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Attic dust assessment near a wood treatment plant: Past air pollution and potential exposure

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ABSTRACT

The wood treatment process uses substances that generate hazardous compounds that may contaminate environmental compartments. In the present study, an area under influence of a deactivated wood treatment plant was investigated to evaluate past air pollution and to try to understand local air dispersion. Attic dust samples were collected from eight residences around the plant and from two residences outside this area, as reference samples. The presence of copper, chromium, arsenic, pentachlorophenol, sixteen priority polycyclic aromatic hydrocarbons and mutagenic activity using *Salmonella*/microsome assay was evaluated. The residences close to the entrance to the plant were the most affected, according to potentially toxic elements analysis. The PCP concentration was 0.49 mg/kg and the total PAHs content ranged from 0.40 to 13.31 μg/g with greater dispersion than potentially toxic elements. The highest mutagenesis values were 15,905 and 10,399 revertants/g of dust in the absence and presence of S9 mix (mammalian metabolic activation), respectively. Samples in which the total PAHs concentration was less than 2 μg/g no mutagenic effects were observed, including the residences in the reference area. The contribution of PAHs to mutagenesis was 10 percent, indicating that other compounds may contribute to the mutagenic effect. These results suggest that the population was or is potentially exposed to substances with strong effects on health.

1. Introduction

The wood treatment process uses substances that generate hazardous compounds that may contaminate soil, surface and groundwater and air. In the wood treatment processes the chemicals used contaminate the environmental compartment through vaporization, leakages and dripping which occur during the treatment and storage processes. The potential for environmental contamination and consequent exposure of organisms to these substances will depend on the preventive and control measures adopted. Over the years, the processes and substances used to treat wood changed because of the discovery of the toxicity of these chemicals. Coal tar creosote is a complex mixture that contains hundreds of compounds, among which aromatic hydrocarbons predominate, including polycyclic aromatic hydrocarbons (PAHs) (Gallego et al., 2008). Creosote oil is solubilized in petrol oil or burned oil for wood treatment. Creosote treated-wood also emits volatile organic compounds and PAHs after treatment in storage fields (Gallego et al., 2008). The International Agency for Research on Cancer (IARC) has determined that coal tar creosote is probably carcinogenic to humans (IARC, 2010). Pentachlorophenol (PCP) is a pesticide primarily used as wood preservative. PCP has not shown strong mutagenic activity but has demonstrated a synergic effect on mutagenicity (Gichner et al., 1998), and it is classified as possibly carcinogenic to humans (IARC, 2010). The sale and use of PCP and its salts for activities aimed at preserving wood have been forbidden in Brazil since 2007 (Brasil, 2006). After the

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use of creosote and PCP, chromated copper arsenate (CCA) was also used to preserve wood. Studies showed that soils from wood treatment plant are contaminated by arsenic due to the use of CCA (Bhattacharya et al., 2002) and that CCA treated wood leaches As, Cu and Cr (Hingston et al., 2001, Moghaddam and Mulligan, 2008) suggesting the risk of contamination of the water resources. Environmental liabilities and contamination related to wood preservation activities have been reported worldwide (da Costa et al., 2012, Dahlgren et al., 2007, Feng et al., 2011, Hensley et al., 2007, Pohren et al., 2012).

In this context, not only occupational, but also environmental exposure to the surrounding population may result in adverse health effects. Previous studies have showed adverse health effects, like mucous membrane irritation, skin, and neurological symptoms besides cancer on long-term residents near a wood treatment plant (Dahlgren et al., 2003a). Elevated levels of polychlorinated dioxins and furans in blood samples from nearby residents of wood treatment facilities were observed around United States of America (Dahlgren et al., 2003b, 2007; Feng et al., 2011; Hensley et al., 2007; Karouna-Renier et al., 2007). PCDD/F congener profiles in individuals that worked and lived in the surrounding neighborhoods of Escambia Treating Company reflected patterns commonly observed in persons exposed to PCP, also hypertension was correlated with PCDD/F levels (Karouna-Renier et al., 2007). These previously mentioned studies also identified specific contamination from the activities of the wood treatment plant in sediment, soil, house dust (Dahlgren et al., 2003b) and attic dust samples (Dahlgren et al., 2007, Hensley et al., 2007).

Particles present in the outside environment are important sources of indoor air quality (Maertens et al., 2004, Mercier et al., 2011). The influence from outdoors in the composition of house dust has been demonstrated (Caravanos et al., 2006; Davis and Gulson, 2005; Gonzalez et al., 2011; Lambert and Lane, 2004). Also, house dust seems to be a sensitive matrix to indicate heavy metals (Callan et al., 2012) and PAHs (Gevao et al., 2007) environmental contamination. The particles from the outside environment infiltrate the attics and indoors these contaminants accumulate and are preserved, since they are less subject to environmental degradation (such as sunlight, temperature changes and decomposition by microbial influence). Due to these properties and complexity, attic dust can be an interesting matrix that can contain a record of undisturbed archived deposited particles and provide an indirect measure of air pollution integrated over varying time periods, thereafter resident's potential for present and past exposure (Davis and Gulson, 2005; Gonzalez et al., 2011; Lioy et al., 2002).

