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Sedimentary archive of Polycyclic Aromatic Hydrocarbons and perylene sources in the northern part of Taihu Lake, China

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## 1 Sedimentary Archive of Polycyclic Aromatic Hydrocarbons and Perylene

## 2 Sources in the Northern Part of Taihu Lake, China

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Abstract: In the present work, we analyzed the concentration patterns of 20 Polycyclic Aromatic 18 Hydrocarbons (PAHs) in 25 surface sediments and 11 sediment cores from the northern part of Taihu 19 Lake, China. Three of the cores were dated based on <sup>137</sup>Cs activity for the deposition age of the 20 sediment. The spatial distributions of the PAH concentrations show that the inflow rivers into Zhushan 21 Bay and Meiliang Bay were the main pathway for PAHs and sediment input to the northern part of the 22 lake. This results in substantially higher PAH concentrations (up to 5000 ng/g) and sedimentation rates 23 24 (higher than the average of 3 - 4 mm/a) in the area close to the river outlets. In addition, results also show that PAH concentrations in the sediments considerably increased from the early 1960s, but the 25 decreasing concentrations in the upper layers of the sediment could be attributed to the introduction of 26 measures on environmental improvement from ca. 2000. There were both anthropogenic and biogenic 27 origins of perylene in the lake sediments, which were distinguished based on spatial distribution 28 patterns and also the concentration proportions of perylene to the sum of the 20 PAHs. In the cores 29 collected close to river outlets, the concentration proportions of perylene typically range from 0.02 to 30 0.18 and there are significant positive linear correlations between the concentration of perylene and 31 32 three anthropogenic PAHs (Benzo[a]pyrene, Benzo[e]pyrene, Pyrene), suggesting that perylene was dominated by anthropogenic input. However, the cores collected further away from the river outlets 33 show the concentration proportions between 0.13 and 0.96, and present significant negative 34 correlations or no correlations between perylene and the three PAHs, suggesting that perylene was 35 36 mainly formed by biogenic activities. Furthermore, the different perylene sources accompanied with the location distributions imply that anthropogenic activities could inhibit its biogenic formation. 37

#### 38 Keywords

39 Taihu Lake in China; Lake sediment; PAH distribution; Perylene source

#### 40 Capsule

This study provides an insight of anthropogenic impacts on PAH deposition in the sediment of Taihu
Lake and also clearly distinguishes biogenic and anthropogenic origins of perylene in the sediment.

43

#### 44 **1. Introduction**

45 Taihu Lake, located in the Yangtze River Delta plain, is a large and shallow freshwater lake with an area of 2338 km<sup>2</sup>. The lake plays an important role for flood control in the region and also for tourism, 46 47 shipping, aquaculture, and as a raw source for drinking water supply in the neighboring cities of 48 Shanghai, Suzhou, Wuxi and Huzhou (Qin et al., 2007) (Fig. 1a). However, decades of rapid 49 development of industrialization and urbanization in this area caused serious pollution in the lake 50 ecosystems, especially the northern part of the lake (Wang et al., 2003; Wilhelm et al., 2011; Jiang et 51 al., 2012; Xu et al., 2014; Tao et al., 2018). This is due to the discharge of large amounts of industrial 52 effluents and improperly treated municipal sewage into the lake, the diffuse pollution from agriculture 53 and aquaculture, or the wet and dry atmospheric deposition of dissolved and particulate matter from traffic and biomass burning. In 2007, e.g., excessive nutrient input into the lake caused a massive 54 55 cyanobacterial bloom with the formation of volatile sulfide compounds, resulting in a serious drinking water crisis in Wuxi City (Zhang et al., 2010). 56

Lake ecosystems are particularly sensitive to anthropogenic impacts as they can act as repositories for 57 contaminants in the aquatic environment. Especially hydrophobic persistent organic pollutants (POPs) 58 59 can easily bind to organic matter and then deposit in sediments. After deposition, such contaminants are less susceptible to microbial degradation due to their strong sorption and ageing, which reduces 60 contaminant bioavailability (Erickson et al., 1993; Hatzinger and Alexander 1995). Lake sediments 61 62 therefore may contain continuous archives of such inputs with an annual to decadal resolution. With 63 this, the analyses of lake sediments that may have accumulated over decades or centuries can provide insights into background conditions, and into historical, present and potentially future anthropogenic 64 impacts (Hollert et al., 2018). 65

