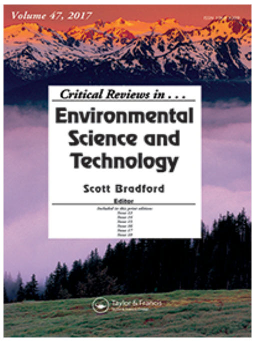

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Sedimentary records of polycyclic aromatic hydrocarbons in China: A comparison to the worldwide

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ABSTRACT

This review focuses on the sedimentary records of polycyclic aromatic hydrocarbons (PAHs) in China and the worldwide. We provide a global image of sedimentary record of PAHs in different regions, including their contents, vertical profiles, and possible sources. Generally, historical PAHs recorded in China, dominated by biomass burning and domestic coal combustion, are moderate on the global scale with an upward trend while elevated levels of historical PAHs are usually found in developed countries with a downward trend. This review ends with some concerns about historical PAHs in sediments and some directions deserved further efforts.

KEYWORDS

Diagnostic ratios; PAHs; sediment core; source

1. Background

Polycyclic aromatic hydrocarbons (PAHs), a family of chemicals with which hundreds of homologues are possible, are ubiquitous in environment (Srogi, 2007). They have received extensive concerns due to their adverse impacts on environment and human beings (Eisler, 1987; Kim et al., 2013; Zhang et al., 2009b). For the same reasons, great efforts have been made to reduce the emission of PAHs in the developed countries during the second half of last century (Mastral and Callen, 2000; Yan et al., 2014). It has been reported that emission of BaP from 33 European countries has fallen by 54.6% from 1970 to 1995 (Pacyna et al., 2003) and the emission of seven carcinogenic PAHs in the United States has also decreased by 30% from 1990 to 1996 (EPA, 1998). Despite this, global emission of PAHs seems constant since the beginning of the industrial period up to 1990s and the reduction of PAH emissions in the developed countries has been offset by the rapid increase in PAH emissions from the developing countries (Masclet et al., 1995; Shen et al., 2011). In addition, it estimated that fossil fuel-derived PAHs have increased 10–30 folds in Norwegian Sea fauna from 1985 to 2010, though that of legacy POPs (such

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as PCBs and DDTs) substantially decreased (De Laender et al., 2011). This means that PAH contamination remains an environmental challenge on the global scale.

The situation may be worse in China. It is well known that China has experienced a rapid economic development after the initiation of the "Reform and Open Policy" since 1978. During this course, China not only enjoyed great economic achievements, but also suffered a lot in environmental deterioration. Large quantities of organic/inorganic pollutants have been released owing to the huge consumption rate of coal for many years (Xu et al., 2006; You and Xu, 2010). Among them, PAHs are of particular interest due to their persistence, long-range transport property, and suspected or proven carcinogenicity and mutagenicity (Boffetta et al., 1997; Bostrom et al., 2002). It has been estimated that the total emission of 16 priority PAHs in China was 25300 tons in 2003 (Xu et al., 2006). More recently, it was updated to 114 Gg, accounting for 21.9% of the total global emission in 2004 (Zhang and Tao, 2009).

PAHs in modern environments are mostly of anthropogenic origin, though natural sources cannot be excluded. To mitigate the adverse impacts of PAHs, the first step is to know how PAHs in environments respond to human activities. Sediment is not only an important sink, but also a valuable natural archive for PAHs (Bigus et al., 2014). Many important pieces of information on PAHs, including historical variation (Guo et al., 2006; Lima et al., 2003) and their sources (Christensen and Arora, 2007; Yan et al., 2014), can be preserved in sediment. Therefore, reconstructing the chronology of PAHs has become a well-established method for obtaining this information, which is of great importance for developing the management strategies and control measures. Up to now, a large number of works on sedimentary record of PAHs have been reported, especially in China (Guo et al., 2010; Li et al., 2015; Mai et al., 2001) and the United States (Lima et al., 2003; Van Metre and Mahler, 2005, 2010). However, this information is mostly fragmented and/or site/region specific, lacking a global perspective on this issue, including the contents, vertical profiles, and sources of historical PAHs, as well as their relationship with anthropogenic activities. Thus, the aims of this review are: 1) to provide a global image of sedimentary record of PAHs in different regions, involving the contents, vertical profiles, and sources of historical PAHs; and 2) to address some concerns and further efforts on the sedimentary record of PAHs.

2. Data sources

Basic information on PAHs, such as physicochemical properties, environmental processes, and the acute/chronic human health effects, has been summarized systematically (Bigus et al., 2014; Ravindra et al., 2008; Yebra-Pimentel et al., 2015). Thus, we start directly with the sedimentary record of PAHs. Because so many related works are available, works cited in this article have been preselected as far as possible based on the following criteria: 1) wide geographic distribution; 2) high depositional resolution; 3) long depositional timescale; 4) more newly published;

and 5) better dated sediment core. It should be noted that: 1) most data cited in the present work are obtained from the original literature using GetData V2.22 and small differences between the extracted data and the original data cannot be avoided, and 2) PAH composition of each previous study is not exactly the same, and 16 priority PAHs proposed by US EPA are considered to be the standard suite of PAHs in this paper.

2.1. Sedimentary record of PAHs in China

Sedimentary records of PAHs in China are summarized in Table 1 and Table 2, most of which were reported within the last two decades, including studies conducted at open seas, estuaries, coastal bays and wetlands, and inland lakes/reservoirs.

In open seas, historical PAHs were analyzed in sediment cores from SCS and YS (Liu et al., 2012a; Peng et al., 2008), in addition to the high-resolution record of PAHs reported at distal mud (Guo et al., 2006) and the continental shelf of ECS (Cai et al., 2016). Meanwhile, vertical profiles of PAHs at the central BHS (Hu et al., 2011) and central YS (Wu et al., 2001) were also presented.

Table 1. Sedimentary record of PAHs along the Chinese coastline.

Location (sampling year)	Depositional year (core)	Range (ng/g)	Peak year	Reference
East China Sea (2003)	1798–2001 (DM1)	27–132[a]	2000	(Guo et al., 2006)
East China Sea (2009)	1928–2009 (DH05)	28.6–96.5[b]	∼1970	(Cai et al., 2016)
East China Sea (2009)	1865–2009 (DH11)	18.8–76.4[b]	2009	(Cai et al., 2016)
South China Sea (2007)	1806–2006 (S1)	32–82.8[b]	1986	(Liu et al., 2012a)
South China Sea (2007)	1928–2006 (S2)	30–80[b]	∼1945	(Liu et al., 2012a)
North South China Sea (2003)	1917–1992	69–216[a]	1992	(Peng et al., 2008)
Yellow Sea (2007)	1816–2006 (YS1)	45–203[b]	∼1996	(Liu et al., 2012a)
Yellow Sea (2007)	1816–2006 (YS2)	19–133[b]	∼1986	(Liu et al., 2012a)
South Yellow Sea (2004)	1920–1998 (D6)	26.3–76.9[a]	1956	(Zhang et al., 2009a)
Bohai Sea (2006)	1942–2004 (BC1)	34.2–202[a]	1999	(Hu et al., 2011)
Bohai Sea (2006)	1902–2004 (BC2)	53.3–168[a]	2004	(Hu et al., 2011)
North Beibu Gulf, SCS (2010)	1950–2010	10.5–87.1[b]	2010	(Li et al., 2015)
Central Beibu Gulf, SCS (2011)	NA	38–74[b]	NA	(Kaiser et al., 2016)
Hailing Bay (2009)	1962–2010 (core B)	5.2–960[b]	2006	(Yu et al., 2012)
Deep Bay (2000)	1901–2004	98.9–674[b]	1992	(Qiu et al., 2009)
Inner Deep Bay (2004)	1963–2003	102–209[a]	1963	(Zhao et al., 2012)
Daya Bay (2003)	1948–2003 (core 8)	77.4–306[a]	1997	(Yan et al., 2009)
Daya Bay (2003)	1953–2003 (core 10)	118–320[a]	2003	(Yan et al., 2009)
Laizhou Bay (2011)	1934–2011	37–211[a]	1986	(Ding et al., 2014)
Haizhou Bay (2010)	1968–2010	72.5–802[a]	2010	(Zhang et al., 2013)
Pearl River Estuary (2000)	1863–1996	59–330[c]	1993	(Liu et al., 2005)
Pearl River Estuary (2003)	1974–2003	312–486[a]	1985	(Peng et al., 2008)
Yangtze River Estuary (2003)	1930–2002 (N6)	33–121[a]	1997	(Guo et al., 2007)
Yangtze River Estuary (2003)	1930–2002 (S5)	54.6–235[a]	2000	(Guo et al., 2007)
Yangtze River Estuary (2011)	NA (CJ03)	141–326[a]	NA	(Wang et al., 2013)
Yangtze River Estuary (2011)	NA (CJ06)	136–262[a]	NA	(Wang et al., 2013)
Yellow River Estuary (2012)	1961–2012	93.2–307[a]	2006	(Yuan et al., 2014)
Liaohe River Estuary (NA)	1720–2012	46–1167[a]	2012	(Guo et al., 2013b)
Yellow Sea (1998)	NA	370–3800[d]	NA	(Wu et al., 2001)
Ma Wan, Hong Kong (2007)	NA	1333–5041[a]	NA	(Li et al., 2009a)

[a] $\sum e_{16}$;
[b] $\sum e_{16}$ subtract NAP;
[c] $\sum ub_{16}$ subtract NAP and plus BeP;
[d] $\sum ub_{16}$ subtract NAP, ACE, ACY, and FLU, plus BeP and PERY.

Table 2. Sedimentary record of PAHs in the Chinese inland lakes or reservoirs.

Lakes (sampling year)	Depositional year (core)	Range (ng/g)	Peak year	Reference
Lake Dianchi (2006)	1937–2005	479–4561[a]	1994	(Guo et al., 2013a)
Lake Dianchi (2009)	1870s–2009	558–6418[a]	~1990s	(Li et al., 2013b)
Hongfeng reservoir (2010)	1963–2010	848–3725[a]	1996	(Li et al., 2013a)
Hongfeng reservoir (2006)	1981–2002	2936–5282[a]	1994	(Guo et al., 2011a)
Lake Yangzonghai (2015)	1952–2014	201–1914[a]	~1998	(Yuan et al., 2017)
Lake Liangtan (2010)	1923–2004	50–1260[a]	2004	(Liu et al., 2012b)
Lake Qinghai (2006)	1860–2000	11–279[a]	1994	(Wang et al., 2010b)
Lake Sugan (2007)	1908–2004	17.6–55.0[b]	2004	(Guo et al., 2010)
Lake Erhai (2006)	1919–2005	72.2–475[b]	2005	(Guo et al., 2010)
Lake Taihu (2000)	1954–2000	180–1600[a]	2000	(Liu et al., 2009)
Lake Nansi (2001)	1910–1995	137–693[a]	1995	(Li et al., 2009b)
Lake Donghu, Wuhan (2006)	1915–2005	33.3–937[a]	2003	(Yang et al., 2011)
Lake Baiyangdian (2009)	1830–2009	97.2–2402[c]	1990	(Guo et al., 2011c)
Lake Baiyangdian (2009)	NA (S1)	140–1754[d]	NA	(Guo et al., 2013c)
Lake Baiyangdian (2009)	NA (S2)	39.5–243[d]	NA	(Guo et al., 2013c)
Lake Baiyangdian (2009)	NA (S3)	40.2–1877[d]	NA	(Guo et al., 2013c)
Lake Nanhu, Wuhan (2006)	NA	328–5343[e]	NA	(Lu et al., 2015)
Dahuofang reservoir (2010)	1955–2009 (N6)	243–1004[a]	1983	(Lin et al., 2012)
Lake Poyang (2006)	NA	163–951[a]	NA	(Wu et al., 2012)
Lake Bosten (2006)	1940–2005	600–1396[a]	2006	(Wu et al., 2012)
Lake Sihailongwan (2009)	1921–2008	474–2289[a]	2008	(Guan et al., 2012)
Lake Lianhua (2007)	1980–2007 (XHL)	477–2247[a]	2004	(Sun and Zang, 2013)
Lake Lianhua (2007)	1980–2008 (HSH)	424–1134[a]	2007	(Sun and Zang, 2013)
Lake Chaohu (2012)	NA (ETY)	144–1080[f]	NA	(Ren et al., 2015)
Lake Chaohu (2012)	NA (EHB)	140–1980[f]	NA	(Ren et al., 2015)
Lake Chaohu (2012)	NA (EYX)	136–2240[f]	NA	(Ren et al., 2015)
Remote Lakes in Tibet (2006–2007)	Core specific (five cores)	98–594[a]	Core specific	(Yang et al., 2016)
Shilianghe Reservoir (2014)	1958–2014	17.4–840[a]	2000	(Zhang et al., 2016)

[a] $\sum an_{16}$;
[b] sum of BaA, CHRY, BbF, BkF, BaP, DBA, and INP;
[c] $\sum PAH_{16}$ subtract INP, DBA, BghiP and plus PERY;
[d] $\sum ub_{16}$ subtract INP, DBA, and BghiP;
[e] $\sum PAH_{16}$ plus BP, BeP, and PERY;
[f] sum of 28 PAH compounds, including $\sum PAH_{16}$.

Along the coastline, initial work associated with PAHs recorded in PRE was reported by Mai et al. (2001), followed by the work conducted at the western shoal of PRE (Liu et al., 2005). Near the YaRE, sedimentary records of PAHs have been reported at the intertidal mud area (Liu et al., 2000), the coastal ECS (Guo et al., 2007), and the shipping route (Wang et al., 2013). More recently, vertical profiles of PAHs at YeRE (Yuan et al., 2014) and LRE (Guo et al., 2013b) were also reported. In addition to the estuaries, sedimentary records of PAHs also can be found in various coastal bays and wetland, such as DB (Qiu et al., 2009), DYB (Yan et al., 2009), HLB (Yu et al., 2012), and BBG (Kaiser et al., 2016; Li et al., 2015) in South China; HZB in East China (Zhang et al., 2013); LZB in North China (Ding et al., 2014); wetland in Hong Kong (Zheng et al., 2002; Ke et al., 2005; Wang et al., 2010a); and coastal margin in Taiwan (Kuo et al., 2014).

Sedimentary records of PAHs in inland lakes/reservoirs are mostly published within the last decade. In Southwest China, historical records of PAHs have been reported at CHH (Guo et al., 2010), EH (Guo et al., 2011b), HF (Guo et al., 2011a; Li et al., 2013a), DC (Guo et al., 2013a; Li et al., 2013b), and YZH (Yuan et al., 2017), as well as remote lakes across Tibet (Yang et al., 2016). In Northwest China, PAHs were analyzed in sediment cores from SG (Guo et al., 2010) and QH (Han et al., 2015; Wang et al., 2010b). In Central and East China, sedimentary records of PAHs have been reported at DH (Yang et al., 2011), PY (Wu et al., 2012), TH (Liu et al., 2009; Tang et al., 2015), CH (Li et al., 2016; Ren et al., 2015), and SLH (Zhang et al., 2016). Besides, published data also included the measurement of PAHs in sediment cores from LH (Sun et al., 2011, 2014), SHLW (Guan et al., 2012), and DHF (Lin et al., 2013) in Northeast China; NS (Li et al., 2009b) and BYD (Guo et al., 2011c) in North China; as well as Feitsui reservoir in Taiwan (Fan et al., 2010).

2.2. Sedimentary record of PAHs in the United States

Initial works on sedimentary record of PAHs in the United States mostly come from its northeastern region, such as Massachusetts (Hites et al., 1977), Rhode Island (Hites et al., 1980), and other sites (Gschwend and Hites, 1981). After the work conducted at Lake Michigan (Simcik et al., 1996), sedimentary records of PAHs were investigated at Milwaukee Harbor Estuary (Li et al., 1998). Besides, historical PAHs were also reconstructed at San Francisco Bay (Nilsen et al., 2015; Pereira et al., 1999), Puget Sound (Louchouarn et al., 2012), and US Gulf Coast (Santschi et al., 2001). At the turn of the new century, high-resolution records of PAHs were developed at Rhode Island (Lima et al., 2003) and Lake Michigan (Schneider et al., 2001), followed by the works performed at Central Park Lake, NY (Yan et al., 2005) and the Great Lakes (Kannan et al., 2005). Meanwhile, a more comprehensive investigation was performed in 38 urban and reference lakes (Van Metre and Mahler, 2005). In addition to these, sedimentary records of PAHs were also reported at other sites, such as Ashtabula River and Black River in Ohio (Gu et al., 2003; Li et al., 2001), Santa Monica Bay, California (Bay et al., 2003), New York/New Jersey harbor complex (Yan et al., 2006), Rocky Mountain (Usenko et al., 2007), and several inland lakes in Michigan (Parsons et al., 2014). Some of them are summarized in Table 3.

2.3. Sedimentary record of PAHs in Europe

Sedimentary record of PAHs in Europe can retrospect to 1980s (Pavoni et al., 1987; Readman et al., 1987). It still draws great attention, even very recently (Arp et al., 2011; Azoury et al., 2013; Bogdal et al., 2011; Vane et al., 2011). Vertical profiles of PAHs have been reported at Scheldt estuary, Netherlands (Van Zoest and Van Eck, 1993). Similar works can be found at Venice Lagoon, Italy (Frignani et al., 2003), Mersey Estuary, UK (Vane et al., 2007), River Elbe, Germany (Götz et al., 2007), and Lake Peipsi (Punning et al., 2008), as well as remote lakes in Scotland (Rose and Rippey, 2002) and Croatia (Mikac et al., 2011). Historical atmospheric PAHs have been reconstructed at high altitude lakes throughout Europe, including the High Tatras and Pyrenees (Fernández et al., 2000; Grimalt et al., 2004). Historical records of PAHs are usually less than two hundred years. However, PAH records as long as 700 years have been reported at Aspvreten, Sweden (Elmquist

Table 3. Sedimentary record of PAHs in the United States.

