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Climate change impacts on polychlorinated biphenyls (PCBs) deposition in a shallow lake, China

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Climate change impacts on polychlorinated biphenyls (PCBs) deposition in a shallow lake, China

Hanxiao Zhang and Shouliang Huo[1]

State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Science, Beijing 100012, China.

[1] Email: huosl@craes.org.cn

Abstract. The deposition of PCBs in lake sediments was affected by climate change and anthropogenic activities. To better understand the mechanism, GAMs models were performed to investigate the relationship between sedimentary PCBs and the two influential factors. The results showed the average contribution value of the sum of temperature and evaporation to PCBs (7.1%) was larger than that of the sum of aquatic TP and GDP (17.9%), indicating human activity is a major factor compared to climate change. The increasing total phosphorus and Gross Domestic Product in Lake Chaohu with the development of urbanization and industrialization promoted the deposition of PCBs in Lake Chaohu sediments before the consideration of environment management. The fitted relationship between sedimentary PCBs concentrations and Temperature was linear for Lake Chaohu, suggesting that increasing temperature enhanced the volatilization from primary and secondary sources and reduced the fugacity capacity of PCBs in water and sediments. The sedimentary PCBs concentrations increased with decreasing evaporation. In consequence, the interactive effect of climate change and anthropogenic activities must be addressed from a scientific perspective for future regulatory actions.

1. Introduction

The potential risks of PCBs pollution were considered widely due to: a) their resistance to degradation, b) their ability of Long Range Atmospheric Transport (LRAT), c) their potential to be bioaccumulated and biomagnificated through the food webs, and d) their potential toxic effects such as carcinogenicity, neurotoxicity and developmental toxicity [1]. Nowadays, PCBs pollution is an extremely urgent problem under the influence of climate change and human activities, with wide consequences for life and ecosystem health [2]. The variation of meteorological factors such as temperature, precipitation, wind speed and evaporation influence directly or indirectly the PCBs environmental fate and mobilization [3]. Increasing temperature can affect the many processes related to PCBs transport: emission rates from primary and secondary sources, the air-surface exchange, the partition between soil, sediment, water and atmosphere, reaction rates [4]. Previous studies suggested that future increases in temperature could reduce PCB contents in the environment but enhance their potential for LRAT from Venice lagoon [1, 5]. The impact of climate change on PCBs at a lake catchment has not been extensively studied. There is a challenge to study the influence of climate change on PCBs pollution at a lake on a long time series with the absence of historical monitoring data [6]. Lake sediment cores, as a store or final destination for PCBs and other pollutants, provided a long-term perspective of historical PCBs pollution [6].

Recent study has confirmed the interactive impacts of climate change and anthropogenic activities on PCBs environmental behaviors in regional scale [7], however, few researchers could clearly quantitatively explain the contribution of climate change and human activities to the deposition of PCBs in lake sediments over long-term time-series. As the relationship between climate change and anthropogenic activities are controlled through complex interactions, significant knowledge gaps about how to separate and quantify the environmental driver's impacts remain [8]. Recently, generalized additives models (GAMs), a non-parametric methods, were applied to separate and quantify the contribution of climate change and human activities on variety of specific environmental indicators [9].

The proposed hypothesis is that climate change and human activities had a synergistic effect on PCBs deposition in lake sediments. The specific objective in the present study were to: (1) reveal the character of PCBs historical disposition and its influential factors; (2) separate and quantify the impact of climate change and anthropogenic activities on PCBs deposition in lake sediments by GAMs.

2. Materials and methods

2.1. Study area

Lake Chaohu is the fifth largest freshwater lake in China, located in the lower reach of the Yangtze River (Figure 1, 31°25′–31°43′N, 117°16′-117°51′E), with a surface area of 780 km^2, a catchment area of 12,938 km^2, and a mean depth of ~3.0 m. The Yuxi River served as an outflow channel for the exchange of water between the lake and the Yangtze River before construction of the Chaohu Dam on the Yuxi River in 1963. The lake has experienced severe eutrophication since 1980s and increasing pollutants input, especially POPs, with the development of urbanization and industrialization [10]. Core C4 was collected from the central area of the western section of the lake.

