Watershed-Scale Phosphorus Balance Evaluation Using a Mass Balance Method

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Abstract. It is crucial for assessing the eutrophication risk of lake by analyzing the phosphorus (P) balance of lake watershed quantitively. A mass balance method was used to calculate P balance of both Yangzonghai lake watershed and the lake itself in one year. The imported P load was 725.1 t in 2010, while the exported P load was 317.3 t, which indicated that 56.2 % (407.8 t yr^{-1}) of P was retained in the lake watershed. Such a high retention load implied that the lake, which was mesotrophic, was under great pressure of further eutrophication. Among all the input pathways, the largest P input contributor was fertilizer, contributing 679.0 t P and accounting for 93.6 % of input P, followed by atmospheric deposition (44.7 t P, 6.2 %). Plant product (264.6 t, 83.4 %) was the largest P output contributor, followed by animal products (50.2 t, 15.8 %).

Keywords: Phosphorus load · Eutrophication · Watershed · Phosphorus balance

1 Introduction

Lake eutrophication is still one of the main water pollution problems in China [1]. Watershed development often increases lake nutrient inputs, leading to lake eutrophication [2]. Excessive lake P input is a major driver of algal blooms in lakes [3, 4]. Phosphorus (P) balance of lake watershed is important to determine if the lake is threatened by eutrophication. When there was more P imported into the watershed than exported, the redundant P retained in the watershed will flow into the lake and may finally result in lake eutrophication, or it will flow into lake through soil erosion [5], increasing the potential of lake eutrophication.

Yangzonghai Lake, a deep-water lake, which located in southwest China is working as a drinking water supply (before 2008) besides providing agricultural and industrial water. Due to nutrient overload, Yangzonghai Lake turns to be mesotrophic currently, which has a potential to threat the safety of drinking water, this is tightly

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related to 60,000 people's health [6]. There are few previous studies reported the lake total P balance. However, it is important to draw up P controlling measures through deeply understanding of the P balance, especially the P flux of each input pathways.

Scholars have made much effort to study P input and output from lake watersheds. P inputs are presented in the form of atmospheric deposition, fertilizer, and other products. P outputs are presented in the form of harvested crops, livestock and its products, and the lake outlet [2, 7–9]. In this study, we identified the P input and output ways of the Yangzonghai Lake on watershed scale, and calculated the P balance of the watershed through mass balance methods, which will provide helpful information to control the lake eutrophication.

2 Materials and Methods

2.1 Study Area

Yangzonghai Lake (102°5′–103°02′E, 24°51′-24°58′N), with an average water area of 3,160 ha and catchment of 19, 200 ha. Has an elevation of 1,769.9 m. The volume of the lake is about 5.69 × 10^8 m^3 [10] and the main soil type is latosol. There are three inflow streams and one channel called Baiyi River outflow from the watershed, but only one outflow river called Tangchi River which is controlled by a sluice gate in the north (Fig. 1). There are 58,743 people living in the catchment (in 2010), most of whom are farmers. Agricultural activities are the main anthropogenic factors in the catchments, and there are only four industrial enterprises (Fig. 1, Brilliant Resort & Spa, Spring City Golf, Yangzong Thermal Power Plant and Yunnan Aluminum Factory) in which the

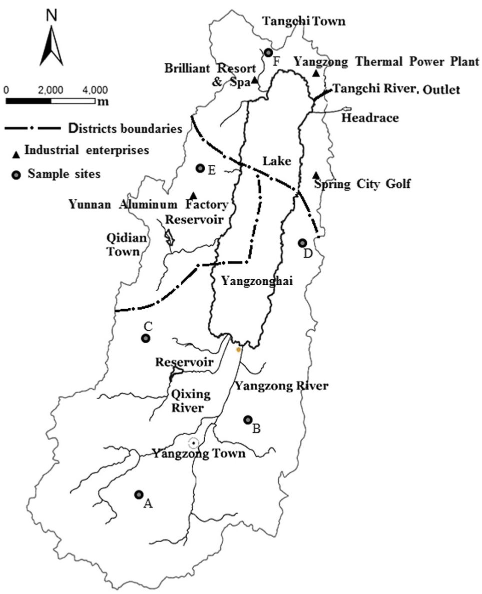

Fig. 1. Map of the Yangzonghai Lake catchment with sampling sites.

