

Environmental & Analytical **Toxicology**

Research Article

Cizmas et al., J Environ Anal Toxicol 2015, 5:2 http://dx.doi.org/10.4172/2161-0525.1000261

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Measurement of Organophosphate Pesticides, Organochlorine Pesticides, and Polycyclic Aromatic Hydrocarbons in Household Dust from Two Rural Villages in Nepal

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Abstract

Although there are few published studies of residential exposures to environmental contaminants in Nepal, there may be substantial exposures to multiple contaminants in Nepali households. Pesticides, which can be harmful to human health, are often used by Nepali farmers, and many farmers lack an understanding of the appropriate procedures for the safe use, handling and storage of pesticides. In addition, many Nepali families use wood burning stoves, leading to the potential for exposure to polycyclic aromatic hydrocarbons from wood smoke. This study measured the levels of four organophosphate pesticides, 22 organochlorine pesticides, and over thirty polycyclic aromatic hydrocarbons in house dust from two rural Nepali villages. Floor dust samples were collected in early summer from a total of 18 households, including nine households in the village of Keraghari, in Kavrepalanchok District, and nine homes in the village of Kafaldanda, in Lalitpur District. These villages have similar environmental features and are located at an altitude of approximately 2,000 meters. In these two villages, many of the homes have improved cookstoves to reduce smoke levels in the homes. The dust samples were collected using pre-ashed glass fiber filter cloths saturated with isopropyl alcohol. In both villages, the organochlorine pesticide that was present in the highest concentrations was the DDT metabolite p,p'-dichlorodiphenyldichloroethylene (4,4'-DDE). Among the organophosphates, methyl parathion accounted for much of the organophosphate mass detected. Across both villages, the median total organochlorine pesticide value (287 ng/m²) was 5-fold higher than the median total organophosphate pesticide value (54.3 ng/m²). The median total polycyclic aromatic hydrocarbon concentration in house dust from both villages was 14,700 ng/m². Interventions are needed to improve safe handling and use of pesticides in Nepali villages. Additional studies are also needed to assess the extent to which improved cookstoves reduce polycyclic aromatic hydrocarbon exposure, during both winter and summer.

Keywords: Nepal; Pesticide; Organochlorine; Organophosphate; Polycyclic aromatic hydrocarbon; Exposure; House dust; Environmental health

Abbreviations: GC/MS: Gas Chromatography/Mass Spectrometry; DDD: Dichlorodiphenyldichloroethane; 4,4'-DDE: p,p'-Dichlorodiphenyldichloroethylene; DDT: Dichlorodiphenyltrichloroethane; FAO-N: Food and Agriculture Organization of the United Nations – Nepal; OC: Organochlorine Pesticide; OP: Organophosphate Pesticide; NGO: Non-Governmental Organization; PAH: Polycyclic Aromatic Hydrocarbon; cPAH: Carcinogenic Polycyclic Aromatic Hydrocarbon; tPAHs: Total Polycyclic Aromatic Hydrocarbons; UNDP: United Nations Development Program; UNWFP-N: United Nations World Food Programme – Nepal; USAID: United States Agency For International Development; USEPA: United States Environmental Protection Agency

Introduction

Pesticides include a wide variety of compounds that serve to control unwanted pests such as insects, weeds and fungi. They are used in agricultural, residential, commercial, and school settings, both indoors and outdoors. Human exposure to pesticides can occur through numerous pathways. Dietary ingestion is an important route of exposure to pesticides [1,2]. Water contamination due to agricultural or non-agricultural runoff can also result in pesticide exposure among humans [3,4]. Children are particularly vulnerable to pesticide exposure because they breathe, drink and eat more per unit body weight than adults [5]. In addition, young children may crawl or exhibit mouthing behavior, during which hands or non-food objects are put

into or touch the mouth, and this can result in ingestion of pesticides from contaminated surfaces [6,7]. Humans are often exposed to multiple pesticides, either at the same time or serially [8].

The families of agricultural workers may experience heightened pesticide exposures due to the take home pathway [9]. One study of farmworkers in the state of Washington in the United States found that during the thinning season, urinary metabolites of organophosphate dimethyl pesticides were significantly higher in the children of farmworkers than in non-farmworker children [10]. In addition, among individuals who work in conventional agriculture where pesticides are used, pesticide concentrations have been found to be elevated in vehicle and house dust when compared to a reference population [10,11].

