



Organochlorine pesticides in the sedimentary core of the southern Tibetan Plateau: The missing pieces induced by lateral remobilization[☆]



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ABSTRACT

The sedimentary core in remote alpine lakes has been recognized as an ideal proxy to reconstruct the emission and air deposition histories of persistent organic pollutants (POPs). Nevertheless, POPs formerly stored in a catchment might also contribute to the variation in the lake sediment by lateral remobilization. In this study, to reveal the relative importance of lateral remobilization, we measured the vertical profiles and isomeric ratios of dichlorodiphenyltrichloroethanes (DDTs), hexachlorocyclohexanes (HCHs) and endosulfan in a dated sedimentary core collected from Lake Yamzho Yumco in the southern Tibetan Plateau. In addition to the flux peaks in the 1970s corresponding to the heavy atmospheric deposition of DDTs and HCHs, these pollutants' fluxes displayed rebounds in the flood periods, with characteristic low ratios of DDT/DDE (α -/ β -HCH). This might indicate a massive remobilization of "weathered" pesticides from catchment soil to the lake because of strong hydro-dynamics. Moreover, the relative contribution of lateral remobilization to the lake sediment in the past decades was recognized through the correlation between DDT/DDE (α -/ β -HCH) ratios and Σ DDT (Σ HCH) fluxes. The results showed that the lateral remobilization contributed to 20–42% of the total fluxes. This study discriminated the air deposition from the contribution of lateral remobilization, which improves current understanding of the vertical POPs profiles in the sedimentary core.

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1. Introduction

Persistent organic pollutants (POPs) in the sedimentary cores of lakes are usually applied to infer the regional pollution history, as lake sediments are effective proxies for environmental components including air deposition and catchment input (Cheng et al., 2014; Liu et al., 2013; Yang et al., 2016). Because of the absence of direct anthropogenic input in remote regions, sedimentary cores have been further used to reconstruct the historical atmospheric fluxes of POPs (Borghini et al., 2005; Malmquist et al., 2003; Muir et al., 1995). The Tibetan Plateau (TP) is an alpine region and is referred to as "the third pole" with an average altitude of higher than

4000 m (Yu et al., 2012). In such a virgin region, POPs were mainly derived from air deposition by cold-trapping after long-range atmospheric transport (LRAT) (Khairy et al., 2016; Yuan et al., 2012, 2014a). Recently, the success in dating the sedimentary core and the travertine profile in the TP (Wu et al., 2011; Yuan et al., 2016) provide the potential to reconstruct the temporal trends of POPs in this region. Although previous studies attempted to interpret the sedimentary records of POPs in the Tibetan lakes (Tao et al., 2014), direct atmospheric deposition was usually regarded as the dominant source, while other factors were neglected.

POPs in sediment should be the chronic combination of POPs from different pathways. Besides direct atmospheric deposition, catchment runoff also contributes POPs that were formerly reserved in the other media of the drainage (Lin et al., 2015). In particular, the unusual soil erosion during the flood periods (Bellucci et al., 2016; León et al., 2017), as well as in the process of human construction (Zhang et al., 2002), might result in a significant amount of POPs released into the runoff and boost their fluxes

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in sediment (Bellucci et al., 2016; Bertrand et al., 2015; León et al., 2017; Zonta et al., 2005). In remote regions at high altitude, the melting glacier was also found to release formerly trapped POPs to sediments of the Alps (Bettinetti et al., 2011; Bogdal et al., 2009; Schmid et al., 2011) and north Canada (Blais et al., 2001; Lafreniere et al., 2006). These above-mentioned processes defined as “lateral remobilization” should be hypothesized to contribute POPs from surrounding reservoirs to sediments in the TP because of the occurrences of strong soil erosion and glacial melting in the TP (Karma, 2000; Shi, 1994; Yu et al., 2012). This indicated that the temporal trends of POP fluxes archived in the lake sediments of the TP could not be limited to only atmospheric deposition but also possibly associated with lateral remobilization. Therefore, a detailed identification of POP sources in the sedimentary cores of the TP is necessary to discriminate the direct air deposition from the contribution of lateral remobilization, whose influence on pollutant flux was previously neglected (Tao et al., 2014). Fortunately, the weathering characteristics of POPs might provide useful information to accomplish this goal. POPs derived from catchment generally undergoes surficial weathering. Thus, they would possess compositional patterns that are different from those of directly atmospheric deposition (Niu et al., 2013).