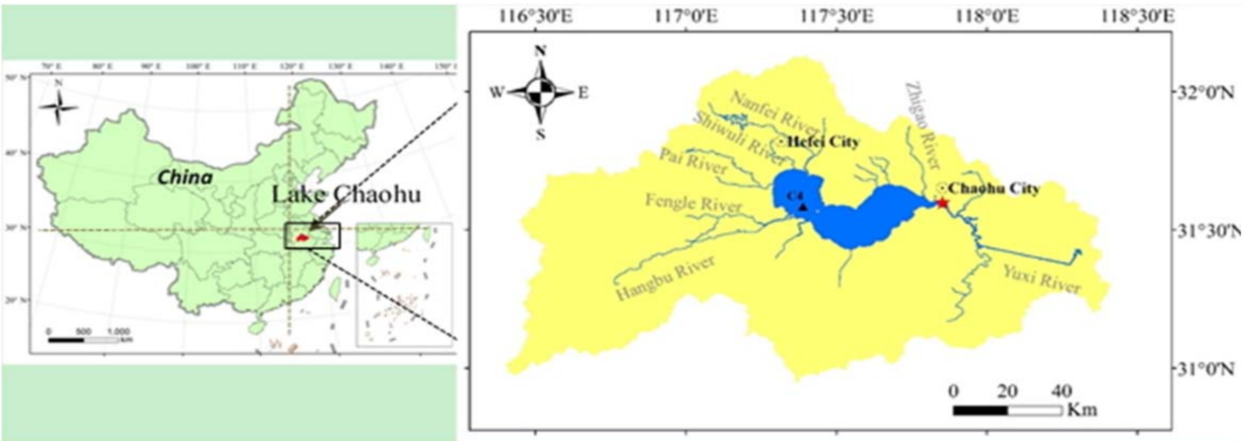

Figure 1. Location of the sampling sites in Lake Chaohu, China.

2.2. Data source

The data of PCBconcentrations in core C4 of Lake Chaohu and sediment dating were collected from Huo et al., [10]. The aquatic total phosphorus (TP) concentrations in Lake Chaohu are from Chen et al. [11], who used sedimentary fossil diatom assemblages to establish a diatom-inferred total phosphorus (DI-TP) transfer function. Meteorological data for this region was extracted from the CN05.1 data set, which is constructed by the anomaly approach during the interpolation with the horizontal resolution of 0.25° * 0.25° [12].

2.3. Generalized Additive Model

Generalized additive models (GAMs) are effective to address the nonlinear relationship between variables and explanatory variables [9]. The general expression is:

$$g(\mu) = \beta_0 + \sum_{i=1}^{k} s_i(x_i) + \varepsilon \quad (1)$$

where μ is the response variable, β_0 is a constant intercept term, $s_i(x_i)$ is the smoothing function of explanatory variable x_i, and ε is the residual.

GAMs were utilized to investigate relationships between PCBs deposition and two influential factors. One of the influential factors was climate change, represented by annual average air temperature (Tem), annual precipitation (Pre), annual average wind speed (Win) and annual evaporation (Evp). The other influential factors were anthropogenic activities, expressed as annual population and Gross Domestic Product (GDP) in Lake Chaohu region, and aquatic TP. Profile of concentrations of sedimentary Sum-PCBs (PCB17, 18, 28, 31, 33, 44, 49, 52, 70, 74, 82, 87, 95, 99, 101, 105, 110, 118, 128, 132, 138, 149, 151, 153, 156, 158, 169, 170, 171, 177, 180, 183, 187, 191, 194, 195, 199, 205, 206, 208 and 209), seven indicator PCBs (∑PCB7; PCB28, PCB52, PCB101, PCB118, PCB138, PCB153, PCB180), tri-PCBs (PCB18, PCB17, PCB28+31, PCB33), tetra-PCBs (PCB52, PCB49, PCB44, PCB74, PCB70), penta-PCBs (PCB59, PCB101, PCB99, PCB87, PCB110, PCB82, PCB118, PCB105) and nona-PCBs (PCB208, PCB206) were selected as the response variables to build six GAMs models. Collinearity diagnostics, the Akaike information criterion (in term of lowest AIC) and generalized cross validation (GCV) score were used to select the predictors and decide on their entry to or exclusion from the model, with collinear factor <10, P<0.1. All six GAMs models were performed by the 'mgcv' package in R. SigmaPlot 9.0 was used for plotting.

