

# The investigation of atmospheric deposition distribution of organochlorine pesticides (OCPs) in Turkey



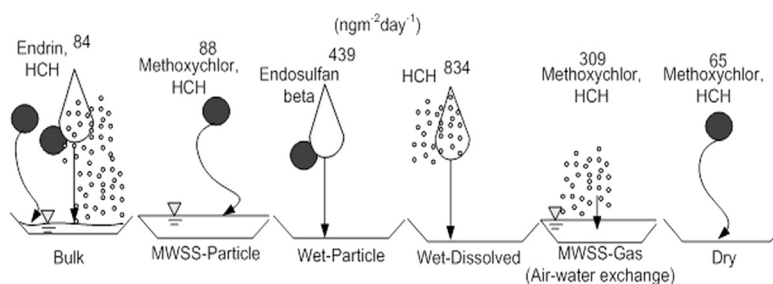
S. Siddık Cindoruk\*, Yücel Tasdemir

Uludag University, Faculty of Engineering and Architecture, Environmental Engineering Department, Bursa, Turkey

## HIGHLIGHTS

- Wet, dry, bulk deposition and absorption fluxes were determined for OCP compounds.
- Wet deposition accounted for the 69% of the total deposition fluxes.
- Different OCP compounds were dominant on different deposition mechanisms.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Atmospheric deposition is a significant pollution source leading to contamination of remote and clean sites, surface waters and soils. Since persistent organic pollutants (POPs) stay in atmosphere without any degradation, they can be transported and deposited to clean surfaces. Organochlorine pesticides are an important group of POPs which have toxic and harmful effects to living organisms and environment. Therefore, atmospheric deposition levels and characteristics are of importance to determine the pollution quantity of water and soil surfaces in terms of POPs. This study reports the distribution quantities of atmospheric deposition including bulk, dry, wet and air–water exchange of particle and gas phase OCPs as a result of 1-year sampling campaign. Atmospheric deposition distribution showed that the main mechanism for OCPs deposition is wet processes with percentage of 69 of total deposition. OCP compounds' deposition varied according to atmospheric concentration and deposition mechanism. HCH compounds were dominant pesticide species for all deposition mechanisms. HCH deposition constituted the 65% of  $\Sigma_{10}$ OCPs.

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## 1. Introduction

Organochlorine pesticides (OCPs), a class of semivolatile organic compounds (SVOCs), were widely used until 1980s and most of them were banned/restricted in Turkey as in other countries, however some OCPs (Endosulfan isomers and Methoxychlor) have been just banned in Turkey. OCPs are stable compounds with high vapor pressure, low water solubility and high lipid solubility which

undergo limited decomposition/degradation ((ATSDR), 2005; Bozlaker, 2008; Cincinelli et al., 2009; Sun et al., 2006). Due to persistence, toxic and bio-accumulative properties of OCPs, research studies focused on investigation of levels in different environmental compartments (water, air, soil, sediment, and biota).

The OCPs in atmosphere can partition between particle and gas phases, transport to remote areas and deposit to water surfaces, soils and vegetation with dry/wet deposition and air–water exchange processes causing contamination of clean environment (Hillery et al., 1998; Jurado et al., 2005; Odabasi et al., 2008). Wet deposition including rain, snow and dew precipitation and dry deposition including particle phase settling and gas phase air–

\* Corresponding author. Tel.: +90 224 294 2114; fax: +90 224 294 26 35.  
E-mail address: [cindoruk@uludag.edu.tr](mailto:cindoruk@uludag.edu.tr) (S.S. Cindoruk).

water exchange are important pathways for long range transport and removal for SVOCs (Ruzickova et al., 2008; Sun et al., 2006; Wang et al., 2008).

Bulk, wet, dry and air–water exchange deposition fluxes have been determined for SVOCs at different sites of world because of playing a key role on transport and fate. Polychlorinated biphenyl (PCB) and polycyclic aromatic hydrocarbon (PAH) bulk, dry and air–water exchange fluxes were determined using modified water surface sampler (MWSS) in our previous studies for Bursa/Turkey (Cindoruk et al., 2008; Cindoruk and Tasdemir, 2007a, b; Esen et al., 2008; Tasdemir and Esen, 2008; Vardar et al., 2008). The air–water exchange fluxes determined by the MWSS can also be assumed as diffusive absorption because the absorbed OCPs in the gas phase were collected without giving any time for re-evaporation to the atmosphere. Therefore, the MWSS can be defined only diffusive absorption. Determination of atmospheric deposition pattern of SVOCs is of importance in respect of two main aspects: (i) to understand the deposition mechanism of SVOCs at dry/wet air conditions, (ii) to estimate contribution of atmospheric deposition to water/soil pollution and vegetation contamination. Previous studies showed that air–water exchange is effective on wet deposition of easily diffusible SVOCs while dry deposition is important for that one's having an affinity of being on particle phase (Gigliotti et al., 2002; Jurado et al., 2004).

In this study, OCPs deposition fluxes were obtained using different sampling devices. Dry deposition fluxes were determined by modified water surface sampler (MWSS) and wet/dry deposition sampler (WDDS), which both of them were designed by our group. Wet deposition fluxes were obtained using WDDS while bulk deposition fluxes including wet and dry were determined by only a stainless steel collector. By using these samplers, it was tried to identify the deposition distribution of OCPs affected by meteorological conditions. In addition, atmospheric OCP concentrations were determined to calculate washout ratios, deposition velocities and mass transfer coefficients (MTC).