Organochlorine pesticides (OCPs) are one type of POPs produced mainly for agricultural purpose, and much attention has been paid to their toxic and carcinogenic risks, as well as global occurrence (Arinaitwe et al., 2016; Daly et al., 2007; Khairy et al., 2016; Ribes et al., 2002; Yuan et al., 2014a). After their extensive usage, the recent inputs of many OCPs into the TP from direct emissions were minimized due to their bans in most parts of the world (Li et al., 1996; Voldner and Li, 1995). Nevertheless, their long-term burdens are continuously recorded in the lake sediments of the TP, such as dichlorodiphenyltrichloroethanes (DDTs) and hexachlorocyclohexanes (HCHs) (Cheng et al., 2014), which were suitable target pollutants for achieving our purpose. As mentioned above, previous studies focusing on atmospheric deposition (Xiao et al., 2010) cannot clarify the second flux peak of some legacy OCPs in the sedimentary core of the TP. With sufficient river runoff and few anthropogenic marks (Wang and Dou, 1998), a dated sedimentary core from Lake Yamzho Yumco offering a chronological overview of the historical pollutant fluxes could be used to investigate the relative contributions of POPs from lateral remobilization, and to assess the relevance of this process in the history. This would add more understanding to current knowledge and even provide proof to clarify the environmental process of POPs in the TP.

2. Materials and methods

2.1. Sampling and dating

Lake Yamzho Yumco lies in the east of Nagarze County and 90 km away from Lhasa in Tibet. This lake is mainly supplied by surficial runoff, which accounted for approximately 84% of the total supply (Wang and Dou, 1998). The lake catchment is mainly composed of hills and mountains with an average terrain slope of 19° (Chen et al., 2014). Its water area and catchment area are approximately 638 and 6100 km², respectively. The lake with such a high catchment-to-surface area ratio (9.6) could be strongly influenced by the catchment input (Schmid et al., 2011). The input flux may be further significant due to the sparse alpine vegetation (eg. *Kobresia pygmaea*) and poor organic matter in the catchment (Yuan et al., 2012). Thus, the sedimentation generation within Lake Yamzho Yumco might be susceptible to the contribution of catchment soil. At the same time, the sediments in the littoral zone of the lake might also undergo redistribution by hydro-dynamics, which

would cause focusing of deposition in the deeper portion of the lake during the flood periods. In this study, a sediment profile was collected at water depth of 25 m in the northeast part of Yamzho Yumco using a 15-cm diameter gravity corer in August 2013 (90.41°N; 29.05°E, 4420 m). The sampling location was far away from the river inlets, as shown in Fig. 1. Forty-eight samples were collected in the vertical profile from surface to bottom and sectioned onsite at an interval of 0.5 cm. The detailed sampling and storage methods were the same as those previously described (Yuan et al., 2014b). The precipitation and discharge time series data (Fig. S1 in Supporting Material, SM) were obtained from the observatory station located in the Jiangzi area provided by the National Meteorological Administration (<http://data.cma.cn>).

The age-depth relationship for the lake sedimentary core was established based on the analysis of ¹³⁷Cs and ²¹⁰Pb by γ -ray spectrometry. A constant rate of supply (CRS) dating model of ²¹⁰Pb (Appleby, 2002) and the detection of ¹³⁷Cs peaks respectively in 1964 and 1986 were used for the chronology calculation. The investigated core covered six-decade sediment accumulation (1949–2011), and an average sedimentation rate was determined to be 0.39 cm/yr. This rate value was similar with the result previously reported for the Lake Kemen and Namco in the TP during 1950s–2010s (Yang et al., 2016). Detailed information on the dating methods is provided in Fig. S2 and our previous study (Li et al., 2017).

2.2. Chemicals

The 26 OCP standard solutions (Table S1 in SM) were purchased from Cambridge Isotope Laboratories (Andover, MA). In addition, 2,4,5,6-tetrachloro-m-xylene (TCMX) and 4,4'-dichlorobiphenyl (J&K Chemical, U.S.) were used as a surrogate standard and internal standard for OCPs. Hexane and dichloromethane were purchased from LGC Standards (Wesel, Germany). They were residue grade and additionally distilled in a full glass unit prior to use.

2.3. Extraction and analysis

The extraction process and the analytical method used for OCPs strictly followed the standard analytical methods reported in our previous report (Yuan et al., 2015). Ten grams of each sample was freeze-dried and spiked with surrogate standard solution. The samples were then Soxhlet extracted with dichloromethane for 48 h, with the addition of activated copper in order to remove the sulfur. The extracts were reduced, hexane exchanged and separated on a alumina/silica gel column. The target fraction containing the OCPs was eluted using 30 ml of hexane/dichloromethane (3:2). The internal standard was added and the mixture was condensed to 100 μ L under a gentle N₂ stream.

The OCP analysis was performed via a GC/MS system (6890 gas chromatograph/5973 triple quadrupole mass spectrometer), operating in electron capture negative chemical ionization mode. The separation was performed on a fused silica capillary column (HP-5MS, 30 m \times 0.25 mm i.d. and 0.25 μ m film thickness). The injector and ion source temperatures were 230 °C and 305 °C, respectively. The GC column temperature was initiated at 60 °C for 2 min, increased to 150 °C at a rate of 30 °C/min, then increased to 240 °C at a rate of 2 °C/min, followed by a 20 °C/min increase to 300 °C, which was maintained for 10 min.

2.4. Quality assurance and quality control (QA/QC)

A solvent blank, a procedural blank and a standard mixture were run per 10 samples to check for contamination, as well as peak identification and quantification. The target compounds were not

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