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## Quantitative ecological risk assessment of inhabitants exposed to polycyclic aromatic hydrocarbons in terrestrial soils of King George Island, Antarctica

S. Pongpiachan <sup>a, \*</sup>, M. Hattayanone <sup>b</sup>, O. Pinyakong <sup>c</sup>, V. Viyakarn <sup>d</sup>, S.A. Chavanich <sup>d</sup>, C. Bo <sup>e</sup>, C. Khumsup <sup>f</sup>, I. Kittikoon <sup>f</sup>, P. Hirunyatrakul <sup>f</sup>

<sup>a</sup> School of Social & Environmental Development, National Institute of Development Administration (NIDA), Bangkok, Thailand

<sup>b</sup> Faculty of Environmental Management, Prince of Songkla University, Hat-Yai, Songkhla, Thailand

<sup>c</sup> Bioremediation Research Unit, Department of Microbiology, Faculty of Science, Chulalongkorn University, Bangkok, Thailand

<sup>d</sup> Reef Biology Research Group, Department of Marine Science, Faculty of Science, Chulalongkorn University, Bangkok, Thailand

<sup>e</sup> Polar Biological Science Division, Polar Research Institute of China, Shanghai, China

<sup>f</sup> Bara Scientific Co., Ltd., Bangkok, Thailand

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#### ABSTRACT

This study aims to conduct a quantitative ecological risk assessment of human exposure to polycyclic aromatic hydrocarbons (PAHs) in terrestrial soils of King George Island, Antarctica. Generally, the average PAH concentrations detected in King George Terrestrial Soils (KGS) were appreciably lower than those of World Marine Sediments (WMS) and World Terrestrial Soils (WTS), highlighting the fact that Antarctica is one of the most pristine continents in the world. The total concentrations of twelve probably carcinogenic PAHs (SPAHs: a sum of Phe, An, Fluo, Pyr, B[a]A, Chry, B[b]F, B[k]F, B[a]P, Ind, D[a,h]A and B[g,h,i] P) were 3.21  $\pm$  1.62 ng g<sup>-1</sup>, 5749  $\pm$  4576 ng g<sup>-1</sup>, and 257,496  $\pm$  291,268 ng g<sup>-1</sup>, for KGS, WMS and WTS, respectively. In spite of the fact that KGS has extremely low  $\Sigma$ PAHs in comparison with others, the percentage contribution of Phe is exceedingly high with the value of 50%. By assuming that incidental ingestion and dermal contact are two major exposure pathways responsible for the adverse human health effects, the cancer and non-cancer risks from environmental exposure to PAHs were carefully evaluated based on the "Role of the Baseline Risk Assessment in Superfund Remedy Selection Decisions" memorandum provided by US-EPA. The logarithms of cancer risk levels of PAH contents in KGS varied from -11.1 to -7.18 with an average of  $-7.96 \pm 7.73$ , which is 1790 times and 80,176 times lower than that of WMS and WTS, respectively. All cancer risk levels of PAH concentrations observed in KGS are significantly (p < 0.001) lower than those of WMS and WTS. Despite the Comandante Ferraz Antarctic Station fire occurred in February 25th, 2012, both the cancer and non-cancer risks of environmental exposure to PAHs were found in "acceptable level".

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#### 1. Introductions

Polycyclic aromatic hydrocarbons (PAHs), usually acknowledged as a group of persistent organic pollutants (POPs), have been comprehensively investigated in the past decades because these congeners have a profound association with a wide range of adverse health effects and other respiratory diseases (Bhargava et al., 2004; Chalbot et al., 2012; Claxton and Woodall Jr., 2007).

\* Corresponding author. *E-mail address:* pongpiajun@gmail.com (S. Pongpiachan).

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PAHs are widely detected in various types of environmental compartments including marine organisms (Pongpiachan et al., 2013a,b; Pongpiachan, 2013a,b; Sette et al., 2013; Yoshimine et al., 2012). It is well known that PAHs can be generated from both anthropogenic and natural sources (Lu et al., 2012; Okuda et al., 2002; Slezakova et al., 2011; Yang et al., 2002). According to recent studies, particulate PAHs are harmful to human health due to its responsibilities for cancer, endocrine disruption, and reproductive and developmental effects (Liao et al., 2011; Hoyer, 2001; Matsui, 2008; Wickramasinghe et al., 2012). Although the majority of PAHs generally have a low degree of acute toxicity to humans, numerous investigations have demonstrated non-carcinogenic

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effects of PAHs involve primarily the pulmonary, gastrointestinal, renal, and dermatologic systems after chronic exposure (Gupta et al., 1991). It is a strenuous task to evaluate observed health effects in epidemiological studies to certain PAH congener because most exposures are related to PAH mixtures, which include both carcinogenic and non-carcinogenic PAHs. A recent study found that both phenanthrene (non-carcinogenic) and benzo[b]fluoranthene (carcinogenic to experimental animals) induced hepatic histopathological changes that highlight metabolic failure and inflammation, particularly in animals exposed to mixtures (Martins et al., 2015). It is also crucial to underline that two or more PAH congeners within a mixture may compete for receptors or active sites of metabolizing enzymes and chaperones, producing synergistic, antagonistic or additive impacts that may responsible either in cocarcinogenic or chemopreventive effects (Jarvis et al., 2014).

As a consequence of distress over its potential hazard to public health, numerous studies were conducted to investigate the impact of meteorological parameters on its temporal variation and spatial distribution (Akyüz and Çabuk, 2009; Amodio et al., 2009). Further attempts on clarification of factors governing diurnal variation of PAHs have also been carried out in different countries (Chetwittayachan et al., 2002; Gu et al., 2010; Ringuet et al., 2012; Zhang et al., 2012).

Regardless of numerous studies on variations of PAHs in soils and sediments around the world, the knowledge of the magnitude of their contamination in Antarctic soil, which is still considered as one of the most pristine areas of the world, is strictly limited. During the past decades, there are numerous studies have been attempted to elucidate the degree of contamination as well as its emission source strength of PAHs in Antarctic soils (Aislabie et al., 1999; Curtosi et al., 2007; Prus et al., 2015). In order to obtain some references for global migration of PAHs, snow samples were collected in Fildes Peninsula in the Southwestern end of King George Island in the South Shetland Islands of Antarctica. Principal component analysis revealed that Naphthalene, Fluorene and Phenanthrene were three main factors of PAHs detected in snow samples accounted for 61%, 22%, and 10%, respectively (Na et al., 2011). Similar patterns were also observed in marine sediments sampled from Prydz Bay, East Antarctica with two- and three-ring PAHs as the most abundant compounds detected at the majority of the observatory sites of Prydz Bay (Xue et al., 2016). Further source apportionment analysis in sediment cores of Admiralty Bay, Antarctica were conducted by using PAHs and carbonaceous particles (SCPs) as two markers used for assessing the local input of anthropogenic materials as a consequence of the impact of human activities in both sub-Antarctic region and South America. By interpreting the diagnostic binary ratios of PAHs, fossil fuels/petroleum, biomass burning and sewage contribution were generally considered as three main sources of PAHs in this region.

In spite of countless studies focusing on spatial and temporal distributions of PAHs in Antarctic environment, there are no compelling studies and their outcomes demonstrating the magnitude of exposure and ecological risk of PAHs in King George Island. Overall, the objectives of this study are i) to analyse PAH concentrations in terrestrial soils of King George Island, ii) to use a variety of comparable ecological risk assessment models to obtain more advantageous insights of the potential carcinogenic risk in the Sub-Antarctic Region for the first time. It is crucial to underline that the main purpose of this study is to illustrate the general principle of applying PAHs as chemical indicators for cancer risk assessment in King George Island. Neither source apportionment nor the assessment of temporal variation and spatial distribution of PAHs is the prominent goal of this investigation.

#### 2. Materials and methods

#### 2.1. Collections of terrestrial soils

All terrestrial soils (n = 21) were collected from 8th January 2014 to 23rd January 2014 at Southwestern part of King George Island with the depth of 0–10 cm (see Table 1 and Fig. 1A). King George Island is the largest of the South Shetland Islands coordinates at  $62^{\circ}02'S$  58°21'W with the area of 1150 km<sup>2</sup> (i.e. 440 sq mi). King George Island possesses three main bays, namely Maxwell Bay, Admiralty Bay, and King George Bay. Admiralty Bay has three fjords, and is generally protected as an Antarctic Specially Managed Area under the Protocol on Environmental Protection to the Antarctic Treaty. The majority of samples were collected around the Great Wall Station, which lies on the Fildes Peninsula on King George Island, and is approximately 2.5 km from the Chilean Frei Montalva Station. Terrestrial soil samples were wrapped in clean aluminum foil, placed in a glass bottle, and kept frozen at -20 °C in order to avoid sample degradation caused by heat, ozone, NO<sub>2</sub>, and ultraviolet (UV) during sample transport. They were freeze-dried prior to being grounded and sieved to homogenize the samples, and then kept in the refrigerator at -4 °C until analysis. Procedural precautions (e.g. special precautions for trace contaminant soil sampling), sample homogenization, dressing soil surfaces, sampling methodology for low concentrations (<200 ng  $g^{-1}$ ), and quality control/quality assurance (QA/QC) were clearly explained in soil sampling US-EPA Method 5035 and will not be discussed here (US-EPA, 2002).

#### 2.2. PAHs analysis

All organic solvents (i.e. DCM and Hexane) are HPLC grade, purchased from Fisher Scientific. A cocktail of 15 PAHs Norwegian Standard (NS 9815: S-4008-100-T) (phenanthrene (Phe), anthracene (An), fluoranthene (Fluo), pyrene (Pyr), 11h-benzo[a]fluorene (11H-B[a]F), 11h-benzo[b]fluorene (11H-B[b]F), benz[a]anthracene (B[a]A), chrysene (Chry), benzo[b]fluoranthene (B[b]F), benzo[k] fluoranthene (B[k]F), benzo[a]pyrene (B[a]P), benzo[e]pyrene (B[e] P), indeno[1,2,3-cd]pyrene (Ind), dibenz[a,h]anthracene (D[a,h]A), benzo[g,h,i]perylene (B[g,h,i]P); each 100  $\mu$ g mL<sup>-1</sup> in toluene: unit:  $1 \times 1$  mL) and a mix of recovery Internal Standard PAHs (d<sub>12</sub>-perylene ( $d_{12}$ -Per),  $d_{10}$ -fluorene ( $d_{10}$ -Fl); each 100 µg mL<sup>-1</sup> in xylene: unit:  $1 \times 1$  mL) were supplied by Chiron AS (Stiklestadveine 1, N-7041 Trondheim, Norway). A 30 g free-dried terrestrial soil sample was transferred to pre-cleaned Thimber. The chemical extraction of PAHs was performed by using 250 ml of Soxhlet extractors, spiked with a known quantity of internal standard ( $d_{10}$ -Fl: Phe, An, Fluo, Pyr, B[a]A, Chry; *d*<sub>12</sub>-Per: B[b]F, B[k]F, B[a]P, Ind, D[a,h]A, B[g,h,i]P) and extracted with DCM containing 1 g of copper powder for 8 h. In order to avoid any method interferences, which may be caused by contaminants in glassware, reagents, solvents, and other experimental devices, the laboratory blanks were routinely analyzed to ensure that all mentioned materials were to be free from interferences under the conditions of the analysis.

The fractionation/clean-up technique, blowing down process (e.g. a combination of rotary evaporation with a gentle nitrogen stream) was strictly performed in accordance with the standard protocol applying the difference in solvent polarity (Gogou et al., 1996, 1998). The reduced extract was subsequently diluted in 10 ml of *n*-hexane before being transferred to the top of a column of disposable silica gel, which was activated at 150 °C for 3 h. Two different compound groups (i.e. light molecular weight PAHs and heavy molecular weight PAHs) were classified by eluting 15 ml *n*-hexane and 15 ml toluene-*n*-hexane (5.6:9.4). The gas chromatography (GC) temperature programming coupled with the

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