



Low concentrations of persistent organic pollutants (POPs) in air at Cape Verde



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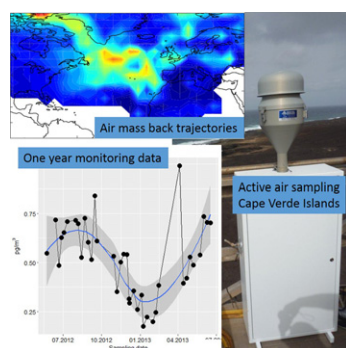
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HIGHLIGHTS

- One year active air sampling of POPs was performed at Cape Verde Islands.
- Concentrations of several POPs were generally low.
- Air mass back trajectories demonstrated Atlantic Ocean as frequent source regions.
- Volatilization from the ocean was a likely source to several POPs.

GRAPHICAL ABSTRACT



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ABSTRACT

Ambient air is a core medium for monitoring of persistent organic pollutants (POPs) under the Stockholm Convention and is used in studies of global transports of POPs and their atmospheric sources and source regions. Still, data based on active air sampling remain scarce in many regions. The primary objectives of this study were to (i) monitor concentrations of selected POPs in air outside West Africa, and (ii) to evaluate potential atmospheric processes and source regions affecting measured concentrations. For this purpose, an active high-volume air sampler was installed on the Cape Verde Atmospheric Observatory at Cape Verde outside the coast of West Africa. Sampling commenced in May 2012 and 43 samples (24 h sampling) were collected until June 2013. The samples were analyzed for selected polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), hexachlorobenzene (HCB) and chlordanes. The concentrations of these POPs at Cape Verde were generally low and comparable to remote sites in the Arctic for several compounds. Seasonal trends varied between compounds and concentrations exhibited strong temperature dependence for chlordanes. Our results indicate net volatilization from the Atlantic Ocean north of Cape Verde as sources of these POPs. Air mass back trajectories demonstrated that air masses measured at Cape Verde were generally transported from the Atlantic Ocean or the North African continent. Overall, the low concentrations in air at Cape Verde were likely explained by absence of

Abbreviations: AAS, active air sampling; CVAO, Cape Verde Atmospheric Observatory; ECMWF, European Centre for Medium-Range Weather Forecasts; FLEXPART-f, forecasted FLEXPART emission sensitivities; FLEXPART-r, retrospective FLEXPART emission sensitivities; GC, gas chromatograph; GFS, Global Forecast System; HCB, hexachlorobenzene; MDL, method detection limit; MS, mass spectrometer; NCEP, National Center for Environmental Prediction; PBDEs, polybrominated diphenyl ethers; PCBs, polychlorinated biphenyls; POPs, persistent organic pollutants; PUF, polyurethane foam; QA-QC, quality assurance-quality control.

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major emissions in areas from which the air masses originated combined with depletion during long-range atmospheric transport due to enhanced degradation under tropical conditions (high temperatures and concentrations of hydroxyl radicals).

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1. Introduction

Many persistent organic pollutants (POPs) have the potential for atmospheric long-range transport and are ubiquitous in the global environment (Poza et al., 2006; Wania and Mackay, 1996). Over the last decades, several regional monitoring networks have been established to assess the occurrence and trends of POPs in air in support of relevant regulatory programs and Conventions (Hillery et al., 1998; Hung et al., 2010; Kong et al., 2014; Tørseth et al., 2012). Historically, these early monitoring networks have been limited to active air sampling (AAS), but more recent advancements in passive air sampling have facilitated increased spatial coverage (Jaward et al., 2004a; Jaward et al., 2005; Shen et al., 2006) and even temporal trends (Schuster et al., 2011). Still, long-term monitoring stations and/or networks targeting POPs in air based on AAS remain scarce and are mainly limited to industrialized regions in the northern hemisphere (Tørseth et al., 2012) and polar regions (Hung et al., 2010). Comparably less data derived on the basis of AAS are available from developing regions and tropical areas although there are notable exceptions in ship-based cruises (Gioia et al., 2008a, 2008b; Jaward et al., 2004b; Lohmann et al., 2012; Lohmann et al., 2013a; Lohmann et al., 2013b; Xie et al., 2011). Among such studies, there is some previous data available for selected compounds measured at the Cape Verde islands off the West African coast (Garrison et al., 2014; Gioia et al., 2011). Also, there are examples of passive air sampling campaigns in tropical areas (Gioia et al., 2011; Klánová et al., 2009; Lammel et al., 2013).

The current study was performed at Cape Verde islands and aimed to (i) monitor concentrations of selected POPs in air outside West Africa based on active sampling during the course of a year, and (ii) to evaluate atmospheric processes and source regions affecting the measured concentrations.