3. Results and discussion

3.1. PCBs historical disposition and its influential factors

Total concentrations of all 41 PCB congeners (Sum-PCBs) in the C4 core ranged from 0.03-5.06 ng g^{-1}. Sum-PCBs in C4 core presented three stages, from bottom to top: (1) PCB concentrations in bottom sediments are quite low (< ~125 ng g^{-1}/TOC) from ~1900 to ~1950, (2) then exponential increase of concentrations, peaking in the period between 1975 and 1995, (3) and a gradual decrease of concentrations towards the sediment-water interface [10]. The profile of variation of PCBs is basically consistent with its use and restriction time. China started to produce PCBs in 1965 and stopped production in 1974, during that time about 9000 tons of tri-PCBs and about 1000 tons of penta-PCBs were produced, for use mainly in capacitors, transformers, coatings and paint additives [13]. Meanwhile, the sedimentary PCBs increased significantly in 1970s to 1980s, which reflected the participation of PCBs in the region from anthropogenic activity. By-products of PCBs from dyestuff processing and waste combustion were still discharged into the environment [14]. Therefore, after the ban of PCB containing industrial products, the sedimentary record of PCBs continued to rise until the 1990s and gradually began to decline.

Noteworthy, anthropogenic activities have a direct impact on PCBs production, while climate change plays an important roles in PCBs environmental behavior and fate [1, 3]. The significant variation of average air temperature (Tem), annual precipitation (Pre), annual average wind speed (Win) and annual evaporation (Evp) were observed during 1961 to 2016 in Lake Chaohu catchment (p<0.05). Tem and Pre increased by 0.9845°C and 148.08 mm respectively from 1961 to 2016 in this catchment (Figure 2 a, b). Win and Evp decreased by 1.1275 m/s and 267.91 mm respectively from 1961 to 2016 in this catchment (Figure 2 c, d). Increasing temperature and evaporation probably enhanced the volatilization from primary and secondary sources [3, 5]. An increase of rainfall can cause an air PCBs wash-out by wet deposition, and wind speed influenced the transport of PCBs in atmosphere [4]. The impact mechanism of meteorological factors on PCBs deposition are interactive and complex to separate and quantify [5].