Recently, studies have attempted to determine the substances, their sources and pathways to estimate the effects of indoor exposure on health (Mercier et al., 2011; WHO, 2010). The World Health Organization (WHO) published a guideline for some selected pollutants in indoors environment; however, these values are expressed as m^3 of air. In the present study, the environmental matrix evaluated was attic dust, which besides supplying information on indoor contaminants, also tries to evaluate the outdoor sources of contamination. Due to the complexity of this matrix, and the absence of reference values, the results obtained in this study were compared to the USEPA Regional Screening Levels for Chemical Contaminants at Superfund Sites for resident soil (USEPA, 2011) and to the guiding values for soil quality as to the presence of chemical substances established by the National Council of the Environment (Brasil, 2009). This current work investigated the presence of hazardous contaminants and mutagenic activity associated with attic dust. The objective was to evaluate past air pollution and try to understand local air dispersion of contaminants.

2. Material and methods

2.1. Study area

The study area is located on the left bank of Taquari River and 8.5 km south of the confluence with Jacuí River, in the northeast of the state of Rio Grande do Sul, Brazil (municipality of Triunfo) (Fig. 1). The wood treatment plant is located on the left bank of the river Taquari, the area is flat with small streams flowing towards the main river drainage. Local geology is characterized by sedimentary rocks, fine to very fine sandstones with intercalations of laminates (argilites and shales). In the surroundings of the plant there is a small residential neighborhood (with local shops and crops are cultivated for subsistence). On the right bank of the river, the predominant use is agricultural. The wood treatment plant operated from 1960 to 2005 and used a variety of processes and substances including creosote, pentachlorophenol (PCP) and chromated copper arsenate (CCA). Initial chemical analyses showed evidence of groundwater and soil contamination by PAHs, Cr, Cu, As, PCF, dioxins and furans (FEPAM, 2010a, 2010b).

2.2. Attic dust sampling

Intending to identify a possible dispersion gradient for the contaminants of interest, the surrounding region was considered the one at risk from the influence of the plant activities. The unavailability of local data referring to the micro-climate encouraged the adoption of this approach, since the dominant regional wind direction is southeast (24.9 percent), which would preferentially transport the pollutants into the Taquari River and into areas that are not heavily settled. On the other hand, the second preferential wind direction, which is northwest (13.7 percent), would influence the area around the plant (data measured by climatological station N° 83954 COPESUL/INMET). The residences were chosen by tracing two concentric circles with a radius of 300 and 600 m, respectively, originating from the main building of the treatment plant (prioritizing the location of the autoclave). The division of the circle into sections delimited different zones which could be sampled, where the residences were chosen randomly for evaluation. Before sampling was verified if the attic had been undisturbed for more than 10 years, no pesticides use in the residence, there was any openings to the outside and if there was accesses to the attic. Recently remodeled homes were not sampled. Within the 300 m radius, houses 4, 5 and 7 were evaluated, and in the 600 m radius, residence 1 (only evaluated for potentially toxic elements (PTEs), since there was not sufficient material to extract the organic compounds), 2, 3, 6 and 9 (Fig. 1). There were no residences in southwestern sections that explain why there is no sample number 8. Two more houses, 1750 m eastward from this area, were also sampled to be used as reference samples (residences numbers 10 and 11). Samples were collected in May and July 2008. Sampling was performed by carefully sweeping the dust accumulated in the attic using natural bristle brushes (Cizdziell et al., 1998; Cizdziel and Hodge, 2000; Silva et al., 1996). Samples were brought refrigerated to laboratory where the impurities were carefully removed and then were weighed and stored in prewashed glass containers protected from light and refrigerated until the preparation for extraction and chemical analysis. The amount of sampled attic dust ranged approximately between 2 g (house 1) and 20 g (house 2). The house characteristics like age, roof type, presence of wood burning stove and others were documented (Table 1).

In a second stage (October 2010) a composite sampling was performed of six nearby residences (located on the same street as samples 5 and 6) to evaluate the presence of PCP. A composite sample (pool samples from different houses) was necessary to collect a sufficient amount of dust to execute the PCP analyses. This sampling followed the same methods and requirements mentioned previously. The dust samples were preserved at 4 °C until analysis that did not take longer than 14 days from the date of sampling, and it was performed in the samples *in natura*. PCP extraction was conducted with methylene chloride, following the method SMWW6410B (APHA, 2005). PCP was determined according to procedures described in the USEPA SW846/8270C method (USEPA, 1996), using gas chromatography coupled to mass spectrometry (GC/MS).

2.3. Potentially toxic elements

The samples were digested for arsenic, copper and chromium analysis according to the USEPA SW846 Method 3050B (USEPA, 1995); inductively coupled plasma mass spectrometry (ICP-MS) was used for quantification. Pearson correlations were used to investigate the relationship between analysed PTEs.

2.4. Organic extraction

The attic dust samples were extracted by sonication (THORNTON—power 1800 W) using dichloromethane (DCM, CASRN. 75-09-2) at a ratio of 1(dust):2 (solvent). After filtration with a Millipore membrane, 0.5 μm porosity, the volume was concentrated in a rotary evaporator at 40 °C to obtain the final extract. The extract was stored in a graduated tube, from which a 1 mL aliquot was