Polycyclic Aromatic Hydrocarbons (PAHs) are such a group of POPs and ubiquitously distributed in ecosystems. They have been emitted since prehistoric time through e.g. wild fires (Vila-Escalé et al., 2007) and volcanos (Kozak et al., 2017). They are also indicators for anthropogenic activities resulting from incomplete combustion of fossil fuels and biomass (Zhang et al., 2008; Shen et al., 2012; Wu et al., 2017) and from various industrial processes (Yang et al., 2002), substantially raising PAH

abundance in the environment. Some of the PAHs such as benzo[a]pyrene, dibenzo[a,h]anthracene, 71 benzo[a]anthracene are classified as mutagenic and carcinogenic and are listed as priority pollutants by 72 73 US EPA (EPA US, 1993; Boffetta et al., 1997; Kim et al., 2013). Consequently, once these PAHs accumulate in the environment and biomagnify in food chains, they ultimately pose considerable 74 threats to human and ecosystem health (Geffard et al., 2003). A specific PAH, perylene, can also be 75 formed biogenically in sediments after deposition, e.g. probably from diatom (Louda and Baker, 76 77 1984), fungi (Suzuki et al., 2010), crinoids (Wolkensein et al., 2006), plants (Marynowski et al., 2013; Grice et al., 2009) or microbial transformations (Silliman et al., 2000). Therefore, its occurrence in 78 sediments may indicate anthropogenic PAH input and also biogenic formation. 79

80 A certain amount of studies on PAHs in the sediment of Taihu Lake has been conducted (Qu et al., 2002; Peng et al., 2005; Qiao et al., 2006; Lei et al., 2014; Tang et al., 2015; Lei et al., 2016), which 81 conclude that main PAH input are from the densely populated and industrialized region in the north of 82 the lake. However, the historical PAH inputs and transport patterns in the lake are still not well known. 83 In addition, pervlene sources and formation have not vet been studied in Taihu Lake. In the present 84 work, we analyzed 25 surface sediment samples and 11 sediment cores from the northern part of Taihu 85 Lake for a more detailed investigation of PAH contents and patterns in this region of the lake. PAH 86 87 concentrations in the surface sediments were used to delineate current PAH input into the lake, and 88 PAH concentrations in the cores to record the temporal changes of PAH input. Specific attention is paid to the concentrations of perylene in relation to the other PAHs to potentially distinguish the 89 anthropogenic sources and biogenic formations of perylene in the environment. 90

#### 91 2. Materials and methods

With a mean depth of 1.9 m and a maximum depth of 2.6 m, Taihu Lake is a characterized by a rather flat lake bed. There are around 120 inflows and outflows around the lake, with the inflows mostly located in the northern and western part and the outflows in the eastern and southern part of the lake (Qin, 2008). Some are occasionally reversing in flow direction, depending on the lake water level. The shallowness of the lake and the hydraulic conditions result in complex flow patterns, and the spatial and temporal current patterns in the lake are not fully understood (Qin et al., 2007).

These complex flow conditions have an influence on the spatial distribution and deposition rates of 98 sediment in the lake. Sediment thickness varies between 0.5 and 2 m in most parts of the lake with the 99 greater in the northern and western parts and lesser sediment thickness in the central parts, reflecting 100 the prevailing wind and resulting water circulation patterns (Luo et al., 2004). In the upper layers, 101 sediment textures are dominated by clayey silt and clay (Jin et al., 2006; Qin et al., 2004). It is, 102 however, likely that during strong winds or floods sediment scouring and resuspension can alter the 103 104 sediment stratigraphy (Qin et al., 2004), complicating the interpretation of dating results and 105 contaminant profiles.