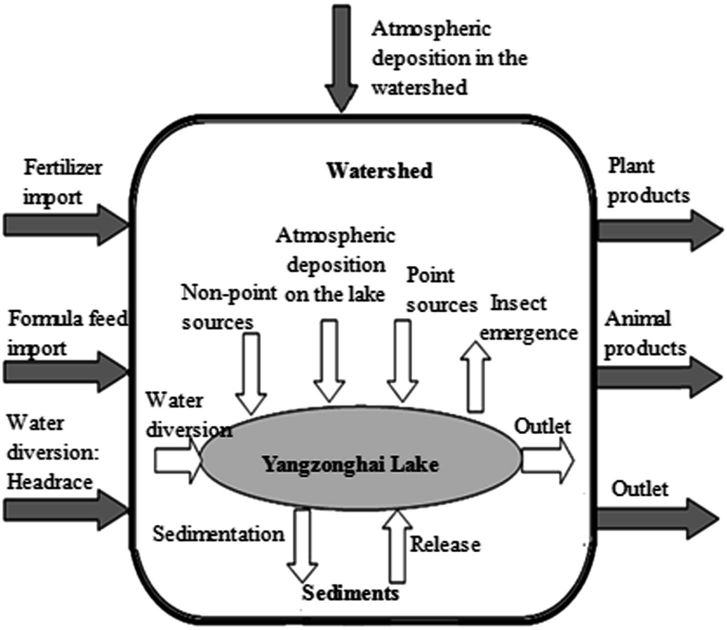

Fig. 2. Concept model of phosphorus balance in the Yangzonghai Lake watershed. In the diagram, solid arrows indicate the phosphorus transfer between the watershed boundaries, and hollow arrows indicates the phosphorus transfer between the lake subsystem boundaries.

sewage discharges into wastewater treatment plants and the treated sewages is discharged into Tangchi River and outflow from the watershed, not into the lake. For convenience, we divided the watershed into three districts: Yangzonghai District, Tangchi District, and Qidian District.

2.2 Methodology

We used a mass balance method [8, 11, 12] in this study to calculate P balance of the watershed which was regarded as a whole in this study and the lake itself in one year (2010). The watershed boundary is the natural catchment area, not including the watershed form where water is channeled. The concept model of the watershed is shown in Fig. 2. In 2010, the P input/output pathways of the watershed in Fig. 2 were studied. The specific methods are described below:

P Input Pathways on Watershed Scale. As Fig. 2 shows, the input pathways taken into consideration in the study included atmospheric deposition, fertilizer use, water diversion, formula feed input.

P Input Through Atmospheric Deposition. In order to calculate the P load from atmospheric deposition, we selected 6 sites (Fig. 1A–F) in the watershed and placed three dust collecting cylinders in each site to collect the total deposition samples for one year. The collecting cylinder is a cylinder with height of 500 mm and a mouth area of 0.0143 m^2. The cylinders were immersed with hydrochloric acid solution before collecting deposition samples. The samples were collected in terms of national standard method of China GB/T15265-94, and analyzed by an ammonium molybdate spectrophotometric method (GB11893-89) once a month. The P load from atmospheric deposition was determined as Eqs. (1) and (2):

$$C_{i2} = [(C_{i1} \times V)]/s. \tag{1}$$

$$P_{ad} = \sum_{i=1}^{n} C_{i2}/10 \times S. \tag{2}$$

where C_{i1} is the P concentration of deposition sample (mg L^{-1}), V is the volume of the sample, s is the area of the cylinder (m^2), C_{i2} is the mass of total P deposited per square meter (mg m^{-2}), Pad is the total mass of P deposited on the whole watershed (t yr^{-1}), and S is the area of the watershed (ha).