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Received December 20, 2014; Accepted January 12, 2014; Published January 16, 2015

Citation: Cizmas L, Ackerman L, Donnelly DA, Donnelly KC, McDonald TJ (2015) Measurement of Organophosphate Pesticides, Organochlorine Pesticides, and Polycyclic Aromatic Hydrocarbons in Household Dust from Two Rural Villages in Nepal. J Environ Anal Toxicol 5: 261. doi:10.4172/2161-0525.1000261

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As of 2012, Nepal had a population of 27.5 million and a gross national income per capita of US\$700 [12]. The United Nations Development Program's Human Development Index is a method for evaluating the development status of a country based on multiple factors such as life expectancy, gross national income per capita, and mean years of schooling [13]. Out of 187 countries that are ranked on the United Nations Development Program's Human Development Index, Nepal was ranked #145 [13]. The US Agency for International Development (USAID) estimates that over 70% of the population is employed in agriculture [14]. Nepal produces a variety of crops, including rice, maize, wheat, millet, soybeans, lentils, potatoes, oilseeds, and vegetables [15]. The first pesticide utilized in Nepal was dichlorodiphenyltrichloroethane (DDT), which was used to fight malaria during the early 1950's [16]. In subsequent years, a number of other pesticides were used, including organochlorines such as dieldrin and chlordane, organophosphates including methyl parathion and malathion, carbamates, and synthetic pyrethroids [16]. Pesticide use in Nepal has steadily increased, and an estimated 178 tons of pesticides were used in this country in 2003 [16]. However, many farmers lack adequate understanding of the health effects of pesticides and appropriate methods for pesticide use and handling [16,17]. Agricultural workers with inadequate information about appropriate safety measures or protective equipment may experience far higher levels of exposure to pesticides than those who have sufficient training and equipment [8]. Multiple problems have been seen in developing countries with agricultural hygiene, including inadequate safeguards on the storage and management of agricultural chemicals, inadequate procedures for disposing of unwanted or outdated pesticides, insufficient safety training, and unsafe procedures for pesticide application, including failure to use personal protective equipment [18-20]. This lack of agricultural hygiene increases the risk of pesticide exposure, for agricultural workers and also their families, due to pesticides that are unwittingly taken home on clothing, shoes and other items.

Adverse health outcomes associated with OP exposure may be acute, typically occurring shortly after exposure, or chronic, due to multiple exposures over a longer time period. Acute OP exposures have been associated with a variety of adverse health effects including tachycardia, anxiety, seizures, muscle weakness, twitching, sweating, salivation, tearing, and diarrhea, bronchospasm, convulsions and coma [8,21]. Death may occur due to pulmonary dysfunction and respiratory failure [22]. The effects of chronic exposure are less well characterized. OP-exposed tree fruit farmers in one study had significantly slower reaction times in the dominant hand than unexposed individuals [23]. In a cross-sectional study, Stephens et al. [24] found that when compared to control individuals, sheep farmers who had been exposed to OPs fared significantly worse on tests to evaluate speed of information processing and sustained attention. Prenatal OP exposure is associated with increased abnormalities in reflexes in neonates [25] and in infants 2 months old or younger [26]. Prenatal exposure to OPs has also been found to be associated with pervasive developmental problems and lower mental development scores at 2 years of age [27], with attention problems at 5 years of age [28], and with lower cognitive scores in 7-year old children [29]. OP exposure has also been associated with changes in semen parameters [30]. Recio-Vega et al. [31] found that among men with high urinary organophosphate pesticide (OP) levels, the total sperm count was significantly decreased. Yucra et al. [32] found a significant association between high levels of ethylated OP metabolites in urine and low seminal volume in Peruvian pesticide applicators. In addition, a case-control study in the People's Republic of China found that men with higher urinary levels of OP dimethylphosphate metabolites were more likely to have low sperm motility and concentration [33].