## 2. Materials and method

### 2.1. Sampling

Sample collection was performed in a residential/urban site of Bursa (Yavuz Selim, YS) (40°11'48.40"N–29°05'46.80"E) (Fig. 1). YS sampling area has mostly small indiscriminate industrial businesses and squatter's houses. The sampler was placed on the roof of a three-story building. Deposition samples and air concentration samples were all collected for 1-year period between June 2008 and June 2009. A high volume air sampler (HVAS) (Thermo Andersen GPS 11 Model, ABD) was used to collect air samples in the particulate and gas phases separately. A glass fiber filter (GFF) with a diameter of 10.2 cm and two pieces of a polyurethane foam (PUF) cartridge with a length of 5 cm and a diameter of 5.5 cm were used for particulate and gas phase OCPs (Kim and Masunaga, 2005; Yeo et al., 2003b). Four different collectors were used for deposition samples. The two of them are in WDDS which can collect rain/snow samples with beginning of precipitation events and can collect dry deposition samples when there was no fall. The third sampler (MWSS), which was used in our previous studies (Cindoruk, 2011; Cindoruk et al., 2008; Cindoruk and Tasdemir, 2007a,b; Esen et al., 2008), is employed for only dry samples determining maximum deposition fluxes. The fourth one is bulk deposition sampler which is widely used by researchers. The WDDS and MWSS are given in Fig. 1. The detail of MWSS is given in Cindoruk and Tasdemir (Cindoruk and Tasdemir, 2007b). The bulk deposition sampler (BDS) is a stainless steel collection pot with 60.5 cm diameter and 19 cm height which gathers both wet and dry deposition in same

sample. In order to prevent turbulence, a leading edge with 20 cm width was placed around the BDS like in MWSS. The BDS is always open to atmospheric deposition during sampling period. The WDDS was designed by our group with some differences from other samplers used in wet/dry or wet-only research studies. Rain or snow fallen into the collection plate is instantly conducted to the filter and resin units without retention. The moving cover of the plate is controlled by an electronic rain sensor. In general, samples were taken for 15 days period.

### 2.2. Analysis

Wet deposition samples obtained by WDDS and MWSS were both extracted with same method. GFF filters and XAD-2 resin extracted with 150 mL 1/1 Acetone (ACE)/Hexane (HEX) mixture using an ultrasonic bath repeating three times. HVAS filters were extracted twice with 25 mL 1/4 dichloromethane/petroleum ether (DCM/PE) mixture for 30 min in the ultrasonic bath while PUF samples were extracted in Soxhlet extractor for 24 h with 1/4 DCM/PE mixture. Dry deposition samples were collected by washing the reservoirs with 1/1 ACE/HEX mixture and they were wiped with some tissues soaked in ACE/HEX in order to remove any deposited particles onto the surface. The samples collected with BDS were extracted due to water content. If there was water in sampler, the water was filtered through XAD-2 resin and the resin was extracted in the ultrasonic bath for 30 min twice with 100 mL 1/1 ACE/HEX mixture. The extract was filtered through sodium sulfate ( $\text{Na}_2\text{SO}_4$ ) to remove any water residues. If there was no water in BDS, the deposited samples were collected as in dry deposition samples cleaning with ACE/HEX soaked tissue.

Gas chromatography (GC) analyses were conducted by using an HP 7890A- $\mu$ ECD (Micro-Electron Capture Detector) (Hewlett–Packard, USA) instrument. The oven temperature program was 80 °C (1 min), with increases of 20 °C  $\text{min}^{-1}$  to 240 °C (5 min), followed by 5 °C  $\text{min}^{-1}$  up to 270 °C, then 20 °C  $\text{min}^{-1}$  up to 300 °C (3.5 min). The inlet temperature was kept at 250 °C, and the detector temperature was 320 °C. The carrying gas was high purity helium, and the make-up gas was high purity nitrogen. HP5-MS, with dimensions of 30 m  $\times$  0.32 mm  $\times$  0.25  $\mu\text{m}$  was used as a capillary column. Total organic carbon (TOC) was analyzed by HT 1300 Solid TOC Analyzer equipped with Analytic Jena Multi N/C 3100 detector on the particle samples taken with GFF according to Turkish standard method of TS 12089 EN 13137. The determination of TOC was done depending on  $\text{TOC} = \text{Total carbon (TC)} - \text{Inorganic carbon (IC)}$  equation. TC content was determined by measuring the carbon dioxide concentration formed as a result of combustion at 1200 °C with pure oxygen gas with an infrared spectrophotometer. IC content was determined by addition of 2 mL  $\text{H}_3\text{PO}_4$  and shaking with aid of magnetic mixer under heat. The carbon dioxide emitted from reaction was measured by spectrophotometer. Finally, TOC was obtained in percent.

### 2.3. Quality assurance/quality control

External recovery standards were used to determine the efficiencies and the recovery values were usually over 69%, therefore, the reported data were not recovery-corrected (Sofuoglu et al., 2004; Zheng et al., 2010). The instrument detection limit (IDL) was determined for each OCP compound. The IDL ranged from 0.04 pg (Heptachlor epoxide) to 0.15 pg (Methoxychlor). The limit of detection (LOD) for every OCP was determined by adding three standard deviations ( $\text{average} + 3 \times \text{SD}$ ) to the average of the blanks. The values smaller than the LOD were not included in the calculation. OCPs in the samples were all blank-corrected. The LOD values varied from 0.71 (alpha-HCH) to 2.13 ng (Endosulfan beta)

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