#### 106 **2.1 Sample collection**

During five sampling campaigns (May 2015, November 2015, June 2016, February 2017, September 107 2017) a total of 25 grab surface sediment samples and 11 sediment cores ranging between 12 cm and 108 109 40 cm in length, were collected from the northern part of Taihu Lake, covering Gonghu Bay, Meiliang Bay and Zhushan Bay. The surface sediment samples were stored in polyethylene bags. The cores 110 were taken using a gravity corer with a core loss preventer (uwitec, Austria), and they were 111 subsequently sliced into 2 cm segments (147 in total) and stored in aluminum screw top jars. Before 112 further analyses, the samples were stored at -20 °C, except during the air shipping from China to 113 Germany. 114



115

Fig. 1 a Overview of the area surrounding Taihu Lake; b Sampling sites for surface sediments and sediment cores. Exact coordinates of the samples and the sampling time are listed in S Tab. 1. The rivers and canals around the sampling area are generally only inflows into the lake. The Wangyu River connecting Taihu Lake and the Yangtze River, however, can act as an outflow in the rainy season.

#### 120 2.2 Sample preparation and PAH analysis

121 All sediment samples were silty-clayey and therefore whole samples were freeze-dried and milled in a vibratory disc mill. Between 7 and 12 g of each sample was then extracted with 35 ml of acetone for 122 123 30 min using Accelerated Solvent Extraction (Dionex ASE 300) in static conditions (100 °C and 10 124 MPa). After extraction, 100  $\mu$ L of an internal standard was spiked to each sample extract. Extracts were then cleaned using a glass column filled with glass wool, 2 g silica gel, 2 g Al<sub>2</sub>O<sub>3</sub> and 0.5 g 125  $Na_2SO_4$  to remove the residual water and interfering compounds. After clean-up, the column was 126 eluted with 15 mL of n-hexane, followed by 5 mL of a 9:1 (v/v) mixture of n-hexane and 127 dichloromethane, and 20 mL of a 4:1 (v/v) mixture of n-hexane and dichloromethane. The eluate was 128 129 concentrated to around 2 mL using an automatic evaporation-dryer with nitrogen. Then 1 µL of the concentrated eluate was injected into a GC-MS (Agilent 7890A/5975C) in pulsed splitless mode and 130 131 detected in the SIM mode of the MS.

For quantification, PAH standards (PAH-mix 14, PAH-mix 45 and deuterated PAH-mix 31) were obtained from Dr. Ehrenstorfer Augsburg, Germany. The standards were diluted together in cyclohexane to four different concentrations for external calibration and response factors calculation. The internal standard (dilution of deuterated PAH-mix 31) with 5 deuterated PAHs was then used to quantify the analytes. All solvents and cleanup chemicals were purchased from Carl Roth GmbH + Co.KG, Germany.

138 Including 16 EPA PAHs, a total of 20 PAHs were analyzed (2-ring: naphthalene, 2-methyl-139 naphthalene, 1-methyl-naphthalene; 3-ring: acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene; 4-ring: fluoranthene, pyrene, benzo[a]anthracene, chrysene; 5-ring: benzo[b]fluoranthene, 140 141 benzo[k]fluoranthene, benzo[e]pyrene (BeP), benzo[a]pyrene (BaP), perylene; 6-ring: 142 dibenzo[a,h]anthracene, benzo[g,h,i]perylene, indeno[1,2,3-cd]pyrene).

#### 143 **2.3 Quality control**

For testing the reliability of this method, a certified soil from a gas works site (European Reference
Material ERM-CC013a) was purchased from the Federal Institute for Materials Research and Testing
BAM (Berlin, Germany). The reported uncertainties for the concentrations of the different PAHs in

the certified soil were between 5% and 20%. The certified soil was extracted and measured identically
to the field samples. Average recovery rates (9 replicates) in our tests were typically between 95% and
130%, indicating good recoveries.

150 The quantification limit for the GC-MS analysis was between 10 and 25 pg of injected mass, 151 depending on the PAH. This limit corresponds to 3 - 7 ng/g of individual PAHs in the soil samples, 152 depending on the amount of soil extracted and the volume of the eluates concentrated.