The effects of acute organochlorine pesticide (OC) exposure include seizures, dizziness, headache, vomiting, twitching, and incoordination [21]. Chronic exposure to OCs has been associated with endocrine disruption [30]. The DDT metabolite p,p'dichlorodiphenyldichloroethylene (4,4'-DDE) exhibits antiandrogenic effects in fetal, pubertal, and adult male rats [34]. A cross-sectional study of young men in a malaria-affected area of South Africa found that lipid-adjusted 4,4'-DDE percentile concentrations in blood plasma were significantly positively associated with low concentrations of spermatozoa, while reduced sperm mobility was significantly positively associated with lipid-adjusted p,p'-DDT percentile concentrations [35]. In a separate study of men in the United States, there was a significant negative association between summed serum level of DDT and 4,4'-DDE and sperm concentration, sperm motility, and sperm

PAH Compound	CAS RN	Molecular Weight	No. of Rings ^b
Naphthalene ^a	91-20-3	128	2
C1-, C2-, C3-, C4-Naphthalenes		142,156, 170,184°	2
Benzothiophene	95-15-8	134	2
C1-, C2-, C3-Benzothiophenes		148, 162, 176°	2
Biphenyl	92-52-4	154	2
Acenaphthylene ^a	208-96-8	152	3
Acenaphthene ^a	83-32-9	154	3
Dibenzofuran	132-64-9	168	3
Fluorene ^a	86-73-7	166	3
C1-, C2-, C3-Fluorenes		180, 194, 208°	3
Carbazole	86-74-8	167	3
Anthracene ^a	120-12-7	178	3
Phenanthrene ^a	85-01-8	178	3
C1-, C2-, C3-, C4-Phenanthrenes/ Anthracenes		192, 206, 220, 234º	3
Dibenzothiophene	132-65-0	184	3
C1-, C2-, C3-Dibenzothiophenes		198, 212, 226°	3
Fluoranthene ^a	206-44-0	202	4
Pyrene ^a	129-00-0	202	4
C1-, C2-, C3-Fluoranthenes/Pyrenes		216, 230, 244°	4
Naphthobenzothiophene	205-43-6	234	4
C1-, C2-, C3- Naphthobenzothiophenes		248, 262, 276°	4
Benz(a)anthracene ^a	56-55-3	228	4
Chrysene ^a	218-01-9	228	4
C1-, C2-, C3-, C4-Chrysenes		242, 256, 270, 284°	4
Benzo(b)fluoranthene ^a	205-99-2	252	5
Benzo(k)fluoranthene ª	207-08-9	252	5
Benzo(e)pyrene	192-97-2	252	5
Benzo(a)pyreneª	50-32-8	252	5
Perylene	198-55-0	252	5
Dibenzo(a,h)anthracene ª	53-70-3	278	5
Benzo(g,h,i)perylene ^a	191-24-2	276	6
Indeno(1,2,3-c,d)pyrene ^a	193-39-5	276	6

a. The US EPA considers these analytes to be priority pollutants. Analytes that are italicized are considered probable human (B2) carcinogens by the USEPA.
b. Number of aromatic 5- and 6-carbon rings. Low molecular weight compounds have 2 or 3 aromatic rings and high molecular weight compounds have 4 or more aromatic rings.

c. The molecular weights for the alkylated compounds are shown in order of C1, C2, C3, etc.

 Table 1: Polycyclic aromatic hydrocarbons (PAHs) quantified in extracts of house dust from two rural villages in Nepal.

morphology [36]. DDT, 4,4'-DDE and DDD are also categorized by the U.S. Environmental Protection Agency (USEPA) as probable human carcinogens (class B2) [37-39].

Polycyclic aromatic hydrocarbons (PAHs) are produced by both natural and human sources including wood fires for cooking, forest fires, coal combustion, tobacco smoking, motor vehicle use, and refuse incineration. They are ubiquitous in the environment and persist indoors and outdoors. In general, their volatility and aqueous solubility are low, and in homes they are typically found adsorbed to particles such as dust. PAHs typically occur as mixtures, and as such, it has not been possible to examine direct evidence regarding the carcinogenicity in humans of individual PAHs. However, the USEPA has designated seven PAHs as probable human carcinogens (Table 1). In addition, in a study of inner city children in New York, New York, USA, high prenatal PAH exposure was associated with an increased risk of cognitive developmental delay, and at 3 years of age was also associated with a lower mental development index [40].