Location (sampling year)	Depositional year (core)	Range (ng/g)	Peak year	Reference
Mississippi river delta (1993)	1892–1992	199–800[a]	1966	(Santschi et al., 2001)
Galveston Bay (1995)	1892–1993	53–479[a]	1968	(Santschi et al., 2001)
Tampa Bay 1993)	1946–1993	769–20365[a]	1989	(Santschi et al., 2001)
Richardson Bay (1992)	1910–1992 (RB92-3)	401–6273[b]	~1950	(Pereira et al., 1999)
San Pablo Bay (1990)	1952–1990 (SP90-8)	157–1298[b]	~1988	(Pereira et al., 1999)
Lake Central Park (1996)	1880–1996 (CPF)	0–27[c*]	1916	(Yan et al., 2005)
Lake Ballinger (1998)	1948–1998	4.8–49[d*]	1992	(Van Metre and Mahler, 2005)
Lake in Chicago (2001)	1942–2001	0.32–23[d*]	2001	(Van Metre and Mahler, 2005)
Lake Crystal M (2000)	1918–2000	159–1456[a]	1992	(Kannan et al., 2005)
Lake Crystal B (2001)	1916–2001	40–511[a]	1986	(Kannan et al., 2005)
Lake Mullett (2001)	1930–2000	7.7–230[a]	1981	(Kannan et al., 2005)
Lake Whitmore (2001)	1969–2001	502–2582[a]	2001	(Kannan et al., 2005)
Lake Cadillac (2001)	1886–2001	417–10438[a]	1994	(Kannan et al., 2005)
Taunton river (2004)	1880–2001(SR)	0.15–14[e*]	1962	(Cantwell et al., 2007)
Taunton river (2003)	1880–2001(MC)	0.2–356[e*]	1968	(Cantwell et al., 2007)
Taunton river (2003)	1875–2001(SJ)	0.7–34[e*]	~1885	(Cantwell et al., 2007)
Lake Michigan (1998)	1870–1998 (HMS1)	634–4257[f]	1942	(Schneider et al., 2001)
Hood Canal (2005)	1818–2004 (HC-3)	60–602[g]	1971	(Louchouarn et al., 2012)
Hood Canal (2005)	1773–2004 (HC-5)	78–1423[g]	1955	(Louchouarn et al., 2012)
Lake Round (2004)	1855–2005	122–1977[h]	1929	(Parsons et al., 2014)
Lake Muskegon (2004)	1967–2004	2250–4663[h]	1969	(Parsons et al., 2014)
Lake Crystal (2004)	1816–2007	149–1707[h]	2006	(Parsons et al., 2014)
Lake Birch (2004)	1833–2004	80–2093[h]	1957	(Parsons et al., 2014)
San Francisco Bay	1892–2004	120–9560[i]	~1947	(Nilsen et al., 2015)
Ashtabula River, Ohio (1998)	1965–1998 (AR-1)	792–4817[j]	1986	(Li et al., 2001)
Black River, Ohio (1998)	Site specific (core BR2~6)	0.1–250[i*]	Site specific	(Gu et al., 2003)
Lake lady bird (2012)	1962–2014 (LBL-4)	272–6338[a]	~2000	(Van Metre and Mahler, 2014)
Milwaukee Harbor Estuary (1990/4)	Site specific (21 cores)	46.6–287.1[l*]	Site specific	(Li et al., 1998)

[*] unit: μg/g;
[a] $\sum gt_{16}$;
[b] $\sum 6gt_{16}$ subtract DBA, plus 4 alkyl-PAHs, triphenylene, and PERY;
[c] $\sum ub_{16}$ plus some C_1-C_4 alkyl homologues, dibenzothiophene series, and retene;
[d] sum of 13 PAHs used for consensus based sediment quality guidelines;
[e] $\sum m_{16}$ subtract NAP, ACE, ACY, BbF, and BkF, plus BeP and PERY;
[f] sum of 33 PAHs compounds;
[g] sum of FLUO, PYR, BaA, CHRY, BbF, BkF, BaP, INP, DBA, and BghiP;
[h] $\sum PAH_{16}$ subtract NAP and BaP, plus BjF;
[i] sum of 24 PAHs compounds;
[j] sum of NAP, FLU, PHEN, ANT, PYR, BaA, and BaP;
[k] $\sum m$ of plus 1-MNAP and 2-MNAP;
[l] average values of 16 PAHs compounds at different sampling sites.

et al., 2007) and Urdaibai estuary, Spain (Leorri et al., 2014), with the longest record of PAHs reported at Eifel, Germany (Musa Bandowe et al., 2014). Besides, historical records of PAHs were also reported in Baltic Sea (Ricking and Schulz, 2002), Ireland (O'Dwyer and Taylor, 2009), Czech Republic (Francu et al., 2010), Italy (Di Leonardo et al., 2009; Feo et al., 2011), and the Mediterranean (Azoury et al., 2013; Tolosa et al., 1996). Some of them are displayed in Table 4.

2.4. Sedimentary record of PAHs in Japan and Canada

Apart from the works mentioned above, investigations on the sedimentary record of PAHs are relatively centralized in Japan and Canada. Most of them

Table 4. Sedimentary record of PAHs in Europe.

Location (sampling year)	Depositional year (core)	Range (ng/g)	Peak year	Reference
Scheldt estuary, Netherland (NA)	1932–1986 (K)	2730–8475[a]	1964	(Van Zoest and Van Eck, 1993)
Scheldt estuary, Netherland (NA)	1896–1986 (E)	2048–6167[a]	1968	(Van Zoest and Van Eck, 1993)
Remote Lake, Scotland (1996)	1830–1987	632–1718[b]	1927	(Rose and Rippey, 2002)
Venice, Italy (1998)	1849–1996 (E)	113–1531[b]	1914	(Frignani et al., 2003)
Venice, Italy (1998)	1871–1996 (F)	66–734[b]	1948	(Frignani et al., 2003)
Venice, Italy (1998)	NA (I1)	2860–16400[b]	NA	(Frignani et al., 2003)
River Elbe, Germany (1995)	1940–1992 (PT)	506–43580[b]	1964	(Götz et al., 2007)
River Elbe, Germany (1995)	1880–1996 (HL)	401–39700[b]	1958	(Götz et al., 2007)
Lake Prosce, Croatia (2003)	1968–2003	285–407[b]	1992	(Mikac et al., 2011)
Lake Kozjak, Croatia (2003)	1951–2003	167–301[b]	1983	(Mikac et al., 2011)
Aspvreten, Sweden (2005)	1343–2005	208–3623[c]	~1962	(Elmquist et al., 2007)
Lake Peipsi, Estonia (2006)	1878–2003 (PS4)	73.7–393[d]	1992	(Punning et al., 2008)
Oslo harbor, Norway (2006)	1830–2004 (H4)	104–27617[e]	1953	(Arp et al., 2011)
Peat core, UK (1990)	1775–1989	1250–25880[f]	1932	(Sanders et al., 1995)
Lake Ladove (2001)	1924–1997	1265–15305[g]	1988	(van Drooge et al., 2011)
Lake Starolesnianske (2001)	1823–1993	1925–17042[g]	1985	(van Drooge et al., 2011)
Lake Dlugi, Staw (2001)	1840–1993	469–12066[g]	1993	(van Drooge et al., 2011)
Clyde estuary, Scotland (2004)	1823–1993	171–3028[h]	1972	(Vane et al., 2011)
Kanala marsh, Spain (2004)	1300–2000	217–4680[i]	1971	(Leorri et al., 2014)
Holzmaar, Germany	600 BC–2010	62–8000[j]	~1960s	(Bandowe et al., 2014)
Gotland basin, Baltic Sea	NA (GB)	52–5200[a]	NA	(Ricking and Schulz, 2002)
Arkona basin, Baltic Sea	NA(AB)	305–3233[a]	NA	(Ricking and Schulz, 2002)
Mersey Estuary, UK (2000–2002)	NA (M102)	1501–3207[h]	NA	(Vane et al., 2007)
Mersey Estuary, UK (2000/2)	NA (M122)	3164–3766[h]	NA	(Vane et al., 2007)

[a] $\sum ne_{16}$ subtract NAP, ACE, ACY, and FLU, plus BeP;
[b] $\sum bt_{16}$;
[c] PHEN, ANT, MP, 1.7-DMP, 2.6-DMP, FLUO, PYR, B(*a*)A, CHRY, B(*b*+*j*+*k*)F, B(*a*)P, B(*e*)P, B(*ghi*)P, DBA, COR;
[d] $\sum{}_{,\,16}$ subtract ACE, ACY, ANT and FLU;
[e] $\sum PAH_{16}$ subtract NAP, ACE, ACY and FLU;
[f] $\sum PAH_{16}$ subtract ACY, BbF, BkF, DBA, INP, plus COR;
[g] $\sum PAH_{16}$ subtract NAP, ACE, and ACY, plus BeP and PERY;
[h] $\sum PAH_{16}$ subtract ACY;
[i] sum of 33 PAHs compounds;
[j] sum of 30 PAHs compounds.

are shown in Table 5. The first work on sedimentary record of PAHs from Japan was conducted at Tokyo Bay (Kannan et al., 2000; Yamashita et al., 2000), followed by the works carried out at Chidorigafuchi Moat (Okuda et al., 2002), moat in Osaka Castle (Moriwaki et al., 2005), and a reservoir in Osaka City (Ishitake et al., 2007). In lacustrine environment, PAHs were measured in sediment cores from Lake Biwa (Itoh et al., 2010) and other lakes (Fukushima et al., 2012). Besides, a record of PAHs over one thousand years was reported at Lake Kitaura (Itoh et al., 2017).

Most works on sedimentary record of PAHs in Canada were reported very recently, such as works conducted at Lakes in Alberta (Donahue et al., 2006; Jautzy et al., 2013; Korosi et al., 2013, 2016), lakes in Athabasca (Ahad et al., 2015; Evans et al., 2016), Eastern Canadian Arctic (Foster et al., 2015), and a slightly earlier, in Lake Tantaré (Gallon et al., 2005). Additionally, Yunker and his co-authors published a lot of works of this kind, conducted at Fraser River Basin, Strait of Georgia, and British Columbia (Yunker and Macdonald, 2003; Yunker et al., 1999, 2014, 2015).

Table 5. Sedimentary record of PAHs in Japan and Canada.

Location (sampling year)	Depositional year	Range (ng/g)	Peak year	Reference
Tokyo Bay, Japan (1995)	1901–1995	38–2000[a]	1982	(Yamashita et al., 2000)
Chidorigafuchi, Japan (1997)	1930–1997	9000–35000[b]	1959	(Okuda et al., 2002)
Moat, Osaka, Japan (1995)	1671–1976	53–26000[c]	1944	(Moriwaki et al., 2005)
Nagaike Pond, Japan (2001)	1935–2000	4200–26000[c]	1945	(Ishitake et al., 2007)
Lake Kizaki, Japan (2005)	1954–2005	451–1874[d]	1963	(Fukushima et al., 2012)
Lake Kasumigaura, Japan (2005)	1941–2004	126–519[d]	1994	(Fukushima et al., 2012)
Lake Shinji, Japan (2005)	1905–2005	97.6–767[d]	1976	(Fukushima et al., 2012)
Lake Suwa, Japan (2005)	1952–2005	273–976[d]	1968	(Fukushima et al., 2012)
Lake Biwa, Japan (2007)	NA	559–1660[e]	NA	(Itoh et al., 2010)
Lake Kitaura, Japan (2002)	988–1999	380–827[f]	∼1990	(Itoh et al., 2017)
Lake Tantaré, Canada (1997)	1678–1996	0.003–0.237[g]	1951	(Gallon et al., 2005)
Sydney harbor, Canada (2001)	1890–1999 (LSR)	6000–487000[a]	∼1976	(Smith et al., 2009)
Lake Simcoe, Canada (2008)	1886–2008	453–4578[h]	1957	(Helm et al., 2011)
Lakes in Athabasca, Canada (2010)	NA	106–324[i]	NA	(Jautzy et al., 2013)
Strait of Georgia, Canada (2003)	NA (GVRD1)	91–573[a]	NA	(Yunker et al., 2015)
Strait of Georgia, Canada (2003)	NA (GVRD2)	779–1427[a]	NA	(Yunker et al., 2015)
Ethel Lake, Canada (2010)	NA	64–436[a]	NA	(Korosi et al., 2013)
Hilda Lake, Canada (2010)	NA	24–1342[a]	NA	(Korosi et al., 2013)
Cold lake in Alberta, Canada (2014)	1872–2015	85–200[j]	∼1915	(Korosi et al., 2016)
Hecate Strait, Canada (2003)	1920–2003 (JP8)	135–215[k]	1964	(Yunker et al., 2014)
Hecate Strait, Canada (2003)	1870–2003 (MS1)	57–665[k]	1990	(Yunker et al., 2014)
Baffin Bay, Canada (2008/9)	NA	21–201[l]	NA	(Foster et al., 2015)
Saskatchewan Lakes, Canada (2010)	NA (10W)	9.3–70.3[m]	NA	(Ahad et al., 2015)
Saskatchewan Lakes, Canada (2010)	NA (13C)	15.5–92.1[m]	NA	(Ahad et al., 2015)

[a] $\sum \text{had}_{16}$;
[b] sum of PHEN, FLUO, PYR, BbF, BkF, BjF, BaP, BeP, INP and BghiP;
[c] sum of PHEN, FLUO, BaA, CHRY, PYR, BbF, BkF, BaP, and BghiP;
[d] $\sum \text{PAH}_{16}$ plus BeP;
[e] $\sum \text{PAH}_{16}$ plus B[j/a]F, BeP, and PERY.
[f] sum of 20 PAHs compounds including $\sum \text{PAH}_{16}$;
[g] $\sum \text{PAH}_{16}$ subtract NAP, ACE, ACY, BaA, plus BeP; (unit: flux: nmol cm^{-2} y^{-1});
[h] $\sum \text{us}_{16}$ plus BeP and PERY;
[i] $\sum \text{us}_{16}$ minus NAP, plus dibenzothiophene;
[j] $\sum \text{nu}_{16}$ plus RET and dibenzothiophene;
[k] 28 Parent PAHs, include $\sum \text{PAH}_{16}$;
[l] $\sum \text{P}_{16}$ subtract ACY;
[m] $\sum$ubtr subtract NAP.

2.5. Sedimentary record of PAHs in other areas

The rest of the works on sedimentary record of PAHs are highly scattered geographically. Most of them are summarized in Table 6. In Korea, sedimentary record of PAHs has been reported by Yim et al. (2005), but seldom reported thereafter. In South Asia, sedimentary record of PAHs is often reported in India (Binelli et al., 2008; Goswami et al., 2016) and Sri Lanka (Ohura et al., 2015), but is lacking dating data. In Southeast Asia, individual work can be found at the Gulf of Thailand (Boonyatumanond et al., 2007), Chini Lake in Malaysia (Bakhtiari et al., 2009), Jakarta Bay in Indonesia (Rinawati et al., 2012), and coastal lagoons in Vietnam (Giuliani et al., 2008, 2015). In addition, historical records of PAHs are also reported at several African lakes, such as Lake Zeekoevlei in South Africa (Das et al., 2008), Lake Ukwa Ibom in Nigeria (Oyo-Ita and Oya-Ita, 2013), and Lake Maryut in Egypt (Barakat et al., 2011). Other studies can be found at White Sea (Savinov et al., 2000), Caspian Sea (Varnosfaderany et al., 2014), Arabian Gulf

Table 6. Sedimentary record of PAHs in other area.