#### 153 2.4 Dating methods

Three of the cores, located in the northern part of Meiliang Bay (ML35), southern part of Meiliang 154 Bay (ML36) and Zhushan Bay (ZS42) were dated using the thermonuclear by-product <sup>137</sup>Cs that has a 155 half-life of 30.17 years. Since a certain mass was required for the determination of <sup>137</sup>Cs activities, 156 some adjacent layers of core samples were combined and each dating sample ranges between 2 and 8 157 cm in length and between 15.1 and 29.9 g in weight. <sup>137</sup>Cs activity measurements were carried out with 158 low-level gamma-spectroscopy based on the distinct <sup>137</sup>Cs gamma emission energy of 661 keV, using 159 a n-type coaxial Low-Energy HPGe detector (ORTEC) with an active volume of 39 cm<sup>3</sup> and a 0.5 mm 160 Be window. The detector efficiency and measuring geometry were calibrated with the certified 161 reference material "IAEA-375 SOIL". Each of the samples was measured at least 24 hours in 32 cm<sup>3</sup> 162 cylindrical capsules. Spectra analysis was performed with the software GAMMA-W<sup>®</sup>. 163

#### 164 **3. Results and discussion**

Riverine runoff and atmospheric deposition are the two main pathways responsible for PAHs and sediment input to a lake (Ferrey et al., 2018; Zakaria et al., 2002). Riverine runoff may potentially contribute more significant pollutant and particle loads, as rivers typically receive treated or untreated domestic wastewater, industrial effluents, and surface runoff (Wolf et al. 2013; Pal et al., 2010). These factors may lead to higher deposition rates and PAH concentrations in the sediment close to river inflows.

#### 171 **3.1 Dating of the cores**

The presence of anthropogenic <sup>137</sup>Cs in the environment of the northern hemisphere can be traced both to the atmospheric testing of nuclear weapons in the late 1950s and early 1960s and to major accidents in nuclear power stations, i.e. the Chernobyl accident in 1986. The latter produced a less uniform and laterally more limited pattern since the fallout was associated with complex short-term weather patterns (in particular rain). Whereas (for this reason) no Chernobyl fallout is known for the southern hemisphere, the nuclear weapon testing fallout is traceable in both the north and the south, appearing in the south about two years later (Rowntree and Foster, 2012).

Sediment deposition rates for the three locations were measured based on <sup>137</sup>Cs activities in the cores 179 taken from that locations. Fig. 2 shows that the <sup>137</sup>Cs activities are rather low and only detectable in the 180 upper around 20 cm of the three cores, which is comparable to the results reported by others (Xue and 181 Yao, 2011; Liu et al., 2004). Although the detected <sup>137</sup>Cs activities in the cores do not show any 182 distinct peak related to any of the events, the deepest layer (at ca. 20 cm depth) at which <sup>137</sup>Cs was 183 detected can be consequently dated to around 1960. This would indicate that an average sedimentation 184 rate at these three locations is around 3 - 4 mm/a, which is similar to the rates reported by Xue and 185 Yao (2011) and Liu et al. (2009), and is also reasonably consistent with the chronological results of 186 spheroidal carbonaceous particles (Cao et al., 2013; Liu et al., 2012). These three locations are some 187 km away from the lakefront and also away from the inflows into the lake. The sedimentation rates 188 189 therefore represent most likely the average rate in the northern part of the lake, but might not be 190 representative for the areas close to the lakefront and river outlets due to more intense sediment input.



**192** Fig. 2 Distribution of <sup>137</sup>Cs activities in the three dated cores

**3.2 PAH in the surface sediments and the cores** 

Fig. 3a shows the spatial distribution of the total PAH concentrations, without considering perylene, in the surface sediments. The concentrations range from around 150 ng/g to around 2300 ng/g. The higher concentrations (above 1000 ng/g) are found close to the inflow in Zhushan Bay (locations 8-23, 4-41 and 3-18) and in the northwestern part of Meiliang Bay (location 8-24). The concentrations at the other locations are typically below 1000 ng/g. These distribution patterns, with the higher concentrations located close to the inflows, reveal that the inflow rivers are the major current pathways for PAH input.