P Input from Fertilizer Use. Based on the investigation of chemical fertilizer used in the watershed, including the application amount and P content of each fertilizer type, *we* calculated the P contribution from fertilizer use by using the Eq. (3) below:

$$F_P = \sum_{i=1}^{n} Q_i \times C_{iP} \times 100. \tag{3}$$

where FP is the P load from chemical fertilizer, Q_i is the application amount of type i fertilizer, and C_{iP} is The P content of type i fertilizer.

P Input Through Water Diversion. The water flow from water diversion channels was continuously monitored by a hydrometric station, and the water quality was analyzed monthly by the local environmental monitoring institution, making it easy to obtain the P input load from water diversion.

P Output Pathways on Watershed Scale. As Fig. 2 shows, the output pathways identified in the study included plant products, animal products, outlet.

P Output Through Plant Products. P exported from one plant product was calculated by the output amount (data provided by the local government) multiplied by the P content of the product. We calculated the total P mass exported from plant products (including food products) by adding the P output of each product. In this study, the P content of the products was determined in terms of the recommended analyzing method of the China Ministry of Agriculture (NY/T1018-2006, the number of replications for each plant products ≥6). The plant products consumed by humans who lived in the watershed are not included.

P Output Through Animal Products. The P exported from one animal product was calculated by the output amount (data provided by the local government) multiplied by the P content of the product. We calculated the total P mass output from animal products by adding the P output of each animal product. In this study, the P content of the main animal products was determined according to the recommended analysis method of the China Ministry of Agriculture (NY/T1018-2006, the number of replications for each animal product ≥6). The animal products consumed by people who lived on the watershed were excluded.

P Output Through Outlet. The water from the outlet was continuously measured by a hydrometric station, and the effluent water quality was monitored monthly by the local environmental monitoring institution. We calculated data each month by multiplying the water flow of each month with the water total P concentration of the month, and obtained the total P output from the outlet by adding all of the data of 12 months.

P Input and Output of the Lake. We identified the P inflows and outflows pathways of the lake and calculated the P flux of each pathways (Fig. 2). The input pathways included sediment release, water diversion, atmospheric deposition, point pollution source input, and non-point pollution source input (Fig. 2). The outflows pathways included outlet, sedimentation and insect emergence [13] (Fig. 2). Similar methods were used to calculate P fluxes of lake through water diversion, atmospheric deposition, outlet. We calculated P load which flowed to the lake in terms of different pollutant types. Point source and non-point source P load was quoted from Yang's research [14]. The P atmospheric deposition was calculated by using Eq. 2, in which S means the surface area of the lake (i.e. S = 3,160 ha). Methods about how to calculate P fluxes through anaerobic sediment release and sedimentation were described below:

P Imported to Lake Through Anaerobic Sediment Release. The P flux from sediment was measured by an experiment carried out in the laboratory (n = 15). Sediments and overlying water were collected from the bottom of the lake (liquid: solid ratio of 20:1, i.e. 4000 mL water: 200 g sediments). N_2 was aerated into a closed glass box to keep environment anaerobic (Dissolved Oxygen <3.0), making sure that it is similar to the bottom environment of lake. The net P release fluxes ($P_{release}$, t) experiments were carried out from February 1, 2010 to January 31, 2011. The P concentrations of overlying water was tested in the 1st day ($C_{background,}$ mg L^{-1}) and then measured once each 15 days, details about analyzing method of P concentration are deccirbed above. The equations we used are described below as Eqs. (4) and (5):

$$C_{Change} = C_{max} - C_{background}. \tag{4}$$

$$P_{release} = \left(C_{Change} \times V_{water}\right)/\ S_{interface} \times S_{lake} \times 10^{-9}. \tag{5}$$

where C_{Change} is changes of P concentrations in the overlying water between the maximum P concentration and the background P concentration (mg L^{-1}); C_{max} is the maximum P concentration value during the whole experiment (mg L^{-1}); V_{water} is the volume of overlying water in experiment (L); $S_{interface}$ is the area of the interface in simulated device (m^2); S_{lake} is the area of lake (m^2).