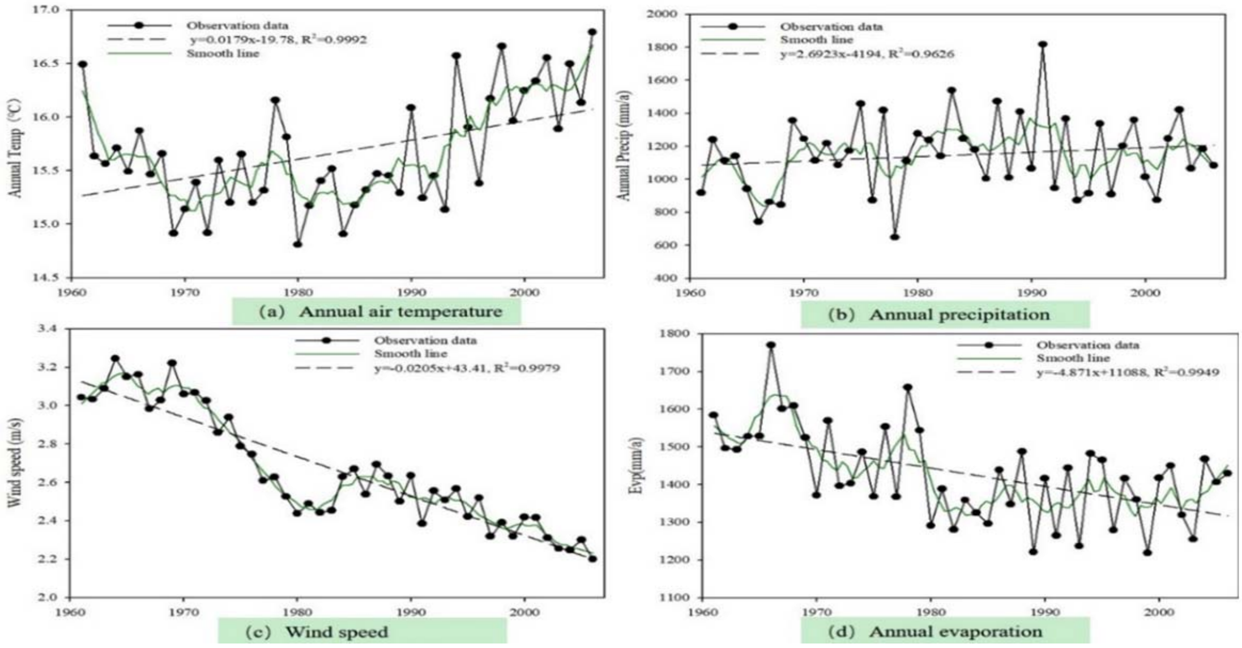

Figure 2. Time series of reconstructed meteorological factor in Lake Chaohu region.

3.2. Contribution of climate change and human activities to PCBs deposition

GAMs analysis was applied to estimate when and to what extent the climate change and anthropogenic activities attributed to the variation of sedimentary PCBs deposition [9]. Tem and Evp were selected as climate change factors. GDP and TP expressed an anthropogenic activitiy factors. The six GAMs models were presented in Table 1. The contribution of predictor variables to sum-PCBs, ∑PCB7, tetra-PCBs and penta-PCBs was, in order TP>GDP>Evp>Tem. The most significant predictor variable for sum-PCBs, ∑PCB7, tri-PCBs, tetra-PCBs and penta-PCBs was TP ($P<0.001$) with intensified anthropogenic activities with increased sedimentary PCBs deposition. Aquatic TP in Lake Chaohu origin from agricultural runoff, industrial and domestic sewage discharge, indicating the intensity of human activities to some extent before environmental management and water pollution control that is considered in recent years [10]. Therefore the decisive influence of human activity intensity on sedimentary PCBs deposition was reflected, though aquatic TP and sedimentary PCBs may not be directly related [15]. While the most significant predictor variables for nona-PCBs was Evp, which is most likely due to the volatility of more-chlorinated homologue groups [16]. Furthermore, the average contribution value of the sum of Tem and Evp to PCBs (7.1%) was larger than that of the sum of TP and GDP to PCBs (17.9%), suggesting that anthropogenic activities explained a larger propotion of sedimentary PCBs deposition than climate change.

Table 1. GAMs analysis results for PCBs deposition in Lake Chaohu.

Response variables	Models	Contribution of predictor variables (%)				P	R^2	Deviance explained
		Tem	Evp	GDP	TP			
Sum-PCBs	g(TPCBs) ~ s(Tem) + s(Evp) + s(GDP) + s(TP)	1.18	1.40	7.20	9.93	<0.001	0.968	97.60%
∑PCB7	g (PCB7) ~ s(Tem) + s(Evp) + s(GDP) + s(TP)	1.55	1.60	3.22	12.43	<0.001	0.938	95.50%
tri-PCBs	g(triPCBs) ~ s(Tem) + s(Evp) + s(GDP) + s(TP)	3.22	3.00	3.85	8.25	<0.001	0.86	87.80%
tetra-PCBs	g(tetraPCBs) ~ s(Tem) + s(Evp) + s(GDP) + s(TP)	3.84	4.26	4.40	11.18	<0.001	0.871	88.90%
penta-PCBs	g(pentaPCBs) ~ s(Tem) + s(Evp) + s(GDP) + s(TP)	1.95	2.19	5.47	17.14	<0.001	0.936	95.30%
nona-PCBs	g(nonaPCBs) ~ s(Tem) + s(Evp) + s(GDP) + s(TP)	1.10	16.73	14.74	9.46	<0.001	0.963	97.40%