Fig. 3 Concentrations of the sum of the 19 PAHs (without perylene) (a) and of perylene (b) in the surfacesediments

Three sediment cores were also taken close to the inflows in Zhushan Bay (ZS23), and in the 204 northwestern and northeastern part of Meiliang Bay (ML7, ML24) (Fig. 1b). In line with the results of 205 206 the surface sediments, the three cores show the by far highest PAH concentrations (without pervlene), 207 reaching up to 5000 ng/g in core ML24 (Fig. 4). These three cores have a length of 40 cm (ZS23), 35 208 cm (ML7) and 18 cm (ML24), respectively, and their concentrations remain high throughout the cores. 209 The other cores (ML6, ML35, ML36, ML43, ZS42) collected further away from the inflows in the two 210 bays, however, show consistently low background concentrations (below around 150 ng/g) in the 211 deeper parts and comparably high concentrations only in the upper 10 - 20 cm. These results suggest that the sedimentation rates in the area close to the inflows are substantially higher than the average 212 213 rate of 3 - 4 mm/a in the other locations, and that the rivers connected to the northern part of these two bays are the main pathway for PAHs and sediment input into the lake. 214

215 These general findings are also reflected in the three cores (ML6, ML35 and ML36) located with 216 increasing distances from the river outlets in Meiliang Bay (Fig. 1b). These cores present similar 217 concentration profiles, with background concentrations in the deeper layers and higher concentrations in the upper layers (Fig. 4). The concentrations increase from a depth of 28 cm (ML6), 26 cm (ML35) 218 219 and 22 cm (ML36), respectively, which indicate that the sedimentation rates in the three locations 220 decrease with increasing distances from the river outlets. In addition, core ML36 has lower 221 concentrations in the upper layers compared to the other two cores, in line with the greater distance from river inflows. The two cores located in the south of Zhushan Bay and Meiliang Bay (ZS42 and 222 ML43) show even lower sedimentation rates and also lower PAH concentrations (350 - 450 ng/g) in 223 224 their upper layers.



225

Fig. 4 Concentration profiles of the sum of the 19 PAHs (without perylene) in the cores (different x axis scale)

Three cores (GH4, GH38 and GH11) were taken from Gonghu Bay (Fig. 1b). Taihu Lake is a crucial 227 drinking water source in this region, especially Gonghu Bay provides water to 4 major water treatment 228 229 works covering 80% of drinking water supply in Wuxi City (Qin et al. 2010; Tao et al., 2010). Gonghu 230 Bay is connected to the Yangtze River through the Wangyu River. In order to alleviate algae blooms 231 in Taihu Lake, a project (WTYT project) was started in 2002 to transfer water from the comparatively 232 low nutrient status Yangtze River to Taihu Lake through the Wangyu River (Zhai et al., 2010). In addition, sediment dredging was conducted around ten years ago to remove contaminated sediments 233 from the bay (Liu et al., 2010; Liu et al., 2016; Chen et al., 2018). These activities likely had an impact 234 235 on the contamination patterns and thickness of the sediments. The three cores show significantly 236 different concentration profiles that consequently might be influenced by the activities in this bay and may not represent the original stratification. 237

In principal, the PAH concentration patterns together with the information on sedimentation rates can 238 be used to reveal the historical input of PAHs into the sediments. Coal and oil are the major energy 239 sources in China (Crompton and Wu, 2005; Wang and Feng, 2003), hence PAH abundance in the 240 241 sediments is a crucial indicator of energy consumption and industrial and economic development. After the People's Republic of China was established in 1949, China started intensifying the 242 development of industry and economy, particularly in coastal areas (Li, 2009; Fan, 1995), which was 243 accompanied with a considerable increase in consumption of coal and oil (Liu, 2008; Jiang and Zhang, 244 245 2005). Furthermore, the reform and opening-up Policy in China, implemented in 1978, was associated 246 with rapidly growing urbanization and industrialization (Yeh et al., 2011; Chen et al., 2013). Therefore, combining the PAH concentration profiles and the dating results with an average 247 sedimentation rate of 3 - 4 mm/a in the sediments away from inflows, the significant increases of the 248 249 PAH concentrations documented in cores ML6, ML35, ML36, ML43, and ZS42 would start from the 250 early 1960s.