Most of the basin is a rural area, the residents on the basin lived a self-sufficient life. According to the information provided by the local government, food imported from outside the basin was negligible and unrecorded; human flows out of the basin and flows in the basin were substantially balanced in 2010; there was little formula feed input from outside the watershed. Thus, the P input from formula feed could be ignored in this balance calculation of Yangzonghai Lake watershed in 2010.

P Exported from Lake Through Sedimentation. The P exported via sedimentation of the lake was calculated by using a mass balance method: inputs - outputs = change in storage of lake water. The formula of P exported from sedimentation (Qs) is as Eq. 6:

$$Q_s = Q_{in} - Q_{outlet} - Q_{IM} - \Delta P. \tag{6}$$

where Q_{in} is the sum of the P load entering into the lake in 2010, Qoutlet is the P load exported from the outlet, and Q_{IM} is the P load exported from insect emergence (to the lake subsystem, the P export pathways are just from sedimentation, outlet, and insect emergence, Fig. 2). ΔP is the P increment in the lake water, which is the difference of P in lake water between December 2009 and December 2010. The water samples from each site were collected at different depths using a water pump (the first at 0.5 m below the surface, then every 1 m down the water column, and the last sample at 0.5 m from the bottom).

Due to the lake was polluted by arsenic in 2008 and was treated with a ferric flocculants ($FeCl_3$) from October 2009 to December 2011, see Liu et al. [10] in details. The P flux of sedimentation has been accelerated by the remediation of arsenic pollution. The Fe-induced sedimentation of P was calculated by establishing relationship model between the amount of arsenic sedimentation and P sedimentation in the overlying water when added the flocculants ($FeCl_3$). The model was established by an experiment carried out in the laboratory (n = 102) in August 30–31, 2012. Two parts of overlying water (8 Barrels × 20 L for each part) were taken from the bottom of the lake in August 30, 2012 in southern and northern of the lake, respectively. Measured the concentration of P and arsenic for each part, then the water samples were poured into 102 glass bottles (3 L for each bottle). Added different amount of the flocculants ($FeCl_3$) into the bottles, sampled and measured the concentration of P and arsenic in the water in the next day and calculated the amount of arsenic and P sedimentation at the same bottle, and gained 102 arsenic and phosphorus corresponding data. Then we established the relationship model. The amount of arsenic removal from the lake in 2010 was 48.43 t (the data estimated by the local environmental monitoring institution). We calculated the amount of P sedimentation induced by iron based on the model and the data of arsenic removal from the lake in 2010. The P concentration analyzed method see above mentioned. The concentrations of arsenic were determined using a 933AFS atomic fluorescence spectrometer (Titan Instruments, Beijing, China; detection limit = 0.5 $\mu g\ L^{-1}$).

Statistical Analysis. SPSS software version 16.0 was used to analyze the data. All comparisons were carried out using analysis of variance (ANOWA, when sample sizes were the same) and General Linear Model (GLM, when sample sizes were different). A significance level of 0.05 was used in the analysis.

3 Results

3.1 P Input of the Yangzonghai Lake Watershed

P Input through Atmospheric Deposition. The total atmospheric deposition P input was 44.7 t in 2010 (44.7 t yr^{-1}). The area weighted flux was 2.3 kg ha^{-1} yr^{-1}. The average monthly flux was 3.7 t. The atmospheric deposition was relatively high in the beginning of the rainy season (Fig. 3).

