 153	PBDE 154	BDE 183	BDE 209
<i>Sediment 2008</i>								
LCN 24_sed	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 10_sed	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	72.73
LCN 9_sed	<LOD	<LOD	15.00	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 5_sed	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	57.14
LCN 6_sed	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	60.00
LCN 31_sed	<LOD	<LOD	3.00	<LOD	<LOD	<LOD	<LOD	100.00
LCN 53_sed	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 161_sed	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	42.86
<i>Soil 2008</i>								
LUK_soil	<LOD	0.93	0.47	<LOD	<LOD	<LOD	<LOD	<LOD
NAM_soil	<LOD	0.71	0.24	<LOD	<LOD	<LOD	<LOD	2.38
PER_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 24_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 10_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 9_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 5_soil	-	-	-	-	-	-	-	-
LCN 6_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 32_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

LCN 54_soil	<LOD	<LOD	1.25	<LOD	2.50	<LOD	<LOD	<LOD
DOL_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
GOK_soil	<LOD	3.33	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LAN_soil	<LOD	<LOD	1.18	<LOD	<LOD	<LOD	<LOD	<LOD
DIG_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
CHH_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
MEL_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

d.	DDx					
	o,p'-DDD	o,p'-DDE	o,p'-DDT	p,p'-DDD	p,p'-DDE	p,p'-DDT
<i>Sediment 2008</i>						
LCN 24_sed	<LOD	<LOD	<LOD	6.00	8.00	<LOD
LCN 10_sed	<LOD	<LOD	<LOD	<LOD	1.82	<LOD
LCN 9_sed	<LOD	<LOD	<LOD	30.00	15.00	<LOD
LCN 5_sed	<LOD	<LOD	<LOD	5.71	12.86	<LOD
LCN 6_sed	<LOD	<LOD	<LOD	8.00	22.00	<LOD
LCN 31_sed	<LOD	<LOD	<LOD	4.00	4.00	<LOD
LCN 53_sed	<LOD	<LOD	<LOD	<LOD	1.25	<LOD
LCN 161_sed	<LOD	<LOD	<LOD	<LOD	3.57	<LOD
<i>Soil 2008</i>						
LUK_soil	<LOD	0.93	<LOD	0.47	2.33	<LOD
NAM_soil	<LOD	0.71	<LOD	1.90	1.90	<LOD
PER_soil	<LOD	<LOD	<LOD	<LOD	2.43	<LOD
LCN 24_soil	<LOD	<LOD	<LOD	<LOD	1.33	<LOD
LCN 10_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 9_soil	<LOD	<LOD	<LOD	<LOD	1.71	<LOD
LCN 5_soil	-	-	-	-	-	-
LCN 6_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
LCN 32_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

LCN 54_soil	<LOD	<LOD	<LOD	1.88	1.88	<LOD
DOL_soil	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
GOK_soil	<LOD	3.33	<LOD	8.89	<LOD	<LOD
LAN_soil	<LOD	<LOD	<LOD	1.18	1.76	<LOD
DIG_soil	<LOD	<LOD	<LOD	1.76	3.53	<LOD
CHH_soil	<LOD	<LOD	<LOD	<LOD	0.39	<LOD
MEL_soil	<LOD	<LOD	<LOD	<LOD	1.25	<LOD

Table S6. LMW/HMW-PAH ratios in sediment and soil (2008-2012) samples.

Valley		Sediments - 2008	
Khumbu	LCN 24_sed	6.7	
Khumbu	LCN 10_sed	36.0	
Khumbu	LCN 9_sed	4.7	
Khumbu	LCN 5_sed	23.8	
Khumbu	LCN 6_sed	32.7	
Imja	LCN 31_sed	9.0	
Imja	LCN 53_sed	7.1	
Imja	LCN 161_sed	10.8	
		Soils - 2008	Soils - 2012
Pharak	LUK_soil	4.0	5.4
Pharak	NAM_soil	0.3	0.5
Khumbu	PER_soil	14.3	0.6
Khumbu	LCN 24_soil	2.2	-
Khumbu	LCN 10_soil	8.4	2.5
Khumbu	LCN 9_soil	6.8	2.2
Khumbu	PYR_soil	-	-
Khumbu	LCN 7a_soil	-	4.3