The contribution of the four predictors, TP, GDP, Evp, and Tem, to the variation of Sum-PCBs, ∑PCB7, tri-PCBs, tetra-PCBs, penta-PCBs and nona-PCBs were shown in Figure 3. The contribution of TP grew rapidly before mid-1970s and then fluctuated steadily for the six GAMs models. The negative effect of TP on response variables changed into positive effect from ~1975. This changes were concomitant with significant upsurge of TP input in Lake Chaohu around 1975 caused by urbanization, industrial and agricultural development [17]. The smooth functions clearly illustrated a moderate response of PCBs deposition to TP, GDP, Evp, and Tem (i.e., y-scale axis of the fitted functions; Figure 4). The fitted relationship between sedimentary PCBs and aquatic TP was nonlinear for Lake Chaohu, showing that the sedimentary PCBs increased with phosphorus enrichment as long as TP concentration did not exceed 60-100 μg L^{-1} (Figure 4). These observations suggest that anthropogenic activity intensity promotes PCB deposition in the absence of environmental management.

For all six models, the contribution of GDP increased and peaked at the late 1990s, then decreased sharply. The negative effect of GDP on responses variables changed into positive effect from around 1980, then changed into negative effect again from pre-2000s (Figure 3). The fitted relationship between sedimentary PCBs and GDP was nonlinear for Lake Chaohu, showing that the sedimentary PCBs increased with GDP rise as long as GDP value did not exceed 20 billion and decreased at the highest GDP (> 20 billion; Figure 4). The parabolic relationship can be explained by industrial emissions leading to the increase of sedimentary PCBs in the early stage of social and economic development, while in the later stage, PCBs pollution is reduced through artificial control [13, 14].

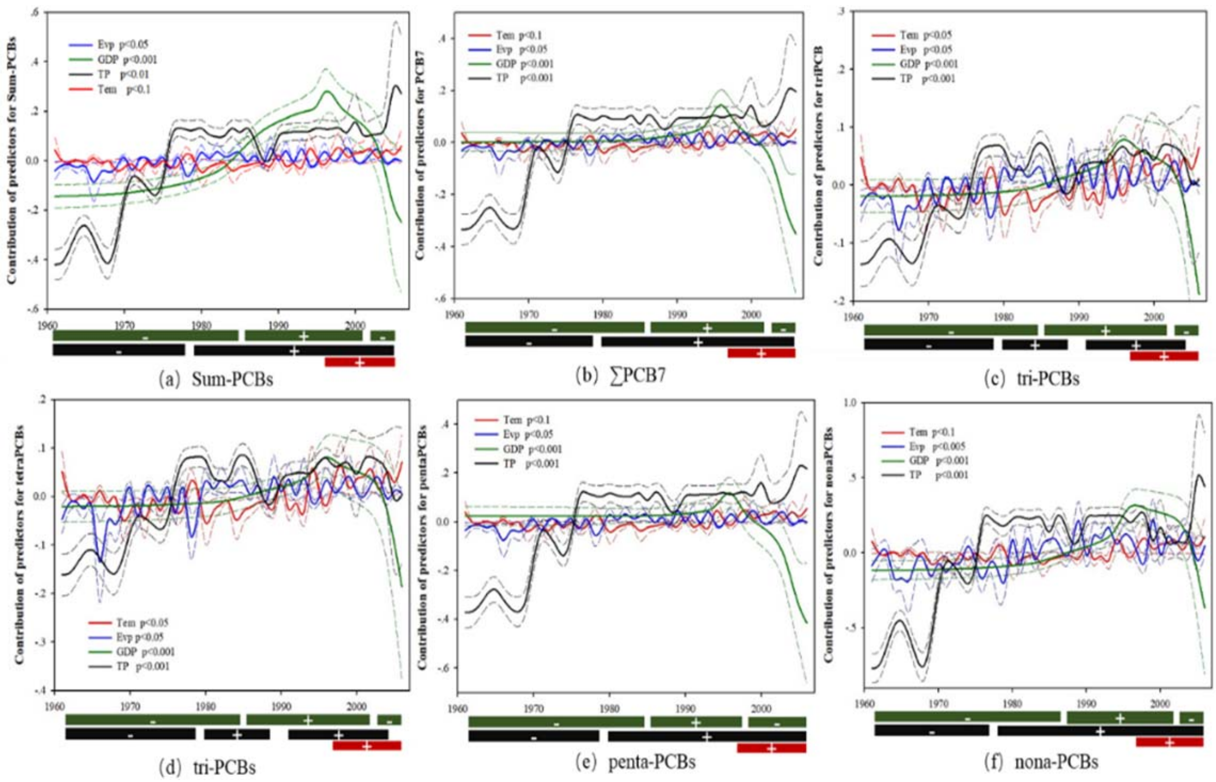

Figure 3. The variations of predictor contribution with time in Lake Chaohu.

The responses of Sum-PCBs, ∑PCB7, tri-PCBs, tetra-PCBs, penta-PCBs and nona-PCBs to Tem were found to be positive since the late 1990s (Figure 3). The fitted relationship between sedimentary PCBs and Tem was linear for Lake Chaohu, showing that the sedimentary PCBs increased with Tem rise (Figure 4). The predominant reason for observation was probably that higher temperatures reduced the fugacity capacity of PCBs in air, water and sediments [15]. Warming also enhanced the volatilization from primary and secondary sources, and diffusive sources to air were stronger [4]. Paul et al. [18] found that the effects of climate induced changes on a sea currents, temperature, wind speed, precipitation. On the fate of PCBs revealed temperature as one of the most influential, suggesting that the increase in temperature could enhance the emission of PCBs primary and secondary sources and lead to alteration in the rates of partitioning, volatilization, degradation and reaction (biodegradation, photolysis and oxidation). Therefore, increased temperature promoted the deposition of sedimentary PCBs in Lake Chaohu, even if complex processes in various media and interface were directly depended on temperature.

The contribution of Evp in all six response variables fluctuated between positive and negative (Figure 3). The fitted relationship between sedimentary sum-PCBs, ∑PCB7, tri-PCBs, penta-PCBs and Evp were linear for Lake Chaohu, while the fitted relationship between tetra-PCBs, nona-PCBs and Evp were nonlinear, showing that the sedimentary PCBs decreased with Evp rise (Figure 4). The larger the amount of evaporation lead to air-water surface exchange and more PCBs volatilization, so the less PCBs deposited into sediments [16]. In general, the environmental fate of PCBs is influenced by the combined effect of variability of many meteorological factors which are difficult to separate [15].