P Input through Fertilizer Use. According to the investigation, the main P fertilizer applied to the watershed was $Ca(H_2PO_4)_2{\cdot}CaHPO_4$, a compound fertilizer (N:P:K, 10:7:8) and a fertilizer specially used on golf courses (Table 1). About 679.0 t P was imported into the watershed in 2010 by fertilizer use.

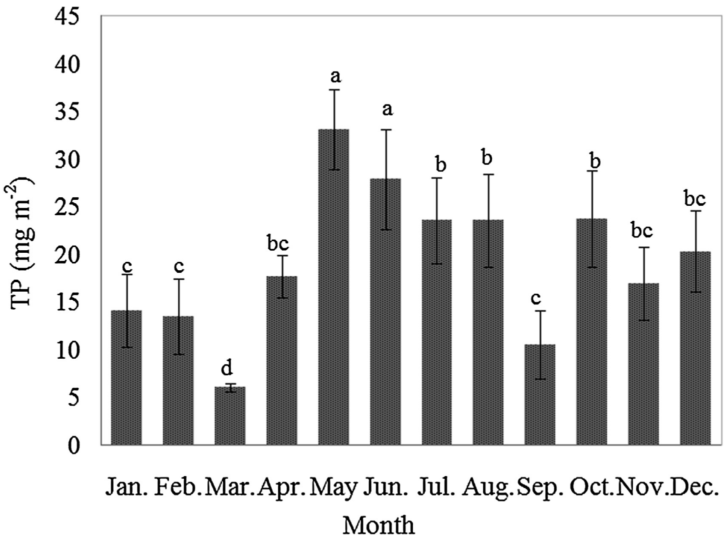

Fig. 3. Trend of phosphorus atmospheric deposition. The different letters displaying the quantity of phosphorus deposition between two months indicated a significant difference (i.e. $p < 0.05$), according to the multiple-comparison tests carried out. Vertical bars are standard deviations (n = 9).

P Input through Water Diversion. The water quality of the diversion water outside the watershed varied seasonally. The worst water quality appeared in September (0.331 mg L^{-1}, Fig. 4A). According to monitoring data, 5,676,566 m^3 of water was from the Baiyi River water diversion in 2010 (Fig. 4B), and it contained 1.4 t P (Fig. 4C), which flowed into the lake. In addition, P from anaerobic sediment source was 1.9 t yr^{-1}.

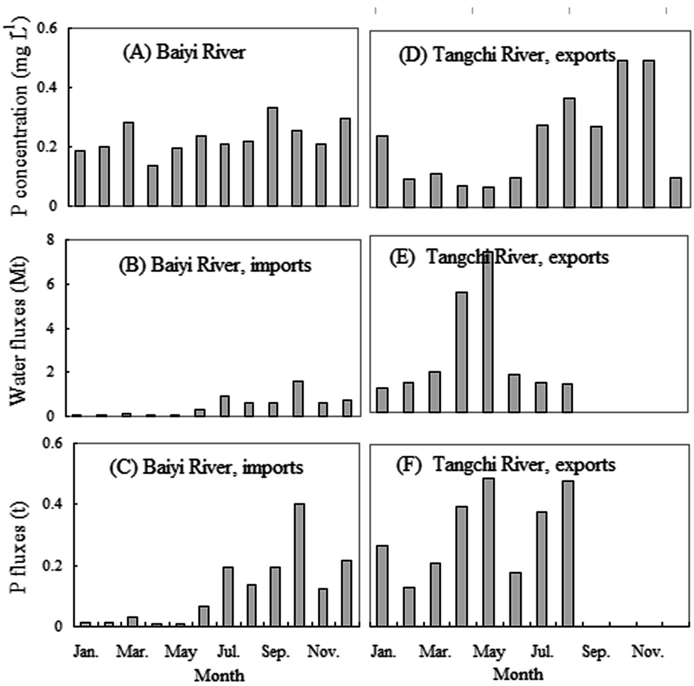

Fig. 4. The amount of imported TP from the Baiyi River (A, B and C) and that of exported TP from the Tangchi River (D, E and F) in 2010.