2. Materials and methods

2.1. Sampling campaign

Sampling was carried out at the Cape Verde Atmospheric Observatory (CVAO; Fig. 1; 16° 51' 49 N, 24° 52' 02 W) which is a World Meteorological Organization-Global Atmospheric Watch station that has served to provide long-term ground- and ocean-based data since 2006. The CVAO is situated on an island >800 km off the coast of Western Africa in the northern trade winds (heading southward from the Canary Islands and North African continent) region of the tropical eastern Atlantic Ocean (Carpenter et al., 2010).

Between 23 May 2012 and 17 June 2013, 43 air samples were collected with median sampling frequency of 8 days (ranging 1–49 days; see details in Table S1). Twenty-four hours integrated air samples were collected with an average sampling volume of 634 m³ (0.44 m³ min⁻¹). The sampling was performed using an active high-volume air sampler (DIGITEL Elektronik AG, Hegnau, Switzerland) installed on the roof of the CVAO station (altitude 3 m, sampler installed 30 m above ground). Further, the sampler collects particle and gas phases using a glass microfiber filter and two polyurethane foam (PUF) disks, respectively. Glass microfiber filters (diameter 1.5 cm, GF/C CAT No. 1822–150 from Whatman GF/C, GE Healthcare, Buckinghamshire, UK) were pretreated at 450 °C for 8 h and wrapped in alumina foil. PUF disks (diameter 4.5 cm, thickness 7.5 cm; from Sunde Søm & Skumplast A/S, Gan, Norway) were pre-cleaned using Soxhlet

extraction with toluene for 24 h, acetone for 8 h and finally with toluene for an additional 8 h. After cleaning, the PUF disks were dried in desiccators at 40 °C under vacuum until they were completely dry. Filter holders and glass sampling chambers were pre-cleaned by soaking in a soap solution overnight and rinsed in tap water, followed by acetone and *n*-hexane.

After sampling was completed, the exposed PUF samples were sealed in a gas tight container and glass fiber filters were wrapped in alumina foil and sealed. The samples were stored in a freezer facility at CVAO and sent to the laboratories of NILU at Kjeller, Norway for analyses.

2.2. Sample clean-up and analyses

The analytical procedures for PCBs, PBDEs and pesticides were similar to those described by Eckhardt et al. (2009) for PCBs. Prior to extraction, the sample material (filters and PUFs combined) were spiked with internal standards (isotopically labeled analogs of selected analytes; listed in Table S2) before they were Soxhlet extracted for 8–10 h in approximately 250 mL of *n*-hexane/10% diethyl ether. Extracts were concentrated on a TurboVap 500 System (Zymark, Hopkinton, MA, USA) to about 50 mL, and transferred to a graded cylinder. For the majority of samples, 10% (volume) of the extracts were transferred to 10 mL stained vials with screw-caps and stored in a fridge for potential future analyses. The remaining extracts were further concentrated to about 0.5 mL, transferred to a centrifuge tube, adjusted to 2 mL with *n*-hexane, and treated with 2 mL concentrated sulfuric acid. During the acid treatment the clear extract turns to dark yellow, and the treatment was repeated until no more color change was visible (usually 2–3 times). The extract was reduced to 0.5 mL in an evaporation glass and further cleaned up with fractionation with a silica column (15 mm in diameter and 200 mm in length). The column consisted of 4 g of activated silica (Silica gel 60 Merck, pretreated for 8 h at 550 °C) topped with 1 cm anhydrous sodium sulfate (pretreated for 12 h at 600 °C). The column was prewashed with 30 mL *n*-hexane before the samples were eluted with 30 mL *n*-hexane/10% diethyl ether. Extracts were reduced to 0.5 mL by evaporation on the Turbovap and solvent exchanged into iso-octane and transferred to a small vial with a screw-cap. Prior to analyses all extracts were further reduced by a gentle stream of nitrogen to approximately 100 µL and 1,2,3,4-tetrachloronaphthalene was added as recovery standard.

Analyses of the i) PCB congeners and HCB were performed using an Agilent 7890A gas chromatograph (GC) coupled to a Waters high resolution mass spectrometer (MS) in electron impact mode (Halse et al., 2011); ii) PBDE congeners were performed with an Agilent 6890 N GC and a low resolution Waters AutoSpec MS in electron impact mode; and, iii) pesticides (*trans* chlordane, *cis* chlordane, *oxy*-chlordane, heptachlor, *trans* nonachlor, *cis* nonachlor and mirex) were performed by a Agilent 6890 N GC coupled to a low resolution Waters Autospec MS in electron capture negative ion mode (Halse et al., 2015). Details of the instrumental conditions (e.g. column and temperature program) and all compounds analyzed are presented in Table S3.

The quantification was performed according to the isotope-dilution method, an internal standard method, which is applied in all modern standard methods dealing with analysis of POPs as for example the US-EPA method 1625B for semivolatile organic compounds (US EPA Method 1625, 1984). These methods inherently correct the analyte

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