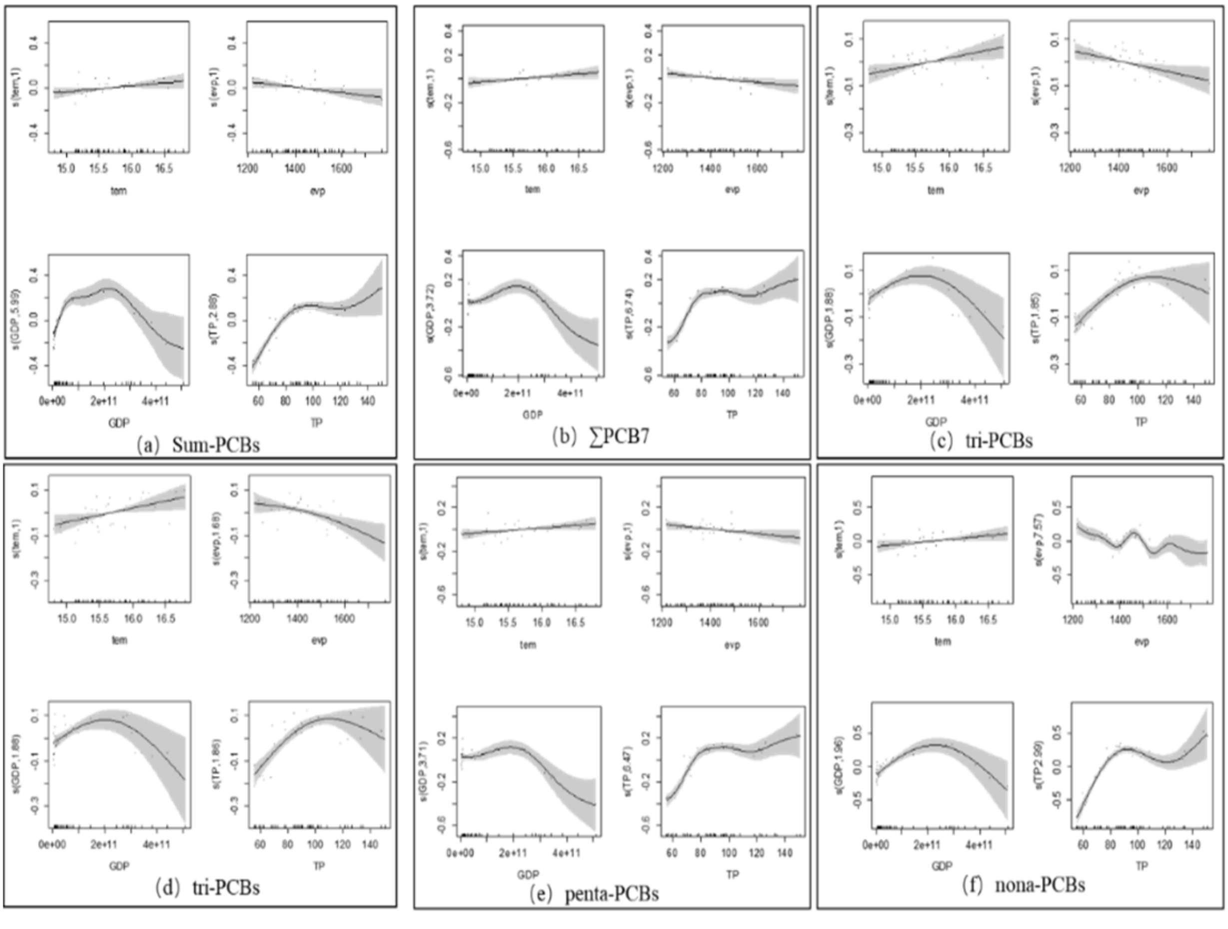

Figure 4. GAMs results for the contribution of predictors in Lake Chaohu.

4. Conclusions

Climate change and anthropogenic activities influenced the deposition of PCBs in Lake Chaohu. The results of GAMs demonstrated that anthropogenic activities explained a larger propotion of sedimentary PCBs deposition than climate change. The contribution of aqatic TP to the proliferation of Sum-PCBs, ∑PCB7, tri-PCBs, tetra-PCBs and penta-PCBs were 9.93%, 12.43%, 8.25%, 11.18%, 17.14%, respectively. These positive effects were observed since ~1975, when the input of TP from human activities increased considerably. The results of the fitted relationship between sedimentary PCBs and TP, GDP indicated anthropogenic activities connected to development of urbanization and industrialization promoted the deposition of PCBs in Lake Chaohu sediments before the consideration of environmental management and water pollution control in recent years. The fitted relationship between sedimentary PCBs and Tem was linear for Lake Chaohu, showing that the sedimentary PCBs increased with Tem rise. The sedimentary PCBs decreased with Evp rise, due to the larger the amount of evaporation lead to air-water surface exchange and more PCBs volatilization. Climate change have a combined influence on PCBs environmental behaviors and fate.

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