Table 1. The amount of fertilizer used, imported TP and exported TP in Yangzonghai Lake watershed in 2010.

District		Yangzong	Qidian	Tangchi	Total
$Ca(H_2PO4)_2 \cdot CaHPO_4$ fertilizer, P content: 25.128 %	Fertilizing amount (t)	1565	17	233	1815
	P (t)	393.2	4.3	58.5	456.1
Compound fertilizer (N:P:K, 10:7:8) P content: 7 %	Fertilizing amount (t)	2582	28	436	2976
	P (t)	180.7	2	30.5	213.2
Golf fertilizer, P content: 15 %	Fertilizing amount (t)	–	–	65	65
	P (t)	–	–	9.7	9.7
P imported in total (t)		573.9	6.3	98.8	679
P-output from plant products (t)		203.24	55.04	6.35	264.63
P-output from animal products (t)		23.26	23.08	3.87	50.21
P exported in total (t)		226.5	78.12	10.22	314.84

3.2 P Output from the Yangzonghai Lake Watershed

P Output through Plant Products. The amount of P exported from the watershed by plant products (including food products) was 264.6 t in 2010 (Table 1). Among all the output pathways, plant products contributed the most (264.6 t yr^{-1}). Among plant products, timber exported 199.4 t yr^{-1} and accounted for 72.6 %, rice exported 38.6 t yr^{-1} and accounted for 14 %, corn exported 19.5 t yr^{-1} and accounted for 7.1 %, and other plant products accounted for 6.3 %.

P Output through Animal Products. The amount of P exported from the watershed by animal products (meat, egg, milk, etc.) was 50.2 t in 2010 (Table 1). Animal products ranked second in all output pathways (50.2 t yr^{-1}). Among animal products, pork exported 31.1 t yr^{-1} and accounted for 59.6 %, eggs exported 7.0 t yr^{-1} and accounted for 13.4 %, poultry exported 5.2 t yr^{-1} and accounted for 10 %, milk exported 3.7 t yr^{-1} and accounted for 6.3 %.

P Output through Outlet. There was 21,389,702 m^3 of water flowed out form the outlet and 2.5 t P in 2010. Variation of P concentration in water, water fluxes and P fluxes of the outlet could be observed in this period of time (Fig. 4D–F).

P Input and Output of the Lake. P retention in lake was 23.4 t at the end of 2009, and was 18.0 t at the end of 2010, so the increment was −5.4 t. By calculation with the mass balance method using Eq. 4, we found that P exported from sedimentation was 44.5 t in 2010. The P budget of the lake subsystem is shown in Fig. 5. As for the lake, non-point sources contributed the largest P input, accounting for 77.7 % of the total P, and sedimentation contributed the largest P output, accounting for 94.6 % of the total P.

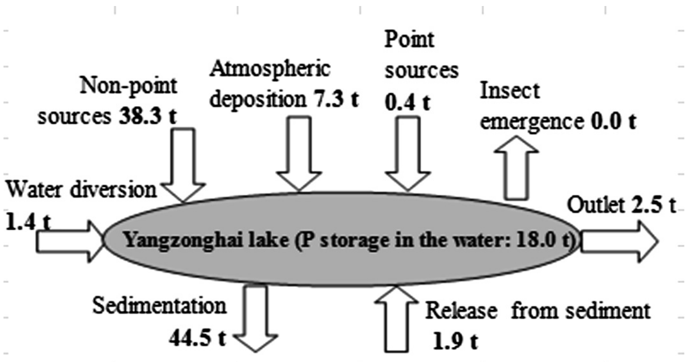

Fig. 5. P budget of the Yangzonghai Lake subsystem in 2010

When adding flocculants ($FeCl_3$) into the lake, the concentrations of P and arsenic were decreased simultaneously ($y = 5.9464x + 0.0006$, $R^2 = 0.9946$, $n = 102$, $p < 0.05$). The Fe-induced sedimentation of P was 7.9 t yr^{-1} based on the relationship model between the amount of P sedimentation and arsenic sedimentation in the water when