Location (sampling year)	Depositional year (core)	Conc. (ng/g)	Peak year	Reference
Masan Bay, Korea (1997)	1922–1999 (C1)	37.8–277[a]	1992	(Yim et al., 2005)
Haengam Bay, Korea (1997)	1934–1999 (C2)	88–756[a]	1977	(Yim et al., 2005)
Lagoon, Sri Lanka (2004)	NA	378–1511[b]	NA	(Ohura et al., 2015)
Kandy Lake, Sri Lanka (2004)	NA	1393–3153[b]	NA	(Ohura et al., 2015)
Sundarban Wetland, India (2005)	NA (S1 and S3–7)	9.4–1144[c]	NA	(Binelli et al., 2008)
Sundarban Wetland, India (2005)	NA (S2)	1098–4250[c]	NA	(Binelli et al., 2008)
Sundarban Wetland, India (NA)	NA (S1–4)	132–2491[d]	NA	(Dominguez et al., 2010)
Lake Nainital, India (2004)	1919–2004	12–216[*]	2004	(Choudhary and Routh, 2010)
Lake Bhimtal, India (2004)	1916–2004	1–217[*]	2004	(Choudhary and Routh, 2010)
Pichavaram, India (NA)	1908–2002	106–273[#]	1989	(Ranjan et al., 2012)
Cooum river, India (2014)	NA (CR3)	31–31423[b]	NA	(Goswami et al., 2016)
Ennore estuary, India (2014)	NA (E1)	5.4–901[b]	NA	(Goswami et al., 2016)
Pulicat lake, India (2014)	NA (P1)	53–520[b]	NA	(Goswami et al., 2016)
Gulf of Thailand (2004)	1931–2004 (GT15)	8.0–117[e]	1977	(Boonyatumanond et al., 2007)
Chini Lake, Malaysia (2007)	NA (core B)	192–1895[f]	NA	(Bakhtiari et al., 2009)
Chini Lake, Malaysia (2007)	NA (core C)	570–6782[f]	NA	(Bakhtiari et al., 2009)
Chini Lake, Malaysia (2007)	NA (core D)	325–4435[f]	NA	(Bakhtiari et al., 2009)
Chini Lake, Malaysia (2007)	NA (core E)	212–1898[f]	NA	(Bakhtiari et al., 2009)
lagoons, Vietnam (2002/5)	NA (ten cores)	95–465[g]	Core specific	(Giuliani et al., 2008)
Nador Lagoon, Vietnam (2009)	1921–2009 (Nad 1)	59–108[g]	2009	(Giuliani et al., 2015)
Lake Maryut, Egypt (2005)	NA	15–5560[b]	NA	(Barakat et al., 2011)
Kuwait Bay, Arabian Gulf	NA	12–45[g]	NA	(Gevao et al., 2016)
Caspian Sea (2012)	1808–2009	202–736[h]	1987	(Varnosfaderany et al., 2014)
Cocó river, Brazil	NA	3.04–2235[b]	NA	(Cavalcante et al., 2009)
Ceará river, Brazil	NA	3.34–1859[b]	NA	(Cavalcante et al., 2009)
Santos Estuary, Brazil (2002)	1963–2002(LCN)	852–7145[b]	1963	(Martins et al., 2011)
Santos Estuary, Brazil (2002)	1963–2002(LSR)	1792–3019[b]	2002	(Martins et al., 2011)
Santos Estuary, Brazil (2002)	1975–2002(CQ1)	3723–6405[b]	2001	(Martins et al., 2011)
Barigui river, Brazil (2011)	1855–2010	1766–14924[b]	1972	(Machado et al., 2014)
Todos os Santos Bay, Brazil (2002)	1904–2002	8.6–334[b]	1963	(Costa et al., 2016)
Guaratuba Bay, Brazil (2005)	NA(GT4)	1.5–3130[i]	NA	(Pietzsch et al., 2010)
Guaratuba Bay, Brazil (2005)	NA(GT12)	78.5–3270[i]	NA	(Pietzsch et al., 2010)
Las Matas, Mexico (2005)	1907–2005 (LSR)	259–1176[b]	1992	(Ruiz-Fernández et al., 2012)
El Tule Lake, Mexico (2010)	NA	50–853[b]	NA	(Ruiz-Fernández et al., 2014)
Santa Elena Lake Mexico (2010)	NA	119–363[b]	NA	(Ruiz-Fernández et al., 2014)
Laja lake, Chili (2002)	1961–2001	226–620[j]	2001	(Quiroz et al., 2005)
George island, Antarctica (2000)	1865–2000 (B)	2.6–10.6[k]	~1993	(Martins et al., 2010)
George island, Antarctica (2005/6)	1951–2004 (C)	5.4–20.4[k]	~2004	(Martins et al., 2010)
George island, Antarctica (2005/6)	1904–2002 (E)	0–34.6[k]	~1979	(Martins et al., 2010)
Jakarta Bay, Indonesia (2009)	NA	39–214[l]	NA	(Rinawati et al., 2012)

[*] PAHs composition is not clear, Unit: ng mg^{-1} C;
[#] $\sum$C;s$_{16}$, Unit: μg cm^{-2} year^{-1};
[a] $\sum$ea$_{16}$ plus 1-MNAP, 2-MNAP, BP, 2,6-DMNAP, 2,3,5-TMNAP, 1-MPHEN, BeP, and PERY;
[b] $\sum$lu$_{16}$;
[c] $\sum$lu$_{16}$ plus 1-MNAP, 2-MNAP, and PERY;
[d] $\sum$lu$_{16}$ subtract NAP, ACE, ACY, and FLU;
[e] sum of PHEN, MPHEN, ANT, FLUO, PYR, BaA, CHRY, B(b+k+j)F, BaP, BeP, INP, BghiP, and COR;
[f] sum of NAP, PHEN, ANT, FLUO, PYR, CHRY, BaA, BkF, BaP, and BeP;
[g] $\sum$um$_{16}$ subtract NAP;
[h] sum of 29 PAHs compounds, including $\sum$PAH$_{16}$;
[i] $\sum$m$_{16}$ subtract ACE and CHRY;
[j] $\sum$bt$_{16}$ subtract ACY and BbF, plus PERY;
[k] $\sum$PAH$_{16}$ subtract NAP, ACE, ACY, FLU, ANT, and PHEN plus BeP;
[l] sum of PHEN, ANT, CPP, FLUO, PYR, BaA, CHYR, BbF, BF, BeP, BaP, INP, BghiP, and COR.

(Gevao et al., 2016), Las Matas, and remote lakes in Mexico (Ruiz-Fernández et al., 2012, 2014). There are many works on the sedimentary record of PAHs from South America, such as the works conducted at Cocó and Ceará rivers (Cavalcante et al., 2009), Barigui river (Machado et al., 2014), Santos Estuary (Martins et al., 2011), and Turvo/Grande watershed in Brazil (Pantano et al., 2016), Río de la Plata Estuary in Argentina (Colombo et al., 2006), and Lake Laja (Quiroz et al., 2005) and Lake Chungará in Chile (Pozo et al., 2014). It is worthwhile noting that there is one work carried out at King George Island, Antarctica (Martins et al., 2010).

In a word, data sources associated with the sedimentary records of PAHs were mostly from China, Europe, and the United States, and to a less extent, from Canada and Japan.

3. Contents of PAHs in sediment cores

3.1. Contents of sedimentary recorded PAHs in China

Contents of sedimentary recorded PAHs are generally low along the Chinese coastline (Table 1), except for PAHs reported at YS (Wu et al., 2001) and Hong Kong (Li et al., 2009a). In the open seas, levels of $\sum PAH_{15}$ ($\sum PAH_{16}$ subtract NAP) in the range of 32–83 ng/g (core S1) and 30–80 ng/g (core S2) were reported in sediment cores from SCS (Liu et al., 2012a). This is apparently less than that of $\sum PAH_{15}$ recorded in the north SCS (69–216 ng/g) (Peng et al., 2008). Guo et al. (2006) reported that concentrations of $\sum PAH_{16}$ at the distal mud area of ECS were in the range of 27–132 ng/g, comparable to the concentrations of $\sum PAH_{15}$ recorded in the continental shelf of ECS (DH05: 29–97 ng/g and DH11: 19–76 ng/g) (Cai et al., 2016). Contents of PAHs recorded in ECS are comparable or less than that of $\sum PAH_{15}$ recorded in YS (YS1: 19–133 ng/g; YS2: 45–203 ng/g) (Liu et al., 2012a), but slightly higher than that of $\sum PAH_{16}$ reported at south YS (26–77 ng/g) (Zhang et al., 2009a). As to BHS, contents of $\sum PAH_{16}$ are in the range of 34–202 ng/g at BS1 and 53–168 ng/g at BS2, respectively (Hu et al., 2011). Generally, historical PAHs concentrations at open seas are higher in sediment cores from the north than from the south.

Along the coastline, concentrations of PAHs are in the range of 59–330 ng/g at sediment core from PRE (Liu et al., 2005). This is close to the result ($\sum PAH_{16}$: 93–307 ng/g) recorded at YeRE (Yuan et al., 2014) and is less than that of PAHs recorded at LRE ($\sum PAH_{16}$: 46–1167 ng/g) (Guo et al., 2013b), but is higher than that of $\sum PAH_{16}$ (core N6: 33–121 ng/g and core S5: 55–235 ng/g) recorded at YaRE (Guo et al., 2007). The relatively low concentrations of $\sum PAH_{16}$ at YaRE (core N6) are probably ascribed to the diluting effect of high sedimentation rate at the sampling site (Guo et al., 2007). Concentrations of $\sum PAH_{15}$ in cores from the north and central Beibu Gulf are in the range of 11–87 ng/g and 38–74 ng/g, respectively (Kaiser et al., 2016; Li et al., 2015), which are much less than that recorded at DYB ($\sum PAH_{16}$: 77–320 ng/g) (Yan et al., 2009), DB ($\sum PAH_{15}$: 99–674 ng/g) (Qiu et al., 2009), and HLB ($\sum PAH_{15}$: 5–960 ng/g) (Yu et al., 2012).

Comparable to that of $\sum PAH_{15}$ recorded at DB, concentrations of $\sum PAH_{16}$ in core from HZB are in the range of 73–805 ng/g (Zhang et al., 2013). This is much higher than that recorded at LZB ($\sum PAH_{16}$: 37–211 ng/g) (Ding et al., 2014) and inner DB ($\sum PAH_{16}$: 102–209 ng/g) (Zhao et al., 2012). In fact, sedimentary records of PAHs were also reported at other sites in PRE (Peng et al., 2008) and YaRE (Liu et al., 2000), with PAHs concentrations in the range of 312–486 ng/g ($\sum PAH_{16}$) and 0.08–12 μg/g (sum of 15 PAH compounds), respectively. They are clearly different from the results mentioned above, though the sampling sites are close to each other. This implies that the sedimentary records of PAHs are site specific. Roughly, PAHs recorded at the estuaries and coastal bays are higher than that recorded at open seas.

Contents of historical PAHs in inland lakes/reservoirs are highly site specific (Table 2). DC and HF are the typical urban lakes in Southwest China. They are heavily polluted by PAHs, with sedimentary record of $\sum PAH_{16}$ in the range of 479–4561 ng/g (Guo et al., 2013a) and 848–3725 ng/g (Li et al., 2013a), respectively. Levels of $\sum PAH_{16}$ detected in sediment core from SHLW (474–2289 ng/g) is compared with that recorded in LH (core XHL: 477–2247 ng/g) (Sun and Zang, 2013) and BYD (97–2402 ng/g) (Guo et al., 2011c). All these are much higher than that of $\sum PAH_{16}$ (98–595 ng/g) recorded in remote lakes across the Tibetan Plateau (Yang et al., 2016). Guo et al. (2010) have analyzed PAHs in sediment cores from several lakes in West China. However, only seven carcinogenic PAHs are included with contents in the range of 18–55 ng/g, 18–74 ng/g, and 72–475 ng/g for SG, Bosten, and EH, respectively. Concentrations of $\sum PAH_{16}$ recorded in QH, another lake in West China, are in the range of 15–355 ng/g (Wang et al., 2010b), which is less than that of $\sum PAH_{16}$ recorded in TH (core ML: 180–1600 ng/g) (Liu et al., 2009) and LT (core ML: 50–1260 ng/g) (Liu et al., 2012b). Clearly, PAHs recorded in inland lakes/reservoirs are higher than that recorded in the coastline, especially for urban lakes.

3.2. Contents of sedimentary recorded PAHs in USA

Concentrations and the analyzed PAH compounds associated with the sedimentary record of PAHs in the United States are highly varied (Table 3). In Milwaukee Harbor, significant amounts of PAHs were deposited in sediment during the second half of the last century, with mean values of 47–287 μg/g depending on the sampling locations (Li et al., 1998). The maximum values of PAHs reported in Taunton River are 14 and 34 μg/g at SR and SJ, and even up to 356 μg/g at MC (Cantwell et al., 2007). High levels of PAHs are also reported in other well-urbanized estuaries along the coast of the United States, such as Tampa Bay (769–20365 ng/g) and San Francisco Bay (120–9560 ng/g). Urban lakes are important sinks for PAHs, such as urban lakes in Seattle (Lake Ballinger: 4.8–49 μg/g) and Chicago (lake in the hills: 0.32–23 μg/g) (Van Metre and Mahler, 2005), as well as the Central Park Lake (0–27 μg/g) in NY (Yan et al., 2005). Besides, significantly high levels of PAHs were also recorded in sediments from Ashtabula River (792–4817 ng/g) and Black River (0.1–250 μg/g), Ohio (Gu et al., 2003; Li et al., 2001). However, along the US Gulf Coast, PAHs recorded in Mississippi River Delta and Galveston Bay are in the range of 199–800 ng/g and 53–479 ng/g, much less than that recorded in Tampa Bay (Santschi et al., 2001). In inland lakes in Michigan, contents of sedimentary recorded PAHs varied significantly. PAHs recorded in Lake Crystal B (40–511 ng/g) and Lake Mullett (7.7–230 ng/g), probably representing the lowest end of PAHs recorded in the United States, are at least one or two orders of magnitude less than that in other inland lakes, such as Lake Cadillac (417–10438 ng/g) (Kannan et al., 2005) and Lake Muskegon (2250–4663 ng/g) (Parsons et al., 2014). This indicates that historical contamination of PAHs in the United States is much heavier than that in China, especially at the coastline and urban lakes.

3.3. Contents of sedimentary recorded PAHs in Europe

Collected data suggest that contents of historical PAHs are generally high in Europe (Table 4). In Germany, significant high levels of PAHs were recorded in River Elbe (core PT: 506–43580 ng/g and core HL: 401–39700 ng/g) (Götz et al., 2007) and Lake Holzmaar (62–8000 ng/g) (Bandowe et al., 2014). Similar situation can be found in the High Tatras, such as PAHs recorded in Lake Ladové (1265–15305 ng/g), Lake Starolesnianske Pleso (1925–17042 ng/g), and Lake Dlugi Staw (469–12066 ng/g) (van Drooge et al., 2011). In Europe, estuaries are usually heavily contaminated. Early on, high contents of PAHs have been found in both cores from Scheldt Estuary, Netherland (core K: 2730–8475 ng/g and core E: 2048–6167 ng/g) (Van Zoest and Van Eck, 1993). They are higher than that of PAHs recorded in sediments from Clyde Estuary, Scotland (171–3028 ng/g) (Vane et al., 2011), Mersey Estuary, UK (core M102: 1501–3207 ng/g and core M122: 3164–3766 ng/g) (Vane et al., 2007), and Urdaibai Estuary, Spain (217–4680 ng/g) (Leorri et al., 2014), but are less than that of PAHs recorded in sediments from Oslo harbor, Norway (core H4: 104–27617 ng/g) (Arp et al., 2011) and San Giuliano Canal, Venice (2860–16400 ng/g) (Frignani et al., 2003). In Baltic Sea, PAHs recorded in Arkona basin (305–3233 ng/g) are less than that in Gotland basin (52–5200 ng/g) (Ricking and Schulz, 2002), but are comparable with that recorded in Aspvreten, Sweden (208–3623 ng/g) (Elmquist et al., 2007). At a remote lake in north-west Scotland, moderate levels of PAHs (632–1718 ng/g) were reported by Rose and Rippey (2002). They are clearly less than the results mentioned above, but are higher than historical PAHs recorded in Lake Prosce (285–407 ng/g) and Lake Kozjak (167–301 ng/g), Croatia (Mikac et al., 2011), and Lake Peipsi, Estonia (74–393 ng/g) (Punning et al., 2008). Generally, historical contamination of PAHs in Europe is comparable to that in the United States and is much heavier than that in China.

3.4. Contents of sedimentary recorded PAHs in Japan and Canada

Contents of sedimentary recorded PAHs in Japan and Canada are summarized in Table 5. Based on the available data, contents of historical PAHs in Japanese lacustrine environment are in the range of 98–1874 ng/g, with relatively high levels found in Lake Biwa (559–1660 ng/g) and Lake Kizaki (451–1874 ng/g) (Fukushima et al., 2012; Itoh et al., 2010, 2017). They are compared with PAHs recorded in Tokyo Bay (38–2000 ng/g) (Yamashita et al., 2000), and are moderate when compared with PAHs recorded in Chinese inland lakes. However, significantly high PAHs levels can be found in Japanese urban environments. Historical PAHs ranging from 53 to 26000 ng/g have been reported in sediments from the Moat in Osaka Castle (Moriwaki et al., 2005). They are compared with PAHs recorded in Nagaike Pond (4200–26000 ng/g), a reservoir in Osaka City (Ishitake et al., 2007), but are still less than PAHs recorded in Chidorigafuchi Moat (9000–35000 ng/g) (Okuda et al., 2002).

Although relatively high levels of PAHs can be found in sediment cores from Lake Simcoe in Ontario (Helm et al., 2011) and Lake Tantaré in Québec (Gallon et al., 2005), reported data suggest that contents of sedimentary recorded PAHs in Canada are generally low in most cases, probably related to the remoteness of the sampling locations. On the west coast of Canada, PAHs analyzed in sediments from Hecate Strait were in the range of 135–215 ng/g and 57–665 ng/g at core JP8 and core MS1, respectively (Yunker et al., 2014). Much less historical PAHs (9–92 ng/g) were detected in Saskatchewan lakes (Ahad et al., 2015), with two of them listed in Table 5. Besides, limited contents of PAHs have also been reported in sediments from Cold Lake in Alberta (85–200 ng/g) (Korosi et al., 2016) and Lakes in Athabasca (106–324 ng/g) (Jautzy et al., 2013). In addition to these, medium-low concentrations of PAHs can also be found at other sites, such as Strait of Georgia (core GVRD1: 91–573 ng/g) (Yunker et al., 2015), Baffin Bay (21–201 ng/g) (Foster et al., 2015), and the Ethel Lake (64–436 ng/g) (Korosi et al., 2013). However, extremely high levels of PAHs (6–487 μg/g) were recorded in Sydney harbor, a heavily industrialized region in Canada (Smith et al., 2009).

3.5. Contents of sedimentary recorded PAHs in other areas

Except that in China and Japan, contents of historical PAHs in other Asian countries are generally low, such as PAHs recorded in the Gulf of Thailand (GT15: 8–117 ng/g) (Boonyatumanond et al., 2007), Jakarta Bay in Indonesia (JKC5: 39–214 ng/g) (Rinawati et al., 2012), lagoons in Vietnam (Coastal lagoons: 59–465 ng/g and Nador Lagoon: 59–108 ng/g) (Giuliani et al., 2008, 2015), and Masan Bay in Korea (core C1: 38–277 ng/g and core C2: 88–756 ng/g) (Yim et al., 2005). PAHs recorded in sediment cores from Sundarban Wetland, India were in the range of 9–1144 ng/g, except that recorded in core S2 (1098–4250 ng/g) (Binelli et al., 2008). At the same studied area, though the sampling sites are not exact the same, gathered data in most cored sediments are in the range of 132–1100 ng/g

(Dominguez et al., 2010), comparable to the results reported by Binelli et al. (2008). Similar situation can be found in Southern India, where concentrations of PAHs in most sediments are within the range of 5–901 ng/g, except that in upper sediments from Cooum river (Goswami et al., 2016). Although the sampling sites are very close to each other, PAHs recorded in Chini Lake, Malaysia varied significantly (Bakhtiari et al., 2009). In Sri Lanka, similar to the historical PAHs recorded in Negombo Lagoon (378–1511 ng/g) (Ohura et al., 2015), PAHs contents measured in core B and core E from Chini Lake were in the range of 192–1895 ng/g and 212–1898 ng/g, respectively (Bakhtiari et al., 2009). This is clearly less than that in other two cores (core C: 570–6782 ng/g and core D: 325–4435 ng/g) and Kandy Lake (1393–3153 ng/g) (Ohura et al., 2015).

Comparable contents of historical PAHs have been measured at Cocó River Estuary (core T1: 3–2235 ng/g) and Ceará River Estuary (core T4: 3–1859 ng/g), Northeast Brazil (Cavalcante et al., 2009). They are much less than that recorded in Santos Estuary (core CQ1: 3723–6405 ng/g, core LSR: 1792–3019 ng/g, and core LCN: 852–7145 ng/g), a well-industrialized area in Southeast Brazil (Martins et al., 2011), and Barigui river in Curitiba (1766–14924 ng/g), a well-urbanized area in South Brazil (Machado et al., 2014). Moderate contents of PAHs (259–1176 ng/g) have been reported in cored sediments from Las Matas lagoon, Gulf of Mexico (Ruiz-Fernández et al., 2012). They are higher than PAHs recorded in Lake Santa Elena (119–363 ng/g) and Lake El Tule (50–853 ng/g), the remote lacustrine environment in the Jalisco Highlands, Central Mexico (Ruiz-Fernández et al., 2014), but are comparable with PAHs recorded in Laja lake in Chili (226–620 ng/g) (Quiroz et al., 2005) and Caspian Sea (202–736 ng/g) (Varnosfaderany et al., 2014). It is noteworthy that extremely low levels of PAHs (core B: 3–11 ng/g, core C: 5–20 ng/g, and core E: 0–35 ng/g) were reported in sediment cores from King George Island, Antarctica (Martins et al., 2010), probably representing the global background level of historical PAHs. It seems that contents of sedimentary recorded PAHs in other area are low to moderate level in most cases, expect that in Brazil.

3.6. *Comparison of contents of sedimentary recorded PAHs in China and the worldwide*

Based on the data gathered in Tables 1–6, contents of sedimentary recorded PAHs are compared within China (Fig. 1) and on the global scale (Fig. 2).

We divide all the Chinese works into six categories according to their geographical locations: open seas, estuaries, coastal bays, remote lakes in Tibet, other inland lakes, and urban lakes. As shown in Fig. 1 (Left), 90% of data of the remote lakes in Tibet are within the range of 170–460 ng/g and are relatively concentrated. As to other suites of data, they are dispersed significantly, especially for the data of other inland lakes. This is probably related to the wide geographic distribution of data sources. Almost all kinds of data are intertwined with each other. This makes

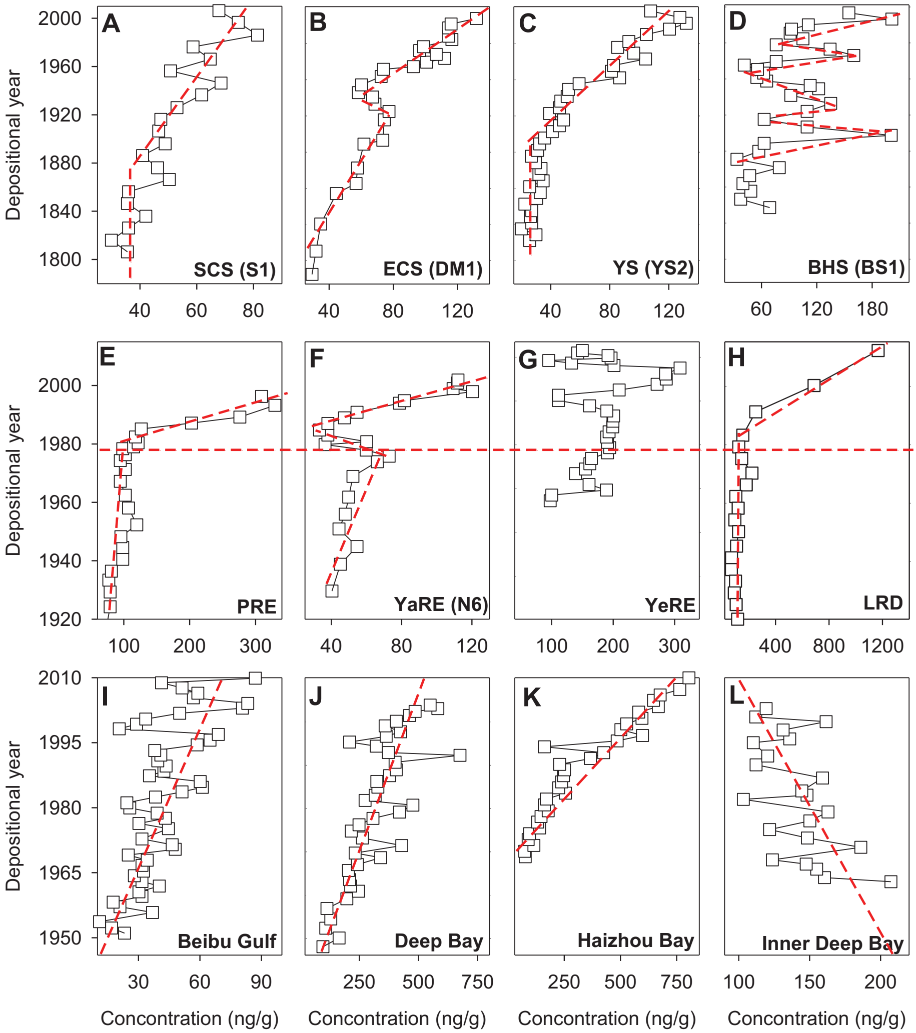

Figure 1. Comparison of sedimentary records of PAHs in different environments in China (left: each open cycle represents an individual figure; right: the boxed data presented in the left, the short lines represent the 10th, 25th, 50th, 75th, and 90th of the percentile values; open cycles represent the 5th and 95th of the percentile values).

difficult to conduct an accurate comparison. To simplify the comparison, we box all the data and the differences between different kinds of data that are revealed (Fig. 1, Right). Clearly, historical contamination of PAHs declined in the order of urban lakes > other inland lakes > remote lakes in Tibet > coastal bays > estuaries > open seas, with the median value of 1585 ng/g, 422 ng/g, 289 ng/g, 201 ng/g, 121 ng/g, and 55 ng/g, respectively.

Within China, contents of historical PAHs in open seas are at the lowest end when compared with that in other regions. Chinese open seas usually receive atmospheric input of PAHs from the adjacent inland, due to the dominant East Asian monsoon. Though atmospheric transport can carry significant amounts of

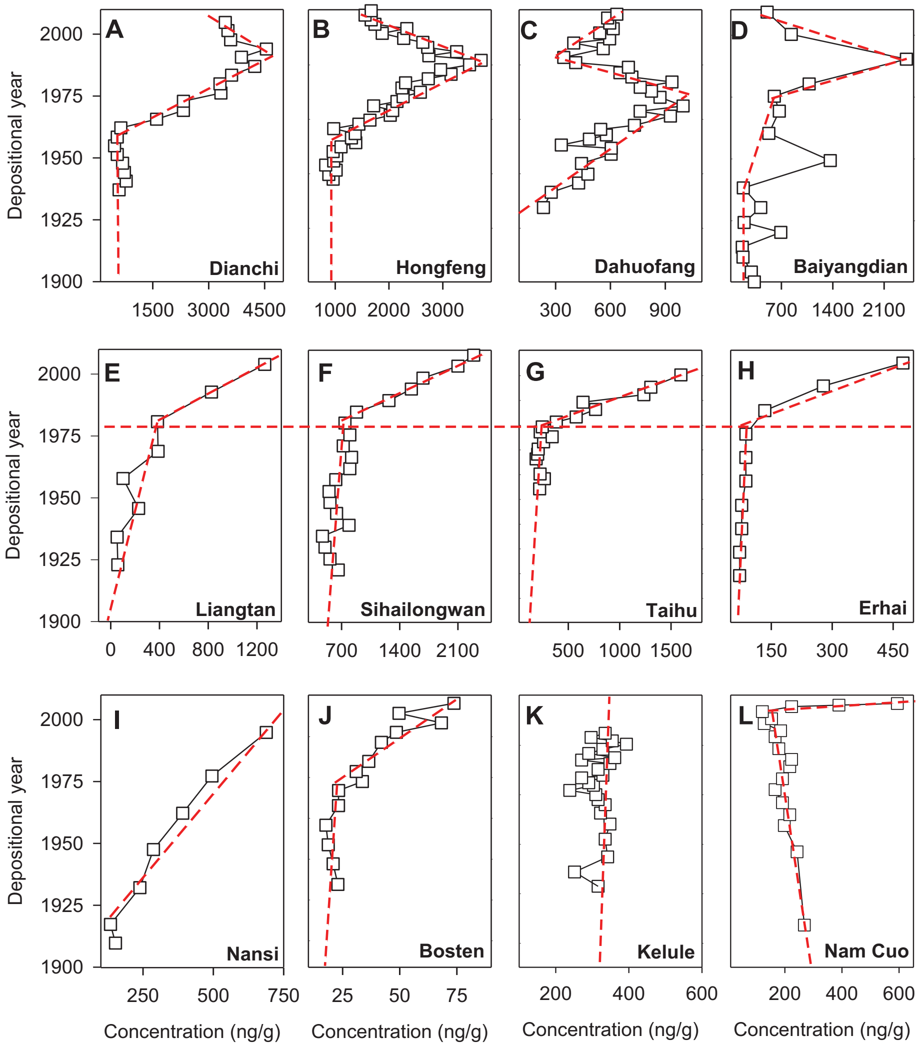

Figure 2. Comparison of sedimentary records of PAHs in China and the worldwide (short lines represent the 10th, 25th, 50th, 75th, and 90th of the percentile values; open cycles represent the 5th and 95th of the percentile values).

PAHs to remote locations, most combustion-derived PAHs are deposited close to their sources. Because the open seas are far from inland and the atmospheric transport potential of pyrogenic PAHs is limited, no wonder that the historical PAHs in Chinese open seas are very limited. In fact, historical PAHs along the Chinese coastline are also limited. They are not only less than that in urban lakes and other inland lakes, but also less than that in remote lakes in Tibet. Historically, China is a typical agricultural country and the human activities are dominated by agricultural activities, which mostly happened in inland area. This is probably the main reason for the low content of historical PAHs along the coastline. We have surmised that remote lakes in Tibet should be less polluted by PAHs due to their remoteness and less influence by anthropogenic activities. However, their median value of historical PAHs is much higher than that in Chinese coastline and open seas. This is beyond our imagination. This is probably related to their high altitude and receiving atmospheric deposition from long-range transport, as illustrated in PAHs recorded in lake sediments from the High Tatras (van Drooge et al., 2011).

On the global scale, we separated the collected data as far as possible according to the country or region and displayed in Fig. 2. Historical PAHs recorded in King George Island are extremely limited, with the median value of 5.4 ng/g. This is at least one order of magnitude less than that recorded in Chinese open seas, with no mention to other data. We note that the historical PAHs recorded in Vietnam (median: 176 ng/g) and India (median: 195 ng/g) are close to each other and similar to the historical PAHs recorded in Chinese coastal bays (median: 201 ng/g). However, data of India is much dispersed than that of Vietnam. Historical PAHs recorded in Canada (median: 202 ng/g) are very close to that in Chinese coastal bays. Nevertheless, data of Canada are highly dispersed, mainly ascribed to the high levels of PAHs recorded in Sydney harbor, a heavily industrialized region (Smith et al., 2009). More than 500 figures on historical PAHs have been collected all over the United States. They are highly varied with the median value (1456 ng/g) close to that recorded in Chinese urban lakes (median: 1585 ng/g). The variation of data from Europe and Japan is somewhat similar to each other, but their median values are totally different (2322 ng/g for Europe and 853 ng/g for Japan). Data on the historical PAHs in Brazil are basically from the industrialized and urbanized area, no wonder that it has the highest median value of 2544 ng/g among the gathered data. As to the historical PAHs recorded in other area, the median value (371 ng/g) is compared to that recorded in Chinese inland lakes (422 ng/g).

In brief, historical PAHs recorded in China are moderate at most, except PAHs recorded in urban lakes. Elevated levels of historical PAHs are usually found in traditional developed countries, such as the United States and Europe, as well as some cases in Japan and Canada. It is worthwhile pointing out that this conclusion is based on the historical data rather than the current data. In fact, situation has changed within the last several decades, because the newly developing countries have become the main emission sources of PAHs, especially China and India (Xu et al., 2006; Zhang and Tao, 2009).

4. Vertical profiles of PAHs in sediment cores

4.1. Vertical profiles of PAHs in China

Typical profiles of PAHs along the Chinese coastline are displayed in Fig. 3. In open seas, basic trends of PAHs recorded in SCS and YS are similar to each other within the last two centuries (Fig. 3a and 3c). This trend consists of two stages: a stable stage in the deeper sediments and a sustainable increasing stage in the upper sediments, though the turning points are not exactly the same (Liu et al., 2012a). The temporal trend of PAHs in ECS is composed of two increasing stages during

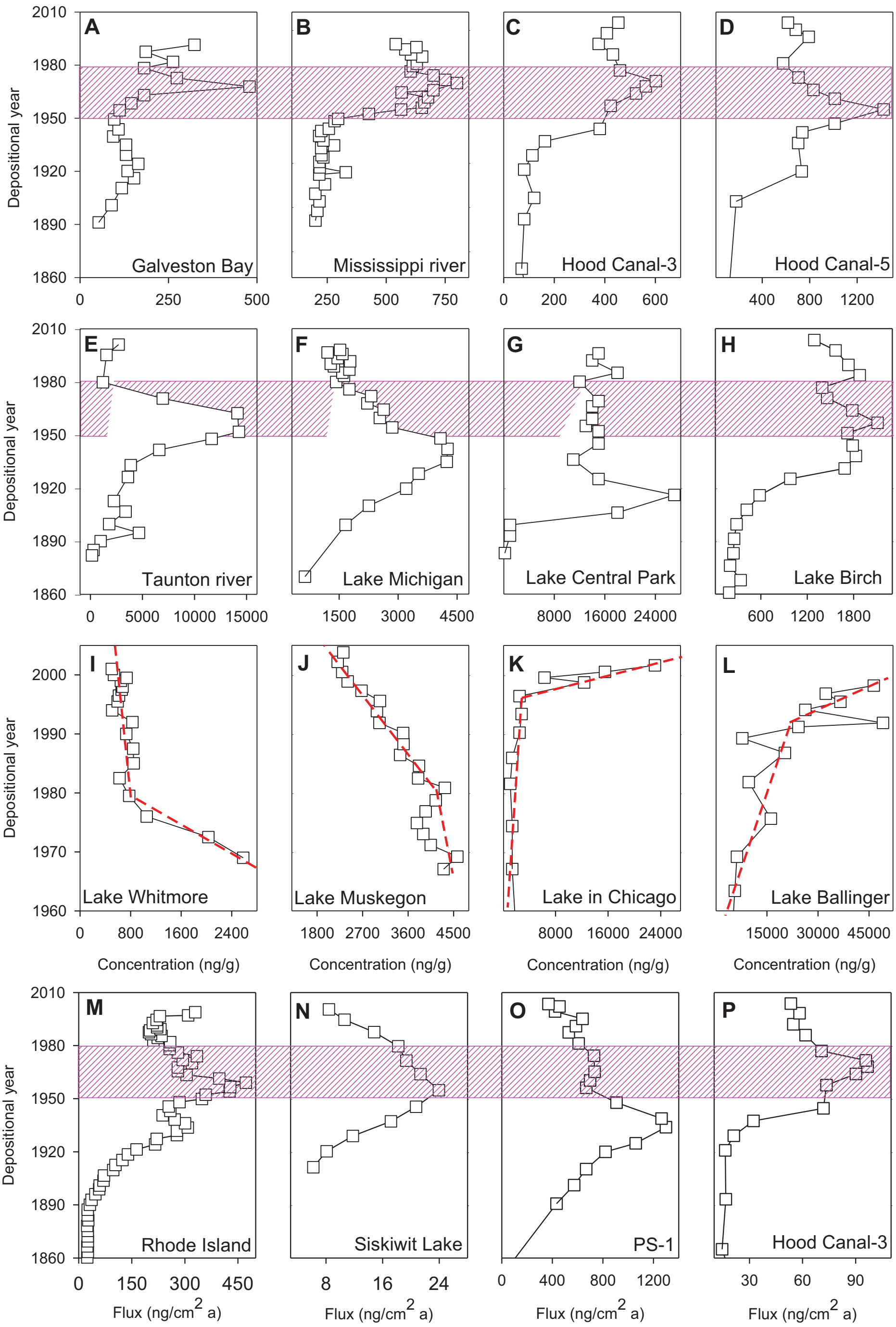

Figure 3. Sedimentary records of PAHs along the Chinese coastline (data sources: a (Liu et al., 2012a); b (Guo et al., 2006); c (Liu et al., 2012a); d (Hu et al., 2011); e (Liu et al., 2005); f (Guo et al., 2007); g (Yuan et al., 2014); h (Ma et al., 2016); i (Li et al., 2015); j (Qiu et al., 2009); k (Zhang et al., 2013); and l (Zhao et al., 2012)).

the period of 1800s to 1920s and 1940s to present, and a declining stage between 1920s and 1940s (Fig. 3b). As for the temporal trend of PAHs recorded in BHS, it increased upward in a zigzag manner with remarkable variation (Fig. 3d). Obviously, the upward increasing trends of PAHs in open seas are the same, but their increasing patterns are different from each other. This is probably a reflection of regional or local characteristics of historical inputs of PAHs.

Typical profiles of PAHs in the estuaries are illustrated in Fig. 3e–3h. Historical trend of PAHs recorded in PRE (Fig. 3e) is similar to that recorded in LRE (Fig. 3h), with a slow increasing stage from the beginning of the last century to the later 1970s and a sharp increase to the upmost sediments. The temporal trend of PAHs recorded in YaRE (Fig. 3f) is similar to that in ECS, composed of two increasing stages during the period of 1920s to 1970s and 1980s to present, and a decline between later1970s and early 1980s. However, they are not fully synchronized. Historical trend of PAHs recorded in YeRE experienced a sharp up and down variation in the upper sediments (Fig. 3g). It is completely out of sync when compared with that in PRE and YaRE. For example, during the period of 1985–1998, historical PAHs in PRE and YaRE have increased 160% and 260%, respectively. In the meantime, PAHs recorded in YeRE have decreased by 80%. After this brief decrease, PAHs recorded in YeRE rose again, leading to a maximum level in 2006. Afterward, concentrations of PAHs decreased abruptly again, returned back to values presented in the early 1960s, then rebounded to 197 ng/g in 2010 and declined to the upmost sediment. Yuan et al. (2014) attribute the unusual PAHs profile to the relocation of Yellow River and the efforts to mitigate the impacts of anthropogenic activities on the environment, but are short of direct evidences to support this.

The typical profiles of PAHs in the coastal bays are displayed in Fig. 3i–l. Temporal trend of PAHs recorded in HZB increased almost linearly from the bottom sediment to the surface sediment (within the last 50 years), with an increase rate of 17.7 ng/a and an unexpected low level at 1994 (Fig. 3k). The vertical profile of PAHs in DB resembles to that in HZB, but the increase rate (8.1 ng/a) is much less than that in HZB and the profile is much fluctuated along the core, with several peaks corresponding to ~1971, ~1981, and ~1992 (Fig. 3j). PAHs recorded in the BBG increased upward in a zigzag manner, with a limited increase rate of 1.0 ng/a (Fig. 3i). The vertical profile of PAHs recorded in LZB is different from that in HZB, DB, and BBG, characterized by a marked peak corresponding to 1986, and then it decreased to the upmost sediment (Ding et al., 2014). In most cases, the temporal trends of PAHs along the Chinese coastline increased from the deeper sediments to the upper sediments. However, opposite trend was found at Mai Po Inner Deep Bay, Hong Kong, where the concentration of PAHs decreased from the early 1960s to the present (Fig. 3l).

The typical temporal profiles of PAHs in Chinese inland lakes/reservoirs are illustrated in Fig. 4. In general, low and constant levels of PAHs are observed in deeper sections till the early 1960s in DC. Concentration of PAHs then starts to

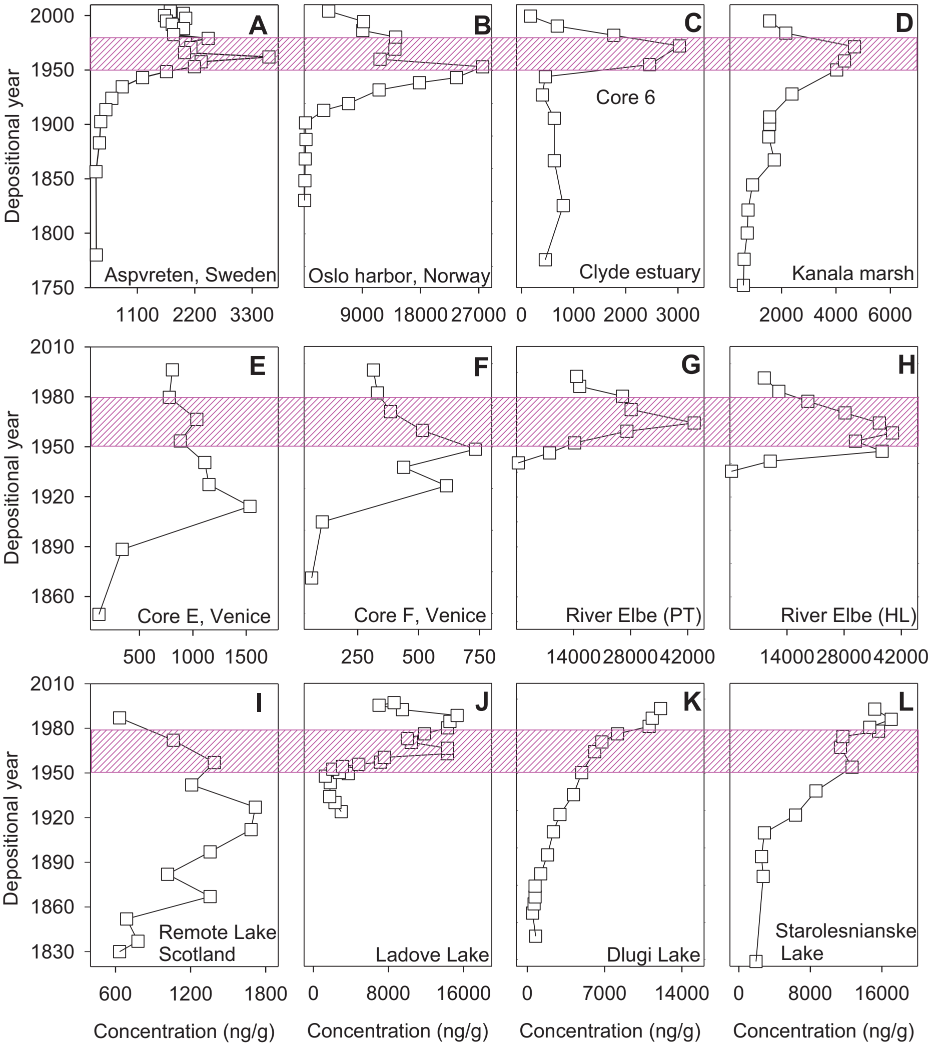

Figure 4. Sedimentary records of PAHs in Chinese inland lakes or reservoirs (data sources: a (Guo et al., 2013a); b (Li et al., 2013a); c (Lin et al., 2012); d (Guo et al., 2011c); e (Liu et al., 2012b); (Li et al., 2009b); f (Guan et al., 2012); g (Liu et al., 2009); h and j (Guo et al., 2010); i (Wang et al., 2010b); k and l (Yang et al., 2016)).

increase at a rate of ~122 ng/a and reach the maximum in 1994 before decreasing till the upmost sediment (Fig. 4a). The notable peak can also be found in other lakes, such as HF (Fig. 4b) and BYD (Fig. 4d), with a peak corresponding to approximately 1996 and 1990, respectively. Within the last century, vertical profile of PAHs in DHF increased successively from the benthic layers to form a plateau between early 1980s and early 1990s, followed by a decreasing stage during 1990–1996 and rebounding again at the subsurface sediments (Fig. 3c). It is interesting to find that vertical profiles of PAHs in LT (Fig. 3e) and SHLW (Fig. 3f) are similar to each other. Additionally, temporal trends of PAHs in TH (Fig. 3g) and EH (Fig. 3h) are also close to each other, though their geographic positions are far from each other. Roughly, historical PAHs in these sediment cores can be divided into two stages: a slow but stable increasing stage in deeper layers and a fast increasing stage in upper layers, with a turning point almost the same (corresponding to the initiation of "Reform and Open Policy" since 1978). However, the increasing rates are different in each core and each increasing stage. In fact, the fast increasing rate of PAHs after the initiation of "Reform and Open Policy" since 1978 not only occurred in lakes mentioned above, but also happened in other lakes, such as QH (Wang et al., 2010b), SG, and Bosten (Fig. 4j). As to the vertical profile of PAHs in NS (Fig. 4i), it increased linearly from the beginning of the last century and reached the maximum at the upmost sediment. It should be pointed out that the vertical profiles of PAHs recorded in remote lakes in Tibet are different from that in other inland lakes. Same to that in other remote lakes, increase in historical PAHs in Lake Keluke is very limited during the whole core (Fig. 4k). As to that in Nam Cuo, it even continuously decreased to the upmost sediments (Fig. 4l).

To sum up, the state of knowledge on the sedimentary record of PAHs in China has advanced significantly within the last two decades. Based on this information, we conclude that in China: 1) PAH contamination in aquatic environments is getting worse in most cases, 2) contamination of PAHs in inland lakes is usually much heavier than that in the coastal area, especially for urban lakes, 3) the basic upward trends are the same for most cases, but details in vertical profiles are different from each other, indicating that the sedimentary records of PAHs are usually site specific, and 4) sharp increase in PAHs levels in sediment mostly occurred from the early 1980s to the present, a period witnessing large changes in socioeconomic development in China.

However, when we finished reading these papers, the feeling was just like a stone in the shoe. When explaining the PAHs recorded in sediment cores, almost all the authors linked the temporal variation of PAHs to the important historical events that happened in China, such as the WWII, the Chinese Civil War, Great Cultural Revolution, and the implementation of the "Reform and Open Policy." The best examples of them are illustrated in the studies conducted at ECS and SCS (Guo et al., 2006; Liu et al., 2012a). Definitely, important historical events have great influence on anthropogenic activities and are crucial for the trend of PAHs recorded in sediment. However, they should not to be the omnipotent causes for the vertical profiles of PAHs recorded in China, because the historical PAHs are not fully synchronized on the national scale (Figs. 3 and 4). Historical PAHs must result from a variety of causes and there must exist other causes for PAHs loading in sediment.

4.2. *Vertical profiles of PAHs in USA*

Initial works in the United States indicated that PAHs in sediments generally increased from the end of nineteenth century and peaked in the mid to late 1950s, then declined thereafter (Gschwend and Hites 1981; Hites et al., 1977, 1980). This is partly supported by the work conducted at Milwaukee Harbor Estuary, where PAH profiles generally peaked in the 1950s through 1980s, then declined in most cases (Li et al., 1998). In 1970s, environmental policy in the United States changed markedly after the establishment of the US EPA (1969) and the passage of the Clean Air Act (1970), the Safe Drinking Water Act (1974), and the Toxic Substances Control Act (1976). It was assumed that this decline would continue, due to the substitution of coal with cleaner fuels and the shift in national policy toward stronger environmental protection. However, other works observed that PAHs in sediment no longer declined in some cases, and either stabilized or rebounded since ~1980s, due to the rapid urbanization and the newly emerged source of PAHs, such as PAHs from coal-tar sealant (Van Metre et al., 2000; Van Metre and Mahler 2005, 2010). Typical profiles of PAHs in the United States are illustrated in Fig. 5a–p.

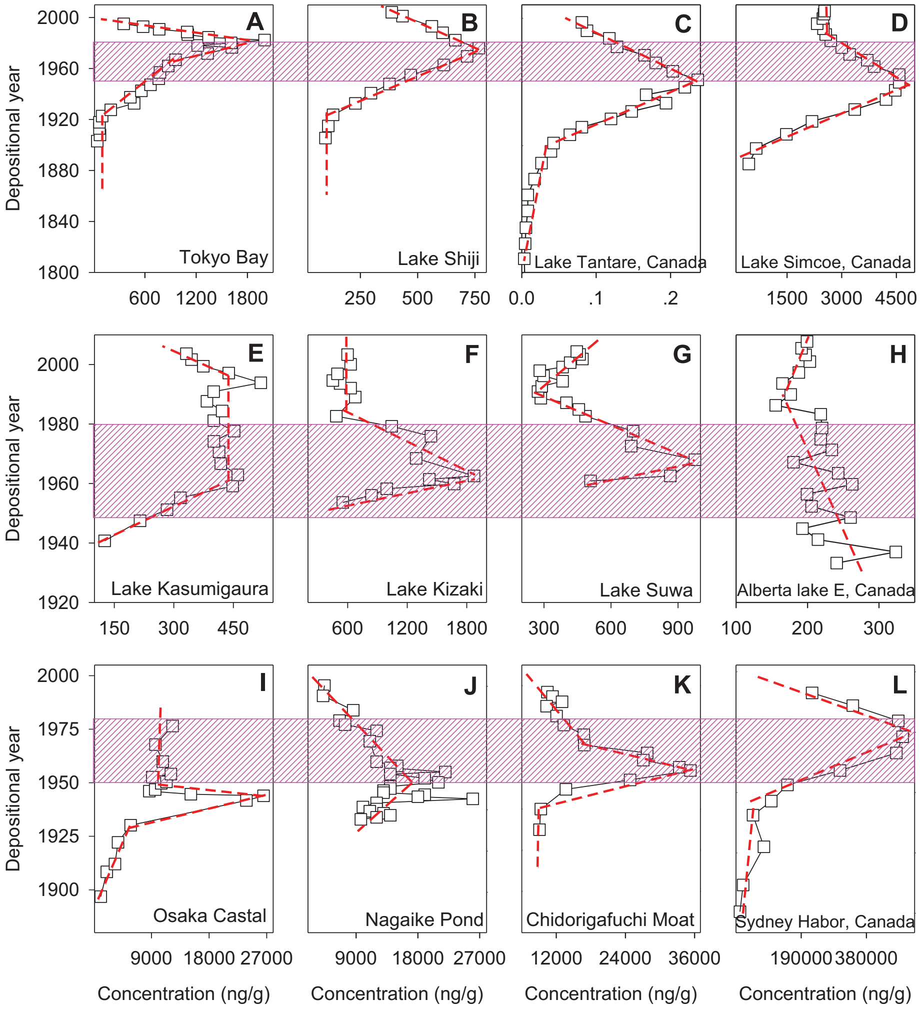

Figure 5. Sedimentary records of PAHs in the United States (data sources: a and b (Santschi et al., 2001); c and d (Louchouarn et al., 2012); e (Parsons et al., 2014); f (Cantwell et al., 2007); g (Schneider et al., 2001); h (Yan et al., 2005); i (Kannan et al., 2005); j (Parsons et al., 2014); k and l (Van Metre and Mahler, 2005); m (Hartmann et al., 2005); n (Slater et al., 2013); o and p (Kuo et al., 2011)).

Santschi et al. (2001) reported the historical PAHs along the US Gulf Coast. Historical PAHs in MRD are low and constant till the early 1940s, followed by a sharp increase resulting in a peak in the early 1970s, then decrease thereafter (Fig. 5b). PAHs recorded in Galveston Bay peaked in the late 1960s, then decreased abruptly to the late 1970s and rebounded at the upmost sediments (Fig. 5a). As to Tampa Bay, contents of PAHs are at least one order of magnitude higher than that in other two sites, with a peak in the 1980s followed by a substantial decrease to the upmost sediment. Although their sampling sites are close to each other, the temporal trends of PAHs are clearly different. Temporal trends of PAHs in hood canal peaked in the middle of 1950s (HC-5, Fig. 5d) and the early 1970s (HC-3, Fig. 5c), with ~15 years offset between two sites (Louchouarn et al., 2012). PAHs recorded in Taunton River are similar to that in HC-3, with a peak corresponding to the early 1960s and a sharp decrease during 1960s–1980s, then slight rebound at recent layers (Fig. 5e). Similar to that reported by Simcik et al. (1996), the profile of PAHs in Grand Traverse Bay, Lake Michigan peaked in the early 1940s, followed by a significant decline to 1980, then remained constant since then (Fig. 5f) (Schneider et al., 2001). PAHs in Lake Central Park started to increase at the initiation of the last century, reaching the maximum in the middle 1910s before decreasing from that point until 1920s, then remained constant till the upmost sediment (Fig. 4g). In Michigan, various types of profiles of PAHs are observed in inland lakes and one of them is shown in Fig. 4h. Figure 4i–l is probably the representatives of PAHs recorded in recent sediments that: 1) decreased till ca. 1980 and then remained relatively constant thereafter (Fig. 5i); 2) remained relatively constant till ca. 1980 and then successively decreased to the upmost sediment (Fig. 5j); and 3) abruptly rebounded at the very recent layers, such as that recorded in Lake in Chicago (Fig. 5k) and Lake Ballinger in north Seattle (Fig. 5l).

Depositional flux is considered to be more meaningful to assess the change in historical PAHs, because the sedimentation rate usually changed over time and concentration values varied as a function of sediment dilution (Fernandez et al., 1999; Lima et al., 2003). Consequently, vertical profiles of PAHs are also presented as depositional flux and some of them are displayed in Fig. 5m–p. In Siskiwit Lake, the profile of PAH fluxes can be divided into two stages within the analyzed sediments: 1) linearly increased from the deeper sediments to a maximum in 1955, and b) linearly decreased from that point to the upmost sediments (Fig. 5n). The quick increase in PyPAHs fluxes in HC-3 started around 1930s and reached a maximum in 1970 followed by a rapid decline in recent sediments (Fig. 5p). PyPAHs fluxes observed in Puget Sound also experienced a sharp increase after 1850s and a rapid decline after the maximum during 1930s–1940s (Fig. 5o). However, they are totally out of sync. High resolution of PAH fluxes recorded in Rhode Island probably elaborates the variation of PAH fluxes during the past 150 years in the US (Fig. 5m). Unfortunately, detailed historical PAH records are not easy to get and the reproducibility of the PAH fluxes recorded in Rhode Island needs further assessment with suitable temporal resolution at other sites.

In a word, vertical profiles of PAHs in the United States, whether expressed as concentration or flux, usually peaked during the period of 1950s–1980s or even earlier. After that, there have been three possibilities: 1) continue to decrease to the upmost sediments, such as PAHs recorded in Lake Birch, Muskegon and Siskiwit, 2) remain constant thereafter, such as PAHs recorded in Lake Michigan and Whitmore, and 3) rebound at the upmost sediments, such as PAHs recorded in Lake in Chicago and Lake Ballinger.

4.3. Vertical profiles of PAHs in Europe

Historical PAHs in Europe are similar to those in the United States, most of which peaked during the period of 1950s–1980s or even earlier (Arp et al., 2011; Götz et al., 2007; Van Zoest and Van Eck, 1993; Vane et al., 2011). However, PAHs ever increasing to the very recent sediments can also be found at individual sites (Punning et al., 2008; van Drooge et al., 2011). The typical temporal trends of PAHs in Europe are schematically shown in Fig. 6a–l.

Compared to the steady loading during the preindustrial age, PAHs varied dramatically within the last century in Oslo harbor, Norway (Arp et al., 2011). Historical PAHs in Oslo harbor started to increase at the beginning of the last century, reaching the maximum at the early 1950s before declining to the upmost sediment (Fig. 6a). Similar to that in Oslo harbor, PAHs recorded in Aspvreten, Sweden also experienced a quick increasing stage before they peaked at the early 1960s, but remained relatively constant during the last several decades (Fig. 6b). Historical PAHs recorded in Clyde estuary, Scotland were relatively constant till the later 1940s, but were sharply up and down during the second half of the last century, with the maximum value in 1972 (Fig. 6c). This is quite different from PAHs deposited in remote lake in Scotland (Rose and Rippey, 2002), where PAHs peaked about 1930s (Fig. 6i). Rapid rise and fall in historical PAHs can also be found in River Elbe, Germany, with the maximum value corresponding to before or immediately after 1960s (Fig. 6g and 6h), which is compared with that recorded in Scheldt estuary, Netherland (Van Zoest and Van Eck, 1993). The vertical profiles of PAHs in Venice, Italy are somewhat similar to those in River Elbe, but peak much earlier (Fig. 6e and 6f).

PAHs deposited in the High Tatras are different from historical PAHs mentioned above, characterized by an ever increase to the upmost sediments as exhibited in Lake Dlugi (Fig. 6k) and Starolesnianske (Fig. 6l), or a later increasing stage as illustrated in Lake Ladove (Fig. 6j). The ever increase in PAHs to the upmost sediments can also be found in Lake Peipsi (Punning et al., 2008). In addition, reported data associated with historical PAHs in Kanala marsh, Spain can retrospect to 1300s (Leorri et al., 2014) and only about 250 years' record is presented in Fig. 6d, exhibiting a slow increase before 1900s and a drastic change within the last century. PAH deposition at Holzmaar (Eifel, Germany) reported by Musa Bandowe et al. (2014) is the longest record of PAHs we ever found. It provides us a

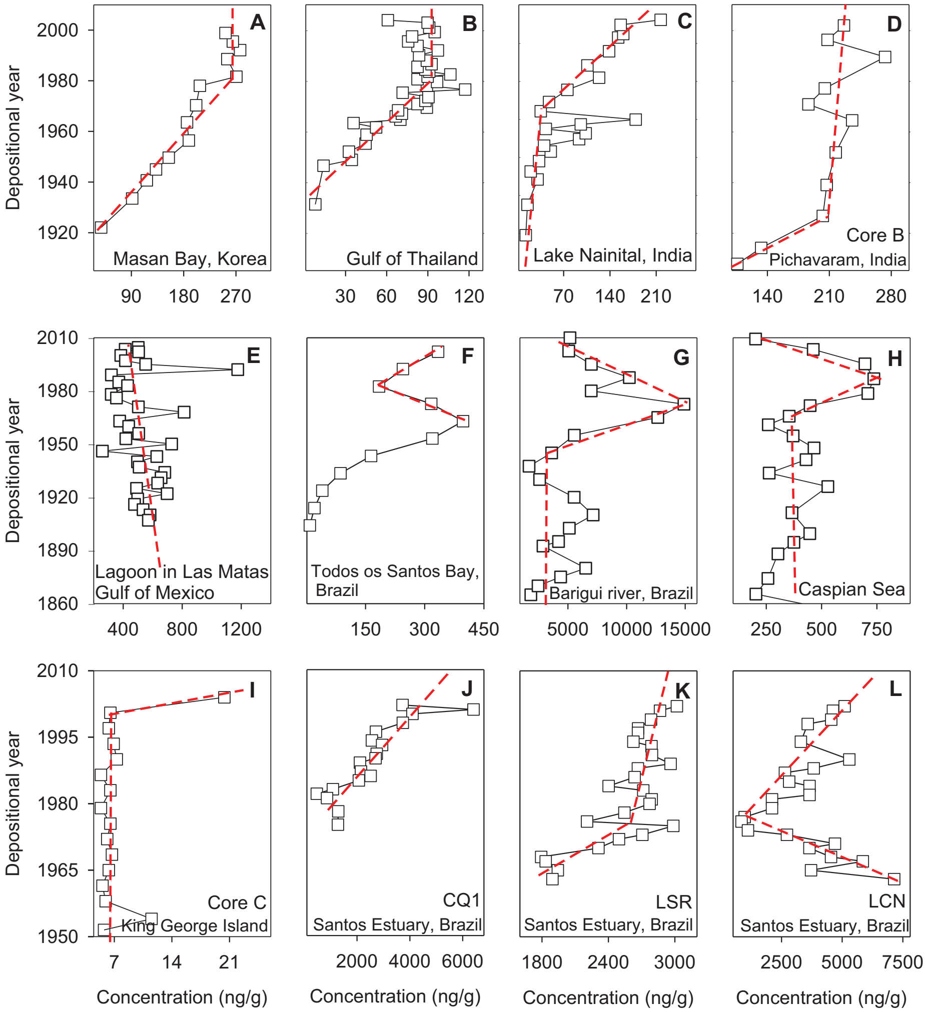

Figure 6. Sedimentary records of PAHs in Europe. (All the cores are from fresh water, except that in b and c. Data sources: a (Elmquist et al., 2007); b (Arp et al., 2011); c (Vane et al., 2011); d (Leorri et al., 2014); e and f (Frignani et al., 2003); g and h (Götz et al., 2007); i (Rose and Rippey, 2002); j, k, and l (van Drooge et al., 2011)).

long-term perspective to understand how serious the impacts of human activities on the environments are over the past two centuries.

4.4. Vertical profiles of PAHs in Japan and Canada

Similar to that in the United States and Europe, Historical PAHs in Japan and Canada are mostly peaked during the period of 1950s–1980s or slightly earlier (Fig. 7a–l). It seems that peaking in the period of 1950s–1980s is the common feature for the vertical profiles of PAHs in traditional developed countries. Collected data on the vertical profiles of PAHs in Japan rarely extend backward beyond

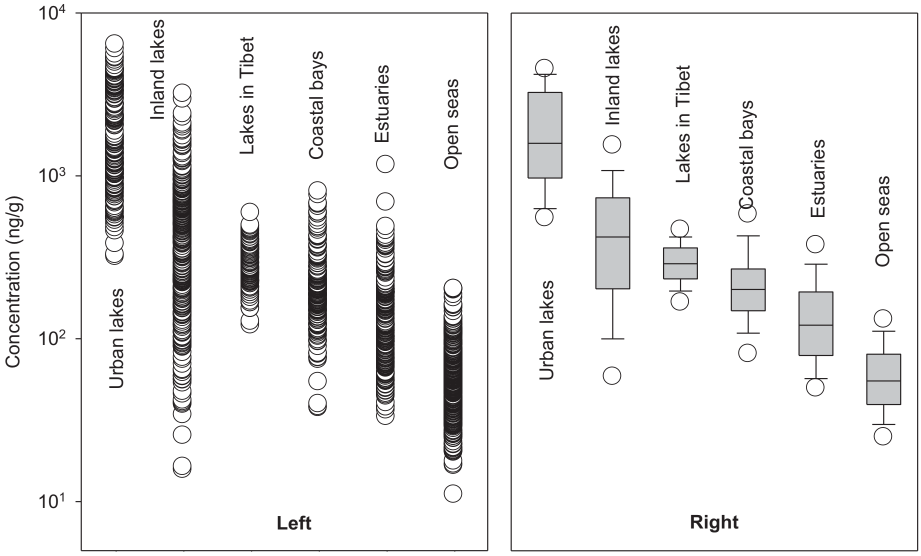

Figure 7. Sedimentary records of PAHs in Japan and Canada. (All the cores are from fresh water, except that in a and l. Data sources: a (Yamashita et al., 2000); b, e, f, and g (Fukushima et al., 2012); c (unit: PAH_{13} nmol cm^{-2} y^{-1}, (Gallon et al., 2005)); d (Helm et al., 2011); h (Jautzy et al., 2013); i (Moriwaki et al., 2005); j (Ishitake et al., 2007); k (Okuda et al., 2002); l (Smith et al., 2009)).

1900. In most cases, historical PAHs in Japan declined immediately after reaching the peak values, except Lake Kasumigaura, where the historical PAHs maintain a platform for more than 30 years (Fig. 7e). Historical PAHs in Japan within the last decades usually continue to decrease till the upmost sediments, such as PAHs recorded in Tokyo Bay (Fig. 7a), Lake Shiji (Fig. 7b), Lake Kasumigaura (Fig. 7e), and Nagaike Pond (Fig. 7j). However, there exists other situation, including remaining constant at the upper layers (Fig. 7f and 7i) or rebounding at the very recent sediments (Fig. 7g).

In Canada, sedimentary record of PAHs in Sydney Harbor (Fig. 7l) resembles that in Lake Tantaré (Fig. 7c), although not fully synchronized. The temporal trends of PAHs in both cores are composed of a slow increasing stage in benthic layers and a fast increasing stage in middle layers, followed by a linear decrease to the upmost sediment. Though there is a lack of preindustrial data, vertical profile of PAHs in Lake Simcoe is akin to that in Lake Tantaré, but remains constant in the very recent sediments (Fig. 7d). Sedimentary PAHs in cold lake in Alberta peaked at ~1915, earlier than the regular peaking time, and then decreased continuously to the upmost sediment (Korosi et al., 2016). This pattern is supported by the work conducted at Alberta lakes (Jautzy et al., 2013), where the contents of PAHs either successively declined to the surface sediment (lake D) or rebounded at the very recent sediments (lake E, Fig. 7h). Other PAH patterns are reported very recently, such as the works conducted at the Hilda and Ethel Lake (Korosi et al., 2013), Hecate Strait (Yunker et al., 2014), Strait of Georgia (Yunker et al., 2015), and Saskatchewan lakes (Ahad et al., 2015). Due to lack of accurate dating data or enough PAH data to develop a clear vertical profile, they were merely listed in Table 5 and not schematically shown in Fig. 7.

In brief, PAH emissions in traditional developed countries have passed its maximum value and have declined significantly, based on the PAHs recorded in sediments. However, new sources of PAHs could emerge, for sedimentary PAHs rebounded very recently in some cases (Jautzy et al., 2013; Van Metre and Mahler, 2005, 2010).

4.5. Vertical profiles of PAHs in other areas

Although many works on the sedimentary record of PAHs are available in other areas, most of them lack dating data. Therefore, sediment cores will be arranged in Fig. 8 provided that the sediment core is dated. Historical PAHs recorded in Masan Bay, Korea (Fig. 8a) and the Gulf of Thailand (Fig. 8b) are similar to each other, both showing an increasing trend in the deeper sediments and remaining relatively constant in the upper sediments, with a turning point about 1980s. Sedimentary PAHs in Lake Nainital, northwest India have shown a slow increasing stage in bottom sediments and a fast increasing stage in upper sediments, with the turning point corresponding to the early 1970s (Fig. 8c). During this period, an unexpected high level of PAHs corresponding to the middle 1960s is observed, probably a reflection of the forest fire happening there (Choudhary and Routh, 2010). However, PAHs in sediment core from Pichavaram mangrove, southeast India consist of a rapid increasing stage in bottom sediments and a slow increasing stage in upper sediments with the turning point corresponding to the middle 1920s (Fig. 8d). As to PAHs recorded in Las Matas, Gulf of Mexico, though frustrated

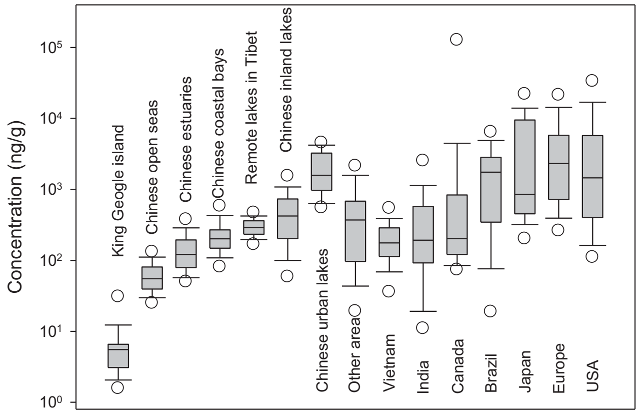

Figure 8. Sedimentary records of PAHs in other area. (All the cores are from marine water, except that in c, d, and g. Data sources: a (Yim et al., 2005); b (Choudhary and Routh, 2010); c (unit: μg cm^{-2} y^{-1}, (Ranjan et al., 2012)); d (Boonyatumanond et al., 2007); e (Ruiz-Fernández et al., 2012); f (Costa et al., 2016); g (Machado et al., 2014); h (Varnosfaderany et al., 2014); i (Martins et al., 2010); j, k, and l (Martins et al., 2011)).

along the whole core, the basic declining trend is apparent (Fig. 8e). PAHs deposited in Santos Bay, East Brazil increased successively from the beginning of the last century to reach the maximum at the early 1960s, then declined to the early 1980s and rebounded thereafter (Fig. 8f). Historical PAHs recorded in Barigui river, Brazil (Fig. 8g) are similar to those in Caspian Sea (Fig. 8h), both exhibiting a dramatic variation during the second half of the last century. Sediment cores (CQ1, LSR, and LCN) from Santos Estuary, Southeast Brazil have dating data within the last 50 years (Fig. 8j–l). Although the sampling sites are close to each other, different patterns of PAHs were presented. For the historical trend of PAHs in Admiralty Bay, King George Island (core C), it is low and constant in benthic sediments till approximate 2000, but shows a considerable increase in most recent sediments (Fig. 8i).

4.6. Comparison of the vertical profiles of PAHs in China and the worldwide

Basic features of the vertical profiles of PAHs in China include: 1) contamination of PAHs in aquatic environments are getting worse in most cases, with a sharp increase in PAHs levels that mostly occurred from the early 1980s to the present, 2) basic upward trends are the same for most cases, but details in vertical profiles are different from each other, and 3) in some urban lakes, declining trends in PAHs profile are found at very recent sediments. The first one is probably a reflection of the sustained socioeconomic development and the continuous increase in energy consumption within the period of time covered by sediments, and a particular period witnessing a sharp increase in socioeconomic development in China since 1980s (Fig. 9a and 9b). The second one indicates that historical PAHs in China are usually a reflection of local or regional sources, which cannot be fully explained by the nationwide socioeconomic data. The recent declining trends of PAHs in some urban lakes do not mean that PAH emission has declined in China (Xu et al., 2006). In fact, historical PAHs result from a variety of causes. Urban lakes in China are usually heavily contaminated by PAHs for their specific geographical location, such as DC and HF. The heavy PAH contamination in DC mainly attributes to the huge amount of pollutants inputs via riverine and municipal runoff from the adjacent cities (Guo et al., 2013a). During the last two decades, great efforts have been made to improve the water quality in DC and the contaminants loading to DC have substantially decreased, including PAHs. This is probably the main reason for the recent decline in historical PAHs in DC (after 1994). The same holds true for HF, an urban lake adjacent to Guiyang city, Southwest China (Li et al., 2013a).