An estimated 2.4 billion people worldwide burn solid fuels including wood for household purposes including cooking [41]. In a critical review of the literature, Smith et al. [42] found a strongly increased risk of acute respiratory infections in young children exposed to smoke from biomass burning, when compared to unexposed children. In the rural areas of Nepal, over 90% of households cook with an open fire with no smoke ventilation out of the home. In addition, over 75% of rural Nepali homes use wood, coal, lignite, or charcoal for cooking [43]. This leads to the potential for sharply elevated levels of PAHs in the home, particularly during the cold winter months. Efforts are being made to promote the use of improved stoves in Nepal, and the majority of the homes in this study were equipped with a pipe to vent smoke from the kitchen and reduce smoke exposure as described in Section 2.1.

Environmental contaminants may be found in contaminated house dust [44], including PAHs [45] and pesticides [6]. House dust can be an important source of exposures indoors, particularly for children who spend time playing or crawling on the floor and who exhibit frequent hand-to-mouth behavior [6,7,46]. Few data have been collected on the levels of PAHs and pesticide residues in the homes of Nepali farmers. The objective of this study was to measure the levels of OPs, OCs and PAHs in residential floor dust in Nepal.

Materials and Methods

Sampling locations

Dust samples were collected from a total of 18 households, including nine homes from the village of Keraghari, in Kavrepalanchok District, and nine households from the village of Kafaldanda, in Lalitpur District. Both villages are located at an altitude of approximately 2,000 meters, and have similar environmental features as they are located on mountain ridges and rely on seasonal rains, rather than irrigation, for agriculture. The samples were collected in early summer (June, 2008). The homes that were sampled for this study used wood burning stoves for heating and cooking. The majority of the homes in this study had stoves that were equipped with *chulos*, a simple pipe which vents wood-fire smoke from the kitchen to outside of the home, thereby reducing smoke and PAH levels inside the home.

Dust sample collection

To evaluate the levels of pesticides and PAHs in the homes,

household dust samples were collected and analyzed. The samples were collected from inside the home in front of the front door, over an area of approximately 30 X 30 cm. The floors were all compacted red clay. The sample area was measured and markers were placed to signify the area. To collect the dust, a pre-ashed glass fiber filter cloth (type A/E, Gellman Sciences, Ann Arbor, MI) was saturated with isopropyl alcohol and used to wipe all the dust from the marked area according to the method of Naspinski et al. [45]. The cloth was then placed into an aluminum foil pouch inside a Ziploc bag, and transported to the laboratory.

Sample extraction

A Dionex ASE2000 accelerated solvent extractor (Dionex, Sunnyvale, CA) was used to extract various organics from pre-dried samples using a modified operating procedure based on USEPA SW-846 Method 3545A [47,48]. The extractions were accomplished with 100% dichloromethane inside stainless-steel extraction cells held at elevated solvent pressure and temperature. Appropriate surrogate standards were added to the accelerated solvent extraction vials prior to extraction. The extracted compounds dissolved in the hot solvent were collected in 60mL glass vials. Extracts were concentrated to a volume of 1-3mL using a Zymark TurboVap II (Caliper Life Sciences, Hopkinton, MA). If necessary, samples were processed through a clean-up column in order to minimize matrix interference. The clean-up procedure was based on USEPA SW-846 Methods 3610B, 3630C, and 3660B [49-51]. Briefly, the method entailed using a Restek RESPREP 6cc polypropylene cartridge, 1000 mg of silica with an addition of 1g of alumina (5% (w/w) reagent water), a small layer (4-5 mm) of sodium sulfate and a 1-2 mm layer of pre-cleaned copper. Extracts were then concentrated to a final volume of 1mL and appropriate internal standards were added in preparation for chemical analysis.

OP and PAH determination

Quantification of PAHs and OPs was accomplished by capillary gas chromatography/mass spectrometry (GC/MS) in selected ion monitoring mode. The PAH quantification method was based on USEPA SW-846 Method 8270D [52,53]. The OP quantification method was based on USEPA SW-846 Methods 8270D and 8141B [52,54]. The PAHs that were quantified are shown in Table 1. The high molecular weight PAHs are those with four or more rings. The OPs that were quantified were diazinon, malathion, methyl parathion, and chlorpyrifos. The gas chromatograph was temperature-programmed and operated in splitless mode. The capillary column was an Agilent Technologies HP-5MS (30 m long by 0.25 mm ID and 0.25 μm film thickness) (Agilent Technologies, Pickering, Ontario, Canada). Carrier flow was accomplished using electronic pressure control. The autosampler was able to make 1 to 5 µL injections. The mass spectrometer had the capacity to scan from 35 to 500AMU every second, or less, and used 70 volts electron energy in electron impact ionization mode. During analysis, the data acquisition system allowed continuous acquisition and storage of all data and was able to show ion abundance versus scan number or time. The reporting limit for PAHs and OPs was 10 ng/m². In cases where analytes were detectable at levels below the reporting limit, a value of <10 was shown, and a value of 5 ng/m² was assigned for calculations.

OC determination

The OCs that were quantified are shown in Table 2. Quantification was performed by gas chromatography/electron capture detector based on USEPA SW-846 Method 8081B [53,55]. The gas chromatograph was

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	II			II				
	Mean	Median	Minª	Max	Mean	Median	Minª	Max
OC pesticide								
Alpha-HCH	15.8	10.8	4.8	53.3	15.5	5.8	<1	98.5
Gamma-HCH	16.2	16.3	5.9	32.2	6.6	6.3	<1	16.5
Beta-HCH	6.7	4.8	<1	23.8	3.8	<1	<1	15.2
Heptachlor	12.4	12.1	1.4	22.2	12.5	10.3	<1	37.4
Delta-HCH	50.5	56.7	<1	110	29.0	8.3	<1	90.4
Aldrin	1.8	1.9	<1	3.8	2.7	1.5	<1	9.9
Heptachlor-epoxide	48.3	37.3	5.3	157	8.1	8.9	<1	14.7
Gamma-Chlordane	5.8	4.2	<1	21.8	2.9	1.8	<1	9.4
Trans-Nanochlor	2.9	3.1	<1	6.5	6.5	5.9	<1	16.8
Endosulfan-I	5.4	4.3	<1	13.4	1.5	1.3	<1	3.2
Alpha-Chlordane	4.3	2.4	<1	10.6	2.1	<1	<1	6.8
Dieldrin	15.5	14.5	1.9	42.1	8.1	8.3	<1	27.0
4,4'-DDE	1609	357	<1	11,500	507	60.1	<1	3,920
DDD	<1	<1	<1	<1	<1	<1	<1	<1
DDT	14.4	13.0	9.2	24.9	8.8	8.9	<1	18.7
Endrin	7.5	<1	<1	50.0	1.0	<1	<1	3.5
Cis-nanochlor	16.3	13.4	7.6	32.5	16.5	15.9	<1	31.5
Endosulfan-II	6.2	3.8	<1	17.3	2.4	1.1	<1	5.7
Endrin-aldehyde	22.4	30.6	2.3	43.7	17.6	19.2	<1	37.1
Endosulfan Sulfate	10.1	5.7	<1	44.6	3.4	2.1	<1	10.4
Endrin Ketone	6.8	<1	<1	44.4	1.8	<1	<1	7.4
Methoxychlor	11.2	11.3	<1	27.6	2.8	<1	<1	10.2
Total OCs ^ь	1890	598	116	11,900	661	231	11.0	4,200

^a Reporting limit is 1 ng/m². When samples had detectable levels of a pesticide below the reporting limit, this was shown as "<1" in the table and included as 0.5 ng/m² in the calculations.

^bThe median, minimum and maximum values for total OCs were calculated by summing the total OC value for each home, then selecting the median, minimum and maximum values from the total OC values for all the homes in either Keraghari or Kafaldanda. Abbreviations: HCH, hexachlorocyclohexane; DDT, dichlorodiphenyltrichloroethane; 4,4'-DDE, dichlorodiphenyldichloroethylene; DDD, dichlorodiphenyldichloroethane.

Table 2: Mean, median, minimum and maximum levels^a of organochlorine pesticides (OCs) and of total OCs in house dust samples from Keraghari and Kafaldanda, Nepal (ng/m²).

	DDT (ng/m²)	DDT as a % of total OCs ^a	4,4'-DDE (ng/m ²)	4,4'-DDE as a % of total OCs ^b	Total OCs (ng/m ²)
Median	11.2	2.9%	66	24.8%	287
Minimum ^c	0.5	0.1%	0.5	0.3%	11.0
Maximum	24.9	14.3%	11,500	97.0%	11,900

^aThe median, minimum and maximum values for DDT as a % of total OCs were calculated by determining this percentage for each home individually, then selecting the median, minimum and maximum values out of the values for all homes in both villages.

^bThe median, minimum and maximum values for 4,4'-DDE as a % of total OCs were calculated in the same manner as the calculations for DDT (see footnote a).

°The reporting limit for the OC pesticides was 1 ng/m². Levels of pesticides that were detectable but below the reporting limit were assigned a value of 0.5 ng/m².

 Table 3: Levels (ng/m²) of dichlorodiphenyltrichloroethane (DDT) and dichlorodiphenyldichloroethylene (4,4'-DDE) across all house dust samples in both Kafaldanda and Keraghari, Nepal, and levels of each as a percentage of total organochlorine pesticides (OCs).

temperature-programmed and operated in splitless mode. The capillary column used was a J&W DB-17HT (30 m long by 0.25 mm ID and 0.25 μ m film thickness). Carrier flow was achieved by electronic pressure control. The auto-sampler was capable of making 1 to 5 μ l injections. The data acquisition system was by HP Chemstation software (Hewlett Packard/Agilent Technologies, Pickering, Ontario, Canada), capable of acquiring and processing gas chromatograph data. The reporting limit for chlorinated pesticides was 1 ng/m². Analytes that were detectable at levels below the reporting limit were shown with a value of <1, and a value of 0.5 ng/m² was assigned for calculations.

Statistical analysis

Statistical analysis was conducted using the SigmaPlot 12.5 software (Systat Software, Inc., Richmond, CA) or Microsoft Excel 2010 (Microsoft, Redmond, WA). Descriptive statistics were calculated for each set of analytes.

Results

OCs and OPs

The levels of OCs in the house dust samples in both locations are presented in Table 2. Median and maximum total OC values were 598 and 11,900 ng/m², respectively, in Keraghari, and 231 and 4,200 ng/m², respectively, in Kafaldanda. In both villages, 4,4'-DDE was present in far higher concentrations that any other individual OC. In Keraghari, the median and maximum values of 4,4'-DDE were 357 and 11,500 ng/m², respectively, while in Kafaldanda, the median and maximum 4,4'-DDE values were 60.1 and 3,920 ng/m², respectively. DDT was present in much lower concentrations, with maximum values in Keraghari and Kafaldanda of 24.9 and 18.7 ng/m², respectively. Table 3 presents the mass per m² floor area of DDT and 4,4'-DDE across both villages. The percentage of total OC mass that was 4,4'-DDE varied widely across



Figure 1: Mass of methyl parathion (ME) and total organophosphate pesticides (total OPs) (ng/m²) in house dust samples from Keraghari (Ke) and Kafaldanda (Ka), Nepal.

	Total OCs (ng/m ²)	Total OPs (ng/m ²)
Median	287	54.3
Minimum	11.0	15.8
Maximum	11,900	543

 Table 4: Comparison of the levels (ng/m²) of total organochlorine pesticides (OCs) and organophosphate pesticides (OPs) in house dust samples from both Keraghari and Kafaldanda, Nepal.



the homes, with median, minimum and maximum values of 24.8%, 0.3%, and 97.0%, respectively. In contrast, the percentage of total OC mass contributed by DDT was lower across all homes, with median, minimum and maximum values of 2.9%, 0.1% and 14.3%, respectively. The level of DDD in all samples was below the reporting limit of 1 ng/ m^2 (Table 2).

Figure 1 presents the levels of methyl parathion and total OPs in both villages. Methyl parathion accounted for much of the OPs detected in both locations. The median values of methyl parathion and total OPs in Keraghari were 19.2 and 25.4 ng/m², respectively, and in Kafaldanda were 136 and 143 ng/m², respectively. Diazinon, malathion, and chlorpyrifos were present in both villages at detectable but very low levels, with the maximum concentrations of these three OPs below the reporting limit of 10 ng/m² in both villages (data not shown).