Khumbu	LCN 7b_soil	-	-
Khumbu	LCN 5_soil	6.1	
Khumbu	LCN 6_soil	10.4	
Imja	LCN 32_soil	1.7	
Imja	LCN 54_soil	4.0	
Gokyo	DOL_soil	5.8	
Gokyo	GOK_soil	31.8	
Thame	LAN_soil	15.2	
Thame	DIG_soil	13.7	
Thame	CHH_soil	0.7	
Thame	MEL_soil	1.4	

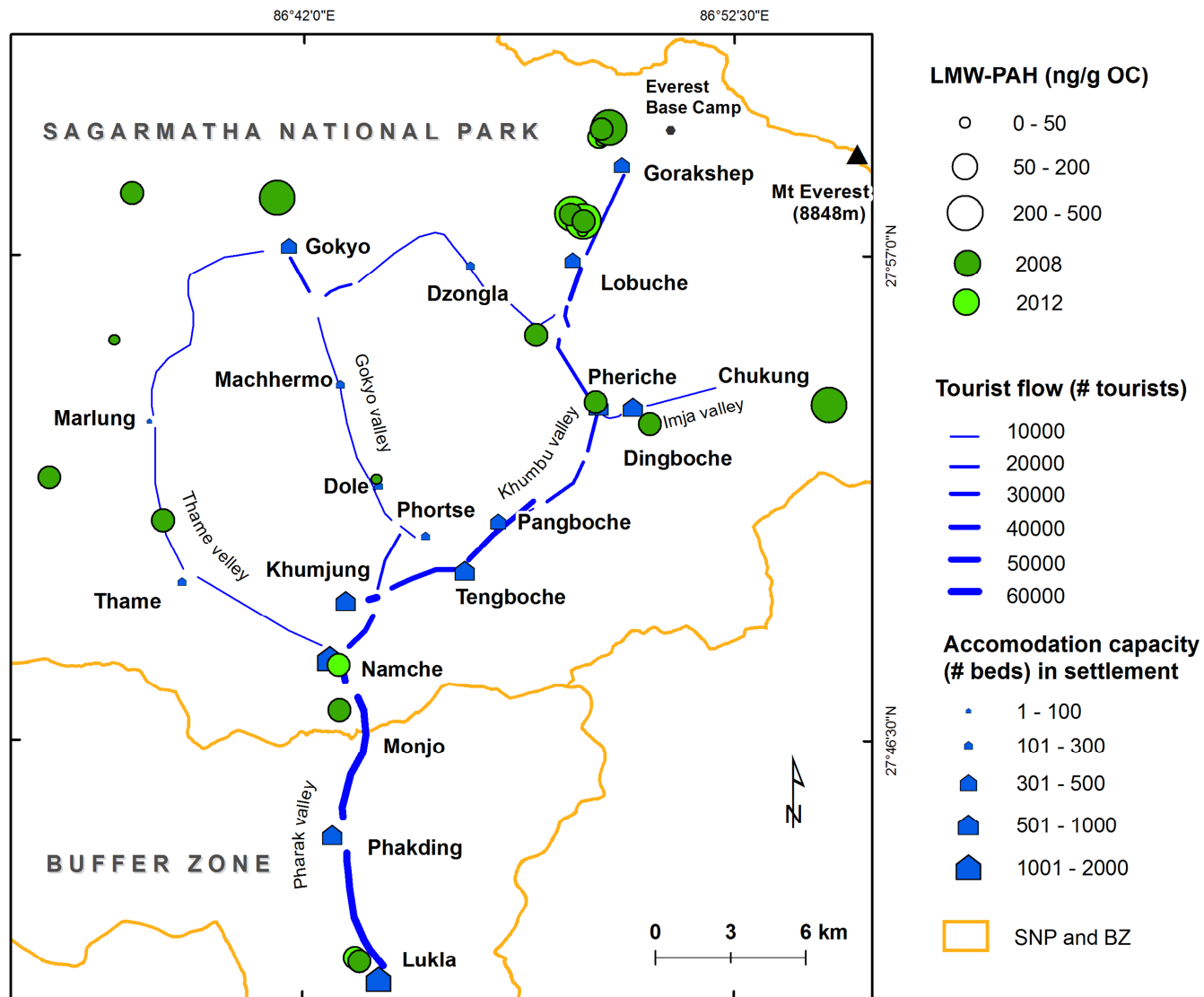


Fig. SM1. LMW-PAHs (ng/g OC) in soil samples (2008 and 2012), touristic flow and accommodation capacity of settlements

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