However, the concentrations are rather stable or somehow decreased in the upper layers of the cores compared to the peak concentration in each of the cores. In contrast, energy consumptions in the catchment are still increasing after 2000. Therefore, the inconsistent PAH concentration distributions

with energy consumption could be accounted for by the fact that the Chinese government has enacted
and enforced a series of laws and regulations regarding ecologically sustainable development and
resources conservation since the 9th Five-Year Plan (1996 – 2000). From the 10th Five-Year Plan
(2001 – 2005), in particular, multiple measures for environmental improvement were introduced
(Schreifels et al., 2012; Chen and Xu, 2010; Zhou et al., 2010; Crompton and Wu, 2005). Especially,
Taihu Lake is one of the prioritized regions for pollution control and treatment.

#### 260 **3.3** Perylene in the surface sediments and the cores

Perylene concentrations in the surface sediments are typically below 200 ng/g, except in the five locations 8-6, 8-7, 8-21, 3-17 and 3-21 with higher concentrations (200 – 300 ng/g) (Fig. 3b). Compared to the other PAHs (Fig. 3a), perylene concentrations are relatively high in most of the sampling locations and the higher concentrations are generally located in the area close to the central part of the lake. The spatial distribution of perylene concentrations is somewhat inverse to the distribution of the other PAHs, which is especially obvious in Zhushan Bay.

As in the case of the surface sediments, pervlene concentrations in the cores are generally below 200 267 268 ng/g, except cores ML35, GH38 and ZS42 (Fig. 5). In some sections of cores ML35 and GH38, 269 perylene concentrations reach over 1000 ng/g, accounting for up to 96% of the sum of the 20 PAH concentrations. Concentration patterns, however, in the three cores are different. In core ML35, the 270 high concentrations occur in the deeper layers and the relatively low and stable ones (around 200 ng/g) 271 272 in the upper layers, while in core GH38 perylene concentrations generally increase from the deeper to the upper layer of the core with the highest concentration at 8 cm. In addition, the concentrations in 273 core ZS42 decrease linearly from the deeper layer (around 450 ng/g) to the upper one (around 150 274 ng/g). Nevertheless, in the other cores, perylene concentrations are low and vary slightly, particularly 275 276 in the upper 15 - 20 cm.



277

278 Fig. 5 Concentration profiles of perylene in the cores (below QL: below the quantification limit)

Compared to the other PAHs, the concentration profiles of perylene are specific in each core, which 279 280 might result from two potential sources, anthropogenic input together with the other PAHs and 281 biogenic formation in the sediments after deposition. Three anthropogenic PAHs (BaP, BeP and pyrene) were selected as references to distinguish between these two sources (Venkatesan, 1988). The 282 five cores (ZS23, ML24, ML7, GH11 and GH4) collected close to the river outlets show significant 283 positive linear correlations between the concentration of perylene and the three PAHs (Fig. 6a), while 284 285 the other six cores (ZS42, ML6, ML35, ML36, ML43 and GH38) collected far from the river outlets show significant negative correlations (Fig. 6b) or no correlations (cores ML36 and ML43 shown in S 286 (supplementary material) Fig. 2). The significant positive linear correlations found close to the river 287

outlets might indicate that perylene was deposited at these locations together with the other PAHs originating from anthropogenic sources in the upstream areas. The detailed results and explanation of the correlation and regression calculations are presented in S Tab. 2 and its following text.



Fig. 6 Concentration correlations between perylene and the three anthropogenic PAHs (BaP, BeP and pyrene) with (a) significant positive linear correlations and (b) significant negative correlations. Four points from core GH4, one point from core ML7, one point from core ZS23 and two points from core ML6 (labelled with grey color) are outliers and are excluded from the correlation and regression calculations, the details about the outliers are explained in supplementary material (below S Tab. 1).