Across both villages, the OCs were present at higher levels (by mass) than the OPs (Table 4). The median total OC value (287 ng/m^2) was 5-fold higher than the median total OP value (54.3 ng/m^2), while the maximum total OC value of 11,900 ng/m² was over 20-fold higher than the maximum total OP value of 543 ng/m^2 .

PAHs

The total carcinogenic PAH (cPAH) and total PAH (tPAH) levels are presented in Figure 2. The cPAHs are the sum of the seven PAHs classified by the U.S. Environmental Protection Agency as probable human (category B2) carcinogens, and include benz(a)anthracene, chrysene, benzo(dibenzo(a,h)anthracene, and indeno(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, and indeno(1,2,3-c,d)pyrene. All dust samples in both villages had detectable levels of cPAHs and tPAHs. The median PAH levels in both villages were similar, with median cPAH levels of 2,060 and 1,890 ng/ m² in Keraghari and Kafaldanda, respectively, and median tPAH values of 14,700 and 14,700 ng/m² in Keraghari and Kafaldanda, respectively.

Discussion and Conclusions

In the Nepali homes in this study, the OCs were present at higher levels by mass than the OPs. This may be due partly to the longer halflives of many OCs when compared to the OPs. Of the OCs that were assessed in these samples, the compound with the highest median and maximum concentrations was 4,4'-DDE. In a study by Bradman and colleagues [56], the median and maximum levels of 4,4'-DDE in floor wipe samples from the homes of California farmworker children were not detectable and 0.15 ng/cm², respectively. In comparison, in the present study, when the data from both villages were combined, the median and maximum values of 4,4'-DDE were 0.0066 ng/cm² and 1.15 ng/cm², respectively, with the maximum value in the Nepali homes more than seven-fold higher than the maximum value in the California farmworker homes. The analyte p,p'-DDT was not detectable in any of the farmworker homes studied by Bradman et al. [56], whereas p,p'-DDT was detected in all the homes studied in Nepal, suggesting more recent inputs of DDT in Nepal than in California.

Methyl parathion was the OP detected in the highest concentrations in these homes in Nepal. Interestingly, the amount of methyl parathion per m² in the dust from these Nepali homes was several orders of magnitude lower than the amount per m² in carpet dust from 10 homes of farmers or pesticide applicators in central New York State, USA [57]. In the ten rural New York homes, the mean and maximum levels of methyl parathion in carpet dust were 12,500 and 52,000 ng/m², respectively. These levels are much higher than the levels seen on the compacted red clay floors in both Nepali villages in this study, where the maximum level of methyl parathion was 532 ng/m². Carpet has been identified as a reservoir that can slow the removal of semivolatile organic compounds [58] such as methyl parathion, and as a source of increased particulate matter exposure when compared to bare floors [59]. In the rural Nepali homes in this study, the compacted red clay floors were cleaned with brooms, and floors were regularly re-mudded once per year during festival times. Pesticides were not routinely applied inside these homes for pest control, though they might have been used to address a pest infestation.

Previous research has identified the importance of limiting exposure to biomass smoke [42,60,61]. The levels of PAHs in house

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dust in these Nepali homes were higher than the levels identified by Naspinski and colleagues [45] in homes in Texas, USA and Sumgayit, Azerbaijan, but lower than the levels detected in homes in Shanxi Province, China. Naspinski et al. [45] found the median total PAH levels in floor dust samples from Texas, USA, Sumgayit, Azerbaijan and Shanxi Province, China were 1.34, 6.1, and 232 µg/m², respectively, while the median levels of the EPA B2 carcinogenic PAHs (cPAHs) were below the detection limit, 1.0 and 53 µg/m² in Texas, Azerbaijan and China, respectively [45]. The median tPAH value in both Keraghari and Kafaldanda was 14.7 μ g/m², above the median value in Azerbaijan but well below the median value in China seen by Naspinski and colleagues [45]. The cPAH levels of 2.06 and 1.89 μ g/m² in Keraghari and Kafaldanda, respectively were also higher than the levels seen by Naspinski et al. [45] in Texas and Azerbaijan but much lower than in China. The samples collected by Naspinski et al. [45] in Azerbaijan were obtained from homes near an industrial complex in an area that was an important petrochemical production location for the former Soviet Union, while the samples from China were collected in a location where coal is an important fuel. The homes in Nepal were located in rural mountain villages where wood burning stoves are used to cook and heat the home. Many of the homes in this study had a conduit connected to the stove to carry the wood smoke outside the home, thereby reducing indoor PAH levels.