By and large, historical PAHs in traditional developed countries have passed its maximum and have declined for many years, although new sources of PAHs, resulting in the rebound of sedimentary PAHs in some cases, cannot be excluded. This is different from that in China. For example, historical PAHs in the United States usually peak in the period of 1950s–1980s, then continue to decrease or

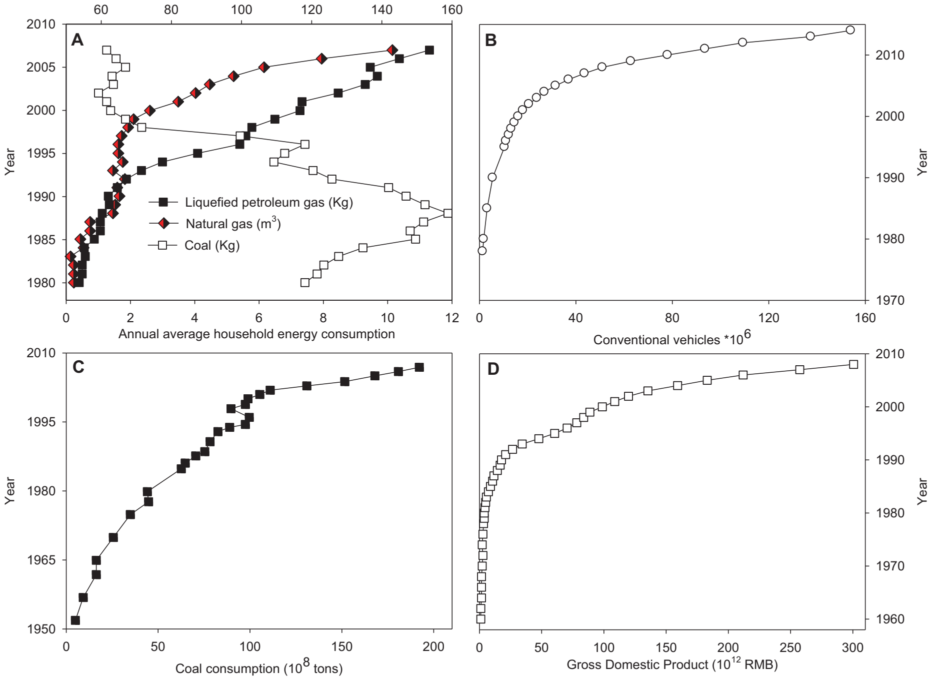

Figure 9. Annual average household energy consumption (a) and conventional vehicles (b) in China (http://www.stats.gov.cn).

remain constant thereafter. As to the historical PAHs in Europe, Japan, and Canada, they are comparable with that in the United States, most of which peaked within the period of 1950s–1980s and declined thereafter in most cases. It seems that peaking in the period of 1950s–1980s is the common feature for the vertical profiles of PAHs in traditional developed countries. For other area, there is no enough data to provide a precise insight on the vertical profile of PAHs. However, a clear PAH peak corresponding to ~1970s is recorded in sediment from Barigui River (Machado et al., 2014), probably a reflection of "Economic Miracle" between 1960 and 1980 that happened in Brazil.

5. Source identification and apportionment

PAHs introduced to the modern environment are mostly of anthropogenic origin. Potential candidates include biomass burning, wood and coal combustion, vehicle exhaust emission, coke and aluminum production, accidental oil spillage, municipal waste incineration and sewage discharge, and domestic activities. Source identification and apportionment is crucial for risk assessment and the development of control strategies. For PAHs, many methods have been developed for source identification and apportionment, such as source markers (Ravindra et al., 2008), parent PAHs (Zemo, 2009), diagnostic ratios (DR) (Yunker et al., 2002), receptor models (Rachdawong et al., 1998; Zhang et al., 2012), and compound-specific carbon isotope analysis (CSCIA) (O'Malley et al., 1996). Due to the complexity of PAH sources, source identification and apportionment of PAHs in sediment is not an easy work and is still a big challenge.

5.1. Method for source identification and apportionment

5.1.1. Source markers

Source markers, also named as tracers, indicators, or signatures, are usually the specific PAH compounds or non-PAH compounds. One of the basis of source identification using source markers is the difference in PAH profiles for certain processes associated with the release of PAHs into the environment. For example, dominance of CHRY and BkF is usually an indication of PAH origins from coal combustion, while high abundance of BghiP, COR, and PHEN is characteristic of vehicle emission, and PAHs from incineration are predominated by PYR, FLUO, and PHEN (Smith and Harrison, 1998). Another basis is assumed that specific PAH source categories have distinct/certain molecular tracers, such as retene (1-methyl-7-isopropylphenanthrene) for wood combustion (Ramdahl, 1983), hopanes for vehicle emission, picene for coal combustion, and levoglucosan for biomass burning (Gao et al., 2012). However, the effectiveness of source markers is limited in source identification, because the source information of PAHs may have changed from the emission source to the sink, and multiple sources are mixed in sediment resulting in the concealment of original source information. Now it is rarely used alone, but usually in combination with other methods for source assignment.

5.1.2. DR

DR is probably the most frequently used method for source identification of PAHs. Beginning with the work of Daisey et al. (1979), numerous works have used DRs to infer the sources of PAHs in environment. The greatest advantage of this method is the simplicity and ease of application. This is of particular appeal to the developing countries where there is no enough data to run complicated models or no extensive measurements to determine the relevant source characteristics. The frequently used DRs include 1,7-DMP/(1,7-DMP + 2,6-DMP), ANT/(ANT + PHEN), BaA/(BaA + CHRY), FLUO/(FLUO + PYR), INP/(INP + BghiP), BaP/(BaP + BeP), BaP/BghiP, $\sum$LMW/$\sum$HMW, BbF/BkF, and RET/(RET + CHRY). For example, ANT/(ANT + PHEN) is often used to distinguish petroleum source from combustion sources and FLUO/(FLUO + PYR) is widely used to discriminate petroleum combustion from other types of combustion (Yunker et al., 2002). 1,7-DMP/(1,7-DMP + 2,6-DMP) is considered to be a good indicator of wood combustion (Benner et al., 1995) and a marker of wood forest burning (Gustafsson et al., 1997), while INP/(INP + BghiP) is believed to have the potential to distinguish petroleum source from pyrogenic sources (Yunker et al., 2002). Most DRs and their source assignments are listed in Table 7.

Table 7. Diagnostic ratios for PAHs source identification.

PAH ratio	Value range	Source	Reference
∑LMW/∑HMW	<1	Combustion	(Yan et al., 2005)
	>1	Petroleum	(Yan et al., 2005)
ANT/ANT + PHEN	<0.1	Petroleum	(Budzinski et al., 1997)
	>0.1	Combustion	(Budzinski et al., 1997)
FLUO/FLUO + PYR	<0.4	Petroleum	(Yunker et al., 2002)
	0.4–0.5	Liquid fossil fuel combustion	(Yunker et al., 2002)
	>0.5	Grass, wood or coal combustion	(Yunker et al., 2002)
BaA/BaA + CHRY	<0.2	Petroleum	(Yunker et al., 2002)
	0.2–0.35	petroleum or coal combustion	(Yunker et al., 2002)
	>0.35	Coal combustion	(Yunker et al., 2002)
INP/INP + BghiP	<0.2	Petroleum	(Yunker et al., 2002)
	0.2–0.5	Liquid fossil fuel combustion	(Yunker et al., 2002)
	>0.5	Glass, wood or coal combustion	(Yunker et al., 2002)
1,7-DMP/(1,7-DMP + 2,6-DMP)	≤0.45	Petroleum burning	(Benner et al., 1995)
	0.65–0.68	Coal combustion	(Yan et al., 2014)
	0.70–0.90	Softwood combustion	(Benner et al., 1995)
RET/RET + CHRY	0.15–0.50	Petroleum burning	(Yan et al., 2005)
	0.30–0.45	Coal combustion	(Yan et al., 2005)
	0.83–0.96	Softwood combustion	(Yan et al., 2005)
BaP/BaP + BeP	~0.5	Fresh particles	(Tobiszewski and Namieśnik, 2012)
	<0.5	Aged particles	(Tobiszewski and Namieśnik, 2012)
BaP/BghiP	<0.6	Non-traffic emission	(Brändli et al., 2007)
	>0.6	Traffic emission	(Brändli et al., 2007)
BbF/BkF	2.5–2.9	Aluminum smelter emissions	(Tobiszewski and Namieśnik, 2012)
∑MPHEN/PHEN	<1	Combustion	(Callén et al., 2011)
	>1	Petroleum	(Callén et al., 2011)
U/R[a]	>4.0	Petrogenic	(Oros and Simoneit, 2000)
	>6.0	Petroleum burning	(Oros and Simoneit, 2000)
	2.9–3.2	Coal combustion	(Oros and Simoneit, 2000)
	0.5–1.4	Softwood combustion	(Oros and Simoneit, 2000)

[a] the ratio of unresolved complex mixture (UCM) to resolved hydrocarbons in saturated fraction.

However, the validity of this method rests on many assumptions (Watson, 1984). The most important assumptions are: 1) DRs of each suspected source or source type are assumed to be unique and different from each other, and 2) DRs of each source or source type are assumed to be strictly conserved between the emission source and the sink. However, both of them are problematic or questionable. Katsoyiannis et al. (2007) have challenged the validity of DRs for PAH source identification in sewage sludge. Further assessment on the validity of DRs has been elaborated in other articles (Galarneau, 2008; Tobiszewski and Namieśnik, 2012). Collected evidences suggest that DRs not only show substantial intrasource variability and intersource similarity (Galarneau, 2008), but also have visible change between the source and the sink (Tobiszewski and Namieśnik, 2012). For example, ratio of BaA/(BaA + CHRY) of different types of coal combustion (0.18–0.49) is seriously overlapped with that of oil combustion (0.12–0.44) (Yunker et al., 2002), and DRs values may change during phase transfers and environmental degradation (Tobiszewski and Namieśnik, 2012). This poses a big challenge to source assignment and obvious misleading could happen. In fact, the critical value for each DR is actually a statistical result, rather than the true values of specific PAH sources (Yunker et al., 2002). Thus, DRs should be used with caution. To be on the safe side, more than one DR and/or other source markers are recommended to confirm the results.

5.1.3. Receptor models

Receptor model is another frequently used method for source identification and apportionment of PAHs. Fundamentals of the receptor model have been described elsewhere (Gordon, 1988; Henry et al., 1984; Hopke, 1991). Receptor model was developed initially for source apportionment of atmospheric PAHs and is now also applied to sedimentary PAHs. It can be divided into two categories: microscopic and chemical (Pant and Harrison, 2012). The former is based on the morphological analysis (Cooper and Watson, 1980) and is less used recently. The latter is based on the chemicals of interest. Available receptor models of this category include chemical mass balance (CMB) model, factor analysis (FA), and improved FA (include principal component analysis (PCA), positive matrix factorization (PMF), and Unmix).

1. *CMB model* As a tool for source apportionment, CMB was first proposed for aerosols in the early 1970s (Friedlander, 1973). The basic idea of CMB is that the marked chemicals at a specific receptor are linearly combined with the chemicals emitted from independent sources (Christensen et al., 1997). The key to a successful CMB application is to obtain a set of source fingerprints consistent with the measurements at the receptor. To obtain a quantitative source contribution, several assumptions, including the consistence of emission source, the intact of marked chemicals, the uniqueness of sources fingerprints, and the independence of each contributing sources, need to be satisfied (Li et al., 2003). The method has been used for the source apportionment of PAHs in dated sediments from Green Bay, Wisconsin (Su et al., 1998), PAHs in Lake Calumet, Chicago (Li et al., 2003), and PAHs in dated sediments from the Black River, Ohio (Gu et al., 2003), and the performance sounds reasonable.

   However, inherent limitations of CMB are inevitable. Firstly, running CMB requires a *priori* knowledge of the fingerprints of all sources, which would be an arduous task for source apportionment of sedimentary PAHs, due to lack of source information of historical PAHs. Secondly, definitive signature of a specific combustion process may not exist, due to the complexity of the combustion process (Li et al., 2003). Thirdly, the predetermined assumptions cannot be strictly obeyed, due to the phase transfer, long-range transport, and environmental degradation of PAHs during the modeling process. Besides, difference in sampling method may introduce additional difference in source signatures.
2. *FA* Compared to CMB, FA can operate with minimal knowledge about source information, meteorological conditions, and details in various environmental processes. The basic idea and assumptions of FA have been elaborated elsewhere (Imamoglu, 2001; Rachdawong et al., 1998). Now, it has been developed into three typical models: PCA, PMF, and Unmix. PMF was introduced in the early 1990s and the details are descripted by Paatero and his co-author (Paatero, 1997; Paatero and Tapper, 1993). Unmix, based

on the self-modeling curve resolution technique (Hopke, 2003), is a new FA receptor model for source apportionment of PAHs recommended by US EPA. Detailed information on Unmix 6.0 model can be found elsewhere (EPA, 2007). PMF has been applied to source apportionment of PAHs in sediments (Sofowote et al., 2008; Zhang et al., 2012), and very recently, Unmix has also been used for source apportionment of PAHs in dated sediments from western China (Xu et al., 2014).

PCA is one of the most widely used FA for source apportionment. The primary function of PCA is to reduce the number of variables while retain the original information as much as possible. It changes the original set of variables into a smaller one accounting for most of the variance of the original set. Variables and characteristics are grouped into factors, which can be interpreted as the emission sources. Generally, each factor extracted from PCA is associated with a source characterized by its representative chemicals. In most cases, mathematical and statistical software are used for source grouping by using PCA with varimax rotation and the retention of principal components with eigenvalue greater than 1. The principal components are used as factors. Loading determines the most representative PAH species in each factor and generally a value greater than 0.5 is selected. This method has been used to apportion the dated PAHs in Black River and Ashtabula River, Ohio (Christensen and Bzdusek, 2005).

Though statistical methods based on FA do not need a comprehensive knowledge of PAH sources (Pant and Harrison, 2012), assignment of factor to specific source category may be questionable due to the disturbance from extreme data and genuine collinearity of diverse sources (Larsen and Baker, 2003; Pant and Harrison, 2012). For a better source assignment, it is helpful to remove outliers using hierarchical cluster analysis before PCA or PMF (Liu et al., 2015). In addition, to obtain accurate results, enough samples are needed for statistical FA analysis (Thurston and Spengler, 1985). Again, detailed information on the emission sources is helpful to discriminating between similar sources.

5.1.4. Compound-specific carbon isotope analysis (CSCIA)

Based on the technology of gas chromatography isotope ratio mass spectrometry, compound- specific stable isotope analysis has become a well-developed analytical method during the last several decades (Schmidt et al., 2004). It has many environmental applications and is in particular true for carbon isotope analysis. CSCIA was firstly introduced for source apportionment of PAHs in sediment by O’Malley (1994), based on the differences in carbon isotope between petrogenic and pyrogenic sources (O’Malley, 1994; O’Malley, et al., 1996). The typical δ^{13}C value of parent PAHs (like FLUO and PYR) from petrogenic sources is around −29 to −27‰, whereas pyrogenic compounds have relatively higher δ^{13}C value of −22 to −26‰ (O’Malley, 1994). This method has been used for source apportionment of PAHs in chidorigafuchi Moat in Japan (Okuda et al., 2002), the New York/New

Jersey harbor complex (Yan et al., 2006), and Alberta Lakes in Canada (Jautzy et al., 2013).

The advantage of CSCIA is that different petrogenic sources, such as coal burning, internal engine, and biomass combustion, can lead to distinct δ^{13}C values (Ballentine et al., 1996; McRae et al., 1998), and δ^{13}C signature of high molecular weight PAHs is not significantly altered by photodegradation (O'Malley, 1994; Okuda et al., 2002) and biodegradation (Mazeas et al., 2002; Morasch et al., 2002). This provides a valuable tool to delineate the sources of PAHs in sediment (O'Malley et al., 1996). For example, PAHs obtained from fluidized-bed coal combustion particulates have a small δ^{13}C variation from -25 to -31%, and larger PAHs are more depleted in ^{13}C. Isotopic fingerprints of PAHs derived from diverse sources may aid source identification of PAHs in environment (McRae et al., 1996, 1998, 1999). The disadvantage of CSCIA is the difficulty in data acquisition, both in sample preparation and instrumental analysis.

In addition to δ^{13}C, radiocarbon (Δ^{14}C) also has the potential to differentiate different pyrogenic PAHs, because the radiocarbon contents are totally different among PAHs' different pyrogenic sources. In general, PAHs from fossil fuel combustion contain no significant radiocarbon ($\Delta^{14}C = -1000$‰) while PAHs from biomass burning contain modern levels of radiocarbon ($\Delta^{14}C = \sim 50$‰) (Kanke et al., 2004; Reddy et al., 2002). Using an isotopic mass balance approach, relative contribution of these sources to PAHs in sediment can be apportioned. This approach has been applied to gain insight into the origin of perylene (Slater et al., 2013).