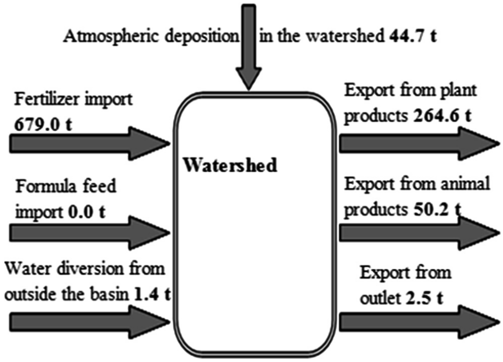

Fig. 6. P budget of Yangzonghai Lake watershed

adding different amount of $FeCl_3$ ($y = 6.1557x + 9E^{-05}$, $R^2 = 0.9946$, $n = 102$, $p < 0.05$) and the amount of arsenic removal from the lake in 2010 (48.43 t).

3.3 P Budget of the Watershed

The imported P load was 725.1 t, and the exported P load was 317.3 t in 2010, which indicates that 407.8 t P was retained in the watershed in this period of time (Fig. 6). Among all the input pathways, fertilizer use ranked first, contributing 679.0 t P and accounting for 93.6 % of input P. Then followed by atmospheric deposition, which contributed 44.7 t P, accounting for 6.2 % of input P. For output pathways, plant products ranked first, exporting 264.6 t P and accounting for 83.4 % of output P. Animal products ranked second in all output pathways (50.2 t yr^{-1}), accounting for 15.8 % of output P.

4 Discussion

4.1 P Budget of the Watershed

Many studies that used a mass balance method have revealed that there is imbalance between P input and output of lakes on watershed scale, and all of the researchs have found that it will lead to P accumulation in the watershed when inputs exceed outputs [2, 7, 15, 16]. This is consistent with Yangzonghai Lake watershed. The P retention rate in the lake watershed was 56.2 % in 2010, which implies more than half of the P inputs was retained in the watershed. The retention rate is comparable with the upper Potomac River basin (60 %, [7]) and Lake Okeechobee (80 %, [16]).

The long term accumulation of P in the watershed will finally result in lake eutrophication [2]. Yangzonghai Lake is a deep lake with an 11.1 year-long water renewal period [17]. The P concentration in the lake has been rising up in the last six years [6], which means that the risk of eutrophication is increasing. Therefore, watershed governors should seek to draw up strategies which can keep the balance of inputs and outputs (or outputs > inputs) in the watershed. Besides reducing fertilizer use, it is necessary to construct wetlands in the riparian zone to decontaminate nutrients in surface runoff on Yangzonghai Lake watershed.

4.2 P Input Fluxes of Watershed and Lake

The average P inflow of the watershed was 37.8 kg ha^{-1} yr^{-1}, and that of Yangzonghai Lake was 15.6 kg ha^{-1} yr^{-1}, which was 2 times higher than that of the eleven recreational lakes in Minnesota State of the United States (0.32–6.0 kg P ha^{-1} yr^{-1}, [2]). Non-point source pollution from agriculture caused river and lake pollution problems in United States, and the control of P from agriculture is still the key aspect in American water pollution treatment [18]. Pollution from agriculture is also responsible for much of the water pollution and drinking water shortages in China. In Yangzonghai Lake watershed, the main P inflow was from fertilizer application, which contributed 679.0 t and accounted for 93.4 % of the total in 2010. In terms of the lake P budget, fertