Worldwide distribution of persistent organic pollutants in air, including results of air monitoring by passive air sampling in five continents

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This article provides an overview of concentrations of persistent organic pollutants (POPs) in ambient air on a global scale, including recent measurements and an extensive compilation of literature data. In this study, passive air samplers (PASs) were successfully employed to assess concentrations of POPs in ambient air from Africa, Latin America, the Caribbean, and the Pacific Islands. The project aimed to extend the knowledge on environmental contamination by POPs in these regions, where the currently available data are still limited to a few monitoring studies.

The ambient air concentrations of PCB in Africa were relatively high when compared to other regions. Waste, in particular electronic waste, exported to Africa from industrialized countries may be a possible source of PCB in Africa, where PCB have never been extensively used or produced. For DDTs, the wide range of concentrations and particularly high levels in some countries of Africa and the Pacific Islands reflect the use of DDT for malaria control in these regions. For PCDD/PCDF, concentrations in Africa and Latin America are similar to or even higher than in Europe, probably due to unfavorable combustion practices of chlorine-containing materials.

The data support the needs for further monitoring in developing countries and countries with economies in transition, and action to reduce environmental contamination by, and human exposure to, hazardous chemicals.

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1. Introduction

Persistent organic pollutants (POPs) represent a group of organic chemicals of particular concern because they are resistant to degradation in the environment (persistent), accumulate in human and animal tissue, biomagnify along food chains, and are associated with chronic (eco)toxicological effects [1]. In 2004, the Stockholm Convention on POPs came into force and aims to protect human health and the environment from negative effects of POPs by reducing or eliminating releases of POPs to the environment [2]. In 2009, the initial list of 12 POPs was extended by nine additional chemicals.

POPs have become ubiquitous contaminants found in not only the environment

and biota of their region of initial use, but also remote areas far away from their primary emission sources [3-7]. POPs used as pesticides are directly emitted into the atmosphere by spraying application. A considerable amount of POPs is also released to air by volatilization from POP-containing applications (open sources) or in fumes from combustion processes (unintentionally formed POPs), and the atmosphere is a key medium for environmental long-range transport of POPs, next to the slower transport by rivers, ocean currents, and sea ice. Ambient air monitoring of POPs is therefore particularly relevant for investigating environmental levels and trends of POPs. Air has also been recognized as a key environmental medium in the Global Monitoring Plan of the Stockholm Convention, which is intended to support evaluation of the effectiveness of the Convention. In addition, several monitoring studies on POPs in the atmosphere on a local, regional, and global scale have been conducted [8-17].

Still, ambient air monitoring of POPs is a challenging task. On the one hand, concentrations of atmospheric contaminants are variable spatially and, in particular, temporarily. On the other hand, the usually low concentrations of POPs in air inevitably require elaborate sampling techniques followed by high-performance, resource-intensive analytical methods, so there are only a few large monitoring studies about POPs in air and the global coverage is still unequal. Whereas fairly extended data sets about atmospheric occurrence of POPs are available for the Northern hemisphere [8,11,12,18–22]; Africa [14,16], Latin America [16,17], and the Pacific region [16] have scarcely been covered.

Herein, we present a recently conducted study about POPs in air in (i) West, East, and South Africa (ii) Latin America and the Caribbean, and (iii) Pacific Islands. We focus on polychlorinated biphenyls (PCBs), dichlorodiphenyl trichloroethane (DDT) and its main transformation products (together referred to as DDTs), as well as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDDs/PCDFs), as representative of industrial POP chemicals, POP pesticides, and unintentionally formed POPs, respectively.

DDTs are of particular interest with respect to the current exemption under the Stockholm Convention, permitting the use of DDT for malaria control in affected regions. Extensive results for 12 POPs measured in ambient air samples, including individual PCB, DDT, and PCDD/PCDF congeners, are presented in a companion document [23]. First, we introduce the applied method based on passive air samplers (PASs). Second, we present and discuss the collected data set in a wider context, including comparison to existing monitoring studies, for which the raw measurement data are available and which rely on the same sampling technique.

2. Determination of levels of POPs in ambient air

To sample air volumes that are large enough to overcome analytical detection limits, PASs have been developed as simple, cost-effective alternatives to active air samplers [24]. The core part of a PAS consists of an adsorbent that is exposed to a passive air flow. Airborne chemicals are taken up by molecular diffusion, resulting in an accumulation of chemicals in the adsorbent. In the initial so-called "linear uptake phase", the amount of airborne chemical sequestered by the adsorbent increases linearly with time and – under the assumptions of a constant volumetric sampling rate of air and a constant chemical concentration in air – is proportional to the sampled air volume (Fig. 1a). In the course of exposure time, the uptake rate decreases until equilibrium between the adsorbent and ambient air is reached. Thus, when PASs are deployed for a period not longer than the linear uptake phase, the amount of chemical sequestered in the adsorbent makes it possible to determine the concentration of the pollutant in ambient air. based on the sampled volume of air.

Polyurethane foam (PUF) disks proved to be an adequate adsorbent in PASs [24]. PASs used in the present project are identical to the devices used in several previous studies (Fig. 1b) [14,16,24] and consist of a PUF disk protected from dry and wet deposition by a stainless-steel casing, which has also a wind-shielding function to reduce fluctuations of the air flow [25,26].

Low acquisition costs, limited maintenance demand, simple handling, and no need for power supply are advantages offered by PASs, especially in countries with limited financial resources or in remote areas. However, the accurate determination of ambient-air concentrations represents a difficulty when PASs are used. The analysis of pollutants in a PUF provides merely their amount sequestered in the PUF during the deployment period. Only after dividing the amount of chemicals (e.g., expressed in pg) by the corresponding air-sampling volume (e.g., expressed in m³) is the concentration of POPs in air obtained (e.g., expressed in pg/m³). The determination of the sampling volume requires extensive calibration exercises. Uptake rates of POPs (e.g., PCB) can vary from 2–8 m³/day [24,27] and typically lie between $3-4 \text{ m}^3/\text{day}$ [14,16,24,27]. The deployment period has to be long enough to obtain the required air-sampling volume, but it should not be too long to avoid the PUF reaching equilibrium with ambient air (Fig. 1a). Typical sampling periods lie within a few weeks, resulting in sampled air volumes of $\sim 100-500 \text{ m}^3$.

For POPs, another limitation needs to be considered. Because of their low vapor pressure, POPs in air partition between the pure gas phase and the particulate phase (aerosols). Uptake rates determined so far apply to only the gas phase. The entrapment of aerosols and associated

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