297 Furthermore, lower perylene abundance relative to other PAHs suggests that perylene mainly originates from pyrolytic processes instead of diagenetic processes (Readman et al., 2002; Baumard et 298 al., 1998). In this study, the concentration proportions of perylene to the sum of the 20 PAHs (S Fig. 1) 299 300 are typically lower (0.02 to 0.18) in the five cores with significant positive linear correlations compared to the other six cores (0.13 to 0.96) with significant negative correlations or without 301 correlations. This can be also supported by the perylene concentration distributions in the surface 302 303 sediment (Fig. 3b) where the lower concentrations are generally located in the areas near the river 304 outlets.

Consequently, these three aspects, the positive linear concentration correlations, the lower concentration proportions and the locations with short distance from inflows, together strongly imply that perylene in these five locations originated mainly from anthropogenic activities and were input by river runoff. In addition, the different slopes of the regressions (Fig. 6a) and the different perylene proportions (S Fig. 1) in the five cores might suggest that there were different anthropogenic PAH sources in the inflow rivers.

However, in the other six locations ML6, ML35, ML36, ML43, GH38 and ZS42, perylene could be mainly formed in situ by biogenic activities without obvious spatial transportation and interaction. This is supported by the negative or no correlations between the PAH concentrations and also the higher perylene concentration proportions on one hand, and on the other hand by the specific perylene concentration profiles.

316 Previous studies suggested that phytoplankton, particularly diatom, could be perylene precursors in deep aquatic sediments (Louda and Baker, 1984; Venkatesan and Kaplan1987; Soma et al., 1996). 317 Massive nutrient inputs have resulted in the rapid proliferation of phytoplankton in Taihu Lake since 318 the 1980s and the occurrence of phytoplankton bloom became more frequent and severe after ca. 2000 319 320 (Dong et al., 2008; Duan et al., 2009; 2015). However, perylene concentrations fluctuate slightly or even decrease in the upper layers of the cores. This implies that the distribution of perylene is not 321 consistent with the accumulation of phytoplankton, which agrees with Silliman's report (1998) that 322 323 there is poor correlation between the mass accumulation rates of perylene and biogenic silica (an 324 indicator of diatom production) in Lake Ontario.

The results also indicate that biogenic formation of perylene might be inhibited in the sediments where anthropogenic impacts are stronger, which is close to the river inflows and in the upper layer of the sediments. Nutrients, organic matter and pollution levels can influence the diversity of microbial communities and mineralization in sediments (Zeng et al., 2005; Haller et al., 2011; Xu et al., 2018) and therefore could also affect biogenic formation of perylene in sediments.

330 4. Conclusions

Analyses of the spatial distribution of PAHs in the sediments in the northern part of Taihu Lake reveal that the main PAH input into the lake occurs through the inflowing rivers into Zhushan Bay and Meiliang Bay. This coincides with higher sediment deposition rates close to the river inflows. PAH concentrations in the sediments in Meiliang Bay are higher compared to the other two bays.

The temporal analyses of the cores show that increasing PAH input into the lake started from the 1960s due to rapid economic and industrial development. Over the years, PAH concentrations in the sediments increased by a factor of 10 or more compared to the background concentrations. However, the decreased or stable PAH concentrations in the upper layers of the sediments might show the effects of environmental measures implemented by the Chinese government from ca. 2000, although more data are needed to prove this.

The spatial distributions of perylene concentrations in the sediments and its concentrations in relation to the other PAHs suggest that Perylene originates from the same sources as the other anthropogenic PAHs in the locations close to the river inflows. However, it may result from biogenic processes in the locations far away from inflows. Consequently, this could imply that the biogenic formation of perylene is inhibited by anthropogenic activities.

#### 346 Declaration of interest: none

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## HIGHLIGHT

- River inflows are the main pathway for PAHs and particles input in the northern part of Taihu Lake
- Anthropogenic activities significantly influenced the PAH abundance in the lake sediment
- Perylene originated from anthropogenic and biogenic activities in different sampling areas

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