There are relatively few studies on residential environmental exposures in Nepal. The studies of human exposures to pesticides in Nepal have typically focused on occupational exposures [16,62-64], or cases of pesticide poisoning and acute symptoms of pesticide exposure [65-69]. Studies of residential use of pesticides in Nepal have generally assessed methods for managing sand fly populations to control visceral leishmaniasis [70-74]. There have also been studies in Nepal of the health effects of residential exposure to indoor smoke [75-80], but these have not quantified individual PAHs or carcinogenic PAHs. To the best of our knowledge, this is the first study conducted in Nepal to quantify the levels of multiple pesticides and PAHs in house dust, which can serve as a source of chronic exposure to household residents, particularly young children. This study examined the levels of OCs, OPs and PAHs in household dust in two rural Nepali villages. The procedure that was used to collect the dust did not require specialized equipment and was easily portable, making it ideal for studies in remote locations where electricity is not available. This semi-quantitative method provided information about the mass of contaminant per unit area of floor. Although this does not provide contaminant concentration in dust, and therefore is not directly relevant to exposure calculations, it can be used to assess the major chemical contaminants, thereby facilitating the hazard identification component in the risk assessment process [45]. Both OCs and OPs were detected in the homes, with higher concentrations per unit area of OCs than OPs. The organochlorine pesticide DDT was detected, suggesting recent inputs of this compound. The Nepali farmers whose homes were sampled for this study had been previously trained in pesticide-free farming methods by a local non-governmental organization (NGO) and the US-based nonprofit organization Health Environmental Learning Program. Prior to this training, many farmers had been unaware of procedures for safe use, handling and storage of pesticides. The majority of farmers in the area continued to use pesticides following the training; however, this training and related discussions about the risks of pesticide exposure may have encouraged these farmers to use pesticides more carefully, resulting in lower pesticide levels in these homes compared to other Nepali village homes where the farmers had not received training. A study by Budhathoki et al. [81] of poisoning in 122 Nepali children found that 45.1% of these children were poisoned by organophosphates and 8.2% by organochlorines. Many farmers do not understand the health effects of pesticides or appropriate procedures for pesticide use and handling [16,17], and many rural villagers are illiterate and unable to read pesticide instructions. This suggests that continued training for Nepali villagers on the safe use, handling and storage of pesticides may be valuable.

The PAH data suggest that occupants of these Nepali homes continue to be exposed to PAHs, though not at the extremely high levels seen in a similar study by Naspinski et al. [45] in China. The samples in Nepal were collected in early summer, when the weather allows greater ventilation with outdoor air. It would be beneficial to further evaluate air quality in the homes with and without the improved stoves, during winter as well as summer, to evaluate the reduction in PAH exposure following use of the improved stoves. In addition, research on longterm homeowner satisfaction with these stoves would be useful, to assess whether the homeowners continue to use and maintain these stoves over a period of years, or whether additional refinements to the stove design would be appropriate. The randomized controlled trial of improved cookstoves currently being conducted by Klasen et al. [82] and the randomized community-based trials recently conducted by Tielsch et al. [83] in Nepal will provide valuable information on the effectiveness of improved cookstoves for reducing indoor contaminants and improving related health indicators.

Acknowledgement

Special thanks to the Health Environmental Learning Program for providing transportation to the sites, and coordination with local farmer groups. We thank Mr. Roshan Prajapati and the staff and farmers' groups of Help and Care, the Nepali sister non-governmental organization of Health Environmental Learning Program (H.E.L.P.). Thanks to Dr. Celia Valdez, Ms. Julliana Valdez, and Ms. Kristen Ackerman, who assisted in collecting the samples. We gratefully acknowledge the families whose homes were sampled in this study.

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Citation: Cizmas L, Ackerman L, Donnelly DA, Donnelly KC, McDonald TJ (2015) Measurement of Organophosphate Pesticides, Organochlorine Pesticides, and Polycyclic Aromatic Hydrocarbons in Household Dust from Two Rural Villages in Nepal. J Environ Anal Toxicol 5: 261. doi:10.4172/2161-0525.1000261