5.2. Sources and their changes of historical PAHs in China

Though knowledge on historical PAHs in China has been advanced significantly within the last two decades, most studies reported the concentrations and/or fluxes without further source identification and apportionment analysis. When sorting out the source information on historical PAHs in China, we found that the available information is mostly qualitative, except one attempt for PAHs in lake sediments from West China (Xu et al., 2014). As for the historical change in PAH sources, a source change from low- and moderate-temperature combustion to high temperature combustion is usually reported (Cai et al., 2016; Guo et al., 2013a; Li et al., 2013a). However, detailed information on the historical sources remains limited. There is still a large information gap on the source identification and apportionment of historical PAHs in China.

DRs and PCA are the most frequently used methods for source identification of historical PAHs in China. Though there are slight differences from others, most works point to the fact that historical PAHs in China are mainly of pyrogenic origin within the period of time covered by sediments, most of which originated from coal combustion and/or biomass burning. This is well consistent with the estimation on the historical emission of PAHs in China (Xu et al., 2006), of which biomass burning and domestic coal combustion contribute almost 80% of the total emission. In China, 45% of the total straw yield is used as fuel (Zeng et al., 2007), providing about 33–45% of the energy source in rural area (Zhang et al., 2008). PAH emission from biomass burning is likely to remain an environmental issue within the foreseeable future in China, due to the low prices and easy availability of firewood and straw (Zhang et al., 2008). Immediately after biomass burning, domestic coal combustion is another major source of PAHs in China. During the period of 1980–1990, emission of $\sum PAH_{16}$ in China has increased 28%, primarily due to domestic coal consumption (Xu et al., 2006). However, this situation is likely to be greatly improved due to the shift in household energy sources, with natural gas and liquefied petroleum gas replacing coal as the major household energy sources, especially after 2000 (Fig. 9a).

In addition to the sources mentioned above, increase in vehicle emission is also mentioned in some cases (Guo et al., 2010; Li et al., 2015; Liu et al., 2012b). China has become the world's largest vehicle market and the number of conventional vehicles has reached 154 million till the end of 2014 (Fig. 9b). It has been speculated that vehicle emission could be an important source of PAHs in the near future. However, although the number of vehicle has sharply increased since 2005 (Fig. 9b), average emission of each vehicle has substantially decreased, due to the mandatory improvement of the national emission standards (Shen et al., 2011). The estimated annual vehicle emission of $\sum PAH_{16}$ in China was peaked between 2010 and 2012 with maximum value of 35 Gg and will be decreased in the future (Shen et al., 2011). Besides, it can be expected that the relative contribution of vehicle emission to total PAH emissions will decrease, because the emission from other sources (including wood and biomass burning, domestic and industrial coal combustion) will not change as quickly as those from motor vehicles.

China's environmental policies have become stricter than they used to be (http://www.mep.gov.cn). Fairly strict regulations have been issued by the central government and the judgment to local governments has been changed from their economic performance to take the environmental regulation into account. Nevertheless, actual monitoring and enforcement largely rely on the local governments, who are still more interested in economic growth. A substantial decrease in PAH emissions may be expected in China, but it will take time.

5.3. Sources and their changes of historical PAHs in the United States

Source identification and apportionment of historical PAHs in the United States have been investigated extensively and the source information is relatively comprehensive, with both qualitative and quantitative information available (Table 8). Historical PAHs deposited in the United States are primarily of pyrogenic origin (Cantwell et al., 2007; Lima et al., 2003; Nilsen et al., 2015; Yan et al., 2006), but detailed information is not exactly the same as that in China. Historical PAHs in China are mostly stemmed from biomass burning and coal combustion, whereas

Table 8. Sources characteristics of historical PAHs in the United States.

Location	Method	Sources characteristics	Reference
Lake Michigan	MLR model	A shift from wood burning to coal combustion, and then petroleum combustion	(Christensen and Zhang, 1993)
Kinnickinnic River, WI	CMB model	Coke oven emission and coal-wood gasification or coal tar are the main sources	(Christensen et al., 1997)
San Francisco Bay, CA	DRs	A gradual replacement of wood burning by fossil-fuel combustion as the main source	(Pereira et al., 1999)
Ashtabula River, OH	CMB model	Mainly form highway dust (78.1–83.8%) and coke oven emission (16.8–22.8%)	(Li et al., 2001)
	FA	Traffic (58%), coke oven (26%), and wood burning/coal tar (16%)	(Christensen and Bzdusek, 2005)
Black River, OH	CMB model	Dominated by highway dust, followed by Coke oven emissions and wood burning	(Gu et al., 2003)
	FA	Traffic (51%), coke oven (44%), and wood burning (5%)	(Christensen and Bzdusek, 2005)
Lake Calumet, CHI	CMB model	Coke oven emission (21–53%), and traffic emission (27–63%)	(Li et al., 2003)
	FA	Coke oven (47%) and traffic (45%) sources and wood burning-coal residential (2.3%)	(Bzdusek et al., 2004)
Central Park Lake, NY	DRs	A shift from softwood combustion toward coal combustion, then petroleum combustion	(Yan et al., 2005)
	CMB model	Wood combustion dominated in preindustrial age, a shift to coal combustion from 1900s to 1950s. Petroleum use began around the 1920s and has dominated since the 1940s.	(Yan et al., 2014)
NY/NJ harbor complex	DRs, CSICA	Combustion sources became more important in most area of the NY/NJ Harbor	(Yan et al., 2006)
Taunton River, MA	DRs	Mainly pyrogenic origin, primary coal and wood combustion	(Cantwell et al., 2007)
40 lakes across USA	CMB model	CT sealcoat contribute one-half of PAHs, followed by vehicle emission and coal combustion	(Van Metre and Mahler, 2010)
Puget Sound, WA	DRs, by-products	From softwood combustion toward coal combustion, and then fossil fuel combustion	(Kuo et al., 2011)
Siskiwit Lake	$\Delta^{14}C$ mass balance	Biomass burning exceed fossil fuel sources in the most recent	(Slater et al., 2013)
Lake Lady Bird, TX	CMB model	Coal-tar sealant is the dominant source of PAHs	(Van Metre and Mahler, 2014)
San Francisco Bay, CA	DRs	A predominant petroleum combustion	(Nilsen et al., 2015)

historical PAHs dominated by fossil fuel combustion are the most probability within the last several decades in the United States (Christensen and Bzdusek, 2005; Kuo et al., 2011; Nilsen et al., 2015; Yan et al., 2014). As for the source change, historical PAHs changing from wood combustion toward coal combustion and shifting toward fossil fuel combustion are probably the typical sources of conversion in the United States, as illustrated in Lake Michigan (Christensen and Zhang, 1993), Central Park Lake, NY (Yan et al., 2005) and Puget Sound, WA (Kuo et al., 2011). However, other situation also happened, such as biomass burning replacing fossil fuel combustion as the major PAH source in very recent sediments from Siskiwit Lake (Slater et al., 2013) and coal-tar sealant as the newly emerging source of PAHs at Lady Bird Lake in Austin, Texas (Van Metre and Mahler, 2014).

Early on, Christensen and Zhang (1993) tried to apportion PAHs in Lake Michigan based on multiple linear regressions. Soon afterward, CMB and FA were introduced for the source apportionment of PAHs deposited in sediments (Bzdusek et al., 2004; Christensen and Bzdusek, 2005; Christensen et al., 1997; Gu et al., 2003; Li et al., 2001). These works involved the PAHs in sediments from Ashtabula River, OH (Christensen and Bzdusek, 2005; Li et al., 2001), PAHs in sediments from Black River, OH (Christensen and Bzdusek, 2005; Gu et al., 2003), and PAHs in the Lake Calumet, Chicago (Bzdusek et al., 2004; Li et al., 2003), as listed in Table 8. The results derived from CMB and FA are sound and can support each other. This attempt provides a quantitative approach to evaluate the relative contribution of various sources to the receptor and is adopted to assess the contribution of coal-tar sealant to PAHs recorded in lakes across the United States (Van Metre and Mahler, 2010, 2014). Another attempt is to develop the mass balance equation using 1,7/(1,7 + 2,6)-DMP, as an indicator of softwood combustion (Benner et al., 1995). It also has the potential to differentiate various pyrogenic PAHs, such as wood, coal, and petroleum combustion (Yan et al., 2005). Based on this, a mass balance equation was developed for source apportionment of PAHs recorded in Central Park Lake, NY (Yan et al., 2014). The results indicated that: 1) PAHs from wood combustion dominated during the preindustrial age in NY, followed by a shift to coal combustion between 1900s and 1920s, and 2) petroleum began to be used around the 1920s and replaced coal as the major contribution since the 1950s. Quantitative information on the apportionment of historical PAHs can also be obtained based on the analysis of $\delta^{13}C$ (Yan et al., 2006) or $\Delta^{14}C$ (Slater et al., 2013). In short, more quantitative information on historical PAHs in the United States can be available.

5.4. Sources and their changes of historical PAHs in other areas

Source information on historical PAHs in Japan, Canada, Europe, and other area is mostly based on the DRs and is qualitative at most. In Japan, source of PAHs recorded in natural lakes, such as Lake Kitaura and Lake Biwa, mainly originated from grass, wood burning, and coal combustion, without significant change over the last few decades (Itoh et al., 2010, 2017). However, in urban area, historical PAHs are highly affected by the WWII and exhibit a source change from coal combustion to fossil fuel combustion (Ishitake et al., 2007; Moriwaki et al., 2005). In Canada, sources of historical PAHs are highly site specific. For example, PAHs recorded in Lake Tantaré mostly come from coal combustion (Gallon et al., 2005), while historical PAHs in Hecate Strait are primarily of natural origin, with little or no anthropogenic input (Yunker et al., 2014). In Saskatchewan lakes, all evidences point out that forest fire is the principal PAH source (Ahad et al., 2015). In Europe, historical PAHs' input to Scheldt estuary, Netherland is mainly from coal combustion (Van Zoest and Van Eck, 1993), while PAHs recorded in a remote lake, Scotland are deduced to be from local sources, i.e., combustion of domestic wood and coal, rather than industrial input (Rose and Rippey, 2002). Historical PAHs recorded at Aspvreten, Sweden covered a long period of time and experienced a source change from wood combustion to coal combustion, and shift toward fossil fuel combustion, as well as the rebound of softwood burning very recently (Elmquist et al., 2007).

As to the sources of historical PAHs recorded in other areas, they are site dependent. Historical PAHs in Masan Bay, Korea were characterized by strong pyrogenic origin (Yim et al., 2005), while it was primarily natural origin for PAHs recorded in Laja Lake, Chile (Quiroz et al., 2005). Sources of historical PAHs deposited in Sundarban Wetland, India are dominated by wood and coal combustion (Dominguez et al., 2010), while petroleum combustion dominated the source of PAHs deposited in Las Matas, Gulf of Mexico (Ruiz-Fernández et al., 2012). In Brazil, historical PAHs put into the Santos Estuary seem chiefly from biomass and coal combustion, but conflicting information is delivered by the ratio of FLUO/(FLUO + PYR) (Martins et al., 2011). As for the sources of historical PAHs in Barigui River, they are dominated by grass and wood combustion before 1920, followed by a mixed source of biomass and petroleum combustion till 1940, and dominated by petroleum combustion since then (Machado et al., 2014).

It is worthwhile noting that, in addition to DRs, spheroidal carbonaceous particles (SCPs) can also act as evidence for specific source of PAHs in sediment (O'Dwyer and Taylor, 2009), because they are by-products of fossil fuel combustion and cannot be formed under natural processes (Rose and Rippey, 2002). Consequently, SCPs are thought to be an indicator of fossil fuel combustion (Rose et al., 1995; Rose and Monteith, 2005) and have been used to track PAHs deposited in Admiralty Bay, King George Island, Antarctica (Martins et al., 2010).

6. Concerns and further efforts on sedimentary PAHs

Although the length of this article is beyond our expectation, there have been several issues that remain unclear and need clarification. The first one is that many works in China usually link the vertical profiles of PAHs to various socioeconomic development indexes, such as energy consumption, population, and gross domestic product. (Guan et al., 2012; Li et al., 2015; Liu et al., 2009; Liu et al., 2012a; Tang et al., 2015). Indeed, these indexes have great influence on the PAH emissions than reflecting in the vertical profiles of PAHs. Good relationship between the vertical profiles of PAHs and these indexes can be obtained and the explanation sounds reasonable, but some statements are farfetched. In fact, there is no direct logical relationship between the vertical profiles of PAHs and socioeconomic development indexes. The increase in energy consumption and GDP is not necessary resulting in the increase in PAH emissions, depending on the cleanliness of energy and GDP. For example, petroleum is cleaner than coal due to its relatively lower emission factor during combustion processes. Large uncertainty in PAH emissions could be introduced even if the same energy source was used. For example, PAHs' emission factors of vehicle can vary more than 4–5 orders of magnitude, due to the differences in vehicle type, fuel type, operation mode, and ambient temperature (Baek et al., 1991). In a word, PAH emission is a complex function of socioeconomic development, energy consumption, emission control techniques, and environmental policies, as well as climate conditions. Available information indicates the synchronism of socioeconomic development and environmental pollution, but large uncertainty still exists. Further efforts are needed for more accurate evaluation.

The second issue is associated with the source assignment. Although many methods, both qualitative and quantitative, have been introduced, it should be borne in mind that each method has its limitations and should be used with caution. For example, when using DRs for source identification, different DRs could provide conflicting source information. The main reason is that the critical value for each DR is actually a statistical result, rather than the true values of specific PAH sources. If this happens, one should be cautious with the conclusion and more evidences are needed before making a decision. For CBM model, representative and accurate source information are the key points for a successful source apportionment. Meanwhile, the basic assumptions should be kept in mind. In actual operation, although ideal situation (satisfying all the assumptions) is not easy to get, it is not desirable when the actual situation is far away from these assumptions. Before operating a model, it is necessary to assess to what extent the actual situation can meet the requirements of the model. As to PCA, a method claims to be able to achieve source apportionment with minimal source information. We privately think that the source information is still very important, especially when the possible sources are similar to each other and not easy to assign.

The third one is concerned about the precise chronology of PAHs. Quantitative reconstruction of contaminants' inputs requires a precise chronology, which can be complicated by postdepositional mixing of sediments through physical, chemical, and/or biological mechanisms and potentially fuzzy the original imprint in sediment. Single radionuclide cannot take all these complications into account. Thus, two or more radionuclides are frequently used in dating analysis. And if necessary, sediment focusing must be assessed for a better chronology. We stress this issue because the sedimentary record of PAHs is occasionally overinterpreted without a precise chronology. This can result in misleading or a wrong decision.

The last one is related to the influence of postdepositional behaviors of PAHs. Once introduced into sediment, PAHs can be absorbed by various geosorbents in sediment simultaneously, leading to the redistribution of PAHs among different geosorbents and finally the sequestration of PAHs. Operationally, the sequestrated PAHs in sediment cannot be extracted by conventional extracting method. This means that the extractable PAHs cannot represent the total PAHs that occurred in sediment. Previous work has confirmed that a large portion of PAHs (33.8–57.4%)

may not be extracted by conventional extracting methods (He et al., 2008). This is supported by another work published very recently (Guo et al., 2017). Crucially, sequestration of PAHs is selective during the postdepositional process. Whether the extractable PAHs can represent whole information of PAHs that occurred in sediment is questionable. At least, we need to prove it.

Abbreviations

ACE	Acenaphthene
ACY	Acenaphthylene
ANT	Anthracene
BaA	Benzo[a]anthracene
BaP	Benz[a]pyrene
BbF	Benzo[b]fluoranthe
BBG	Beibu Gulf
BeP	Benz[e]pyrene
BghiP	Benzo[ghi]perylene
BHS	Bohai Sea
BjF	Benzo[j]fluoranthene
BkF	Benzo[k]fluoranthene
BP	Biphenyl
BYD	Lake Baiyangdian
CH	Lake Chaohu
CHH	Lake Chenghai
CHRY	Chrysene
COR	Coronene
CSCIA	Compound-specific carbon isotope analysis
DB	Deep Bay
DBA	Dibenz[ah]anthracene
DC	Lake Dianchi
DMNAP	Dimethylnaphthalene
DMP	Dimethylphenanthrene
DR	Diagnostic ratios
DYB	Daya Bay
ECS	East China Sea
EH	Lake Erhai
FLU	Fluorine
FLUO	Fluoranthene
HF	Lake Hongfeng
HLB	Hailing Bay
HZB	Haizhou Bay
INP	Indeno[1,2,3-cd]pyrene
LRE	Liao River Estuary
LZB	Laizhou Bay

MNAP	Methylnaphthalene
MPHEN	Methylphenanthrene
NAP	Naphthalene
NS	Lake Nansi
PAHs	Polycyclic aromatic hydrocarbons
PERY	Perylene
PHEN	Phenanthrene
PRE	Pearl River Estuary
PY	Lake Poyang
PYR	Pyrene
QH	Lake Qinghai
RET	Retene
SCS	South China Sea
SG	Lake Sugan
SHLW	Maar Lake Sihailongwan
TH	Lake Taihu
TMNAP	Trimethylnaphthalene
YaRE	Yangtze River Estuary
YeRE	Yellow River Estuary
YS	Yellow Sea
$\sum HMW$	sum of four to six-rings PAHs
$\sum LMW$	sum of two to three-rings PAHs
$\sum PAH_{16}$ *(considered as the standard suite for PAHs in this review)*	sum of sixteen US EPA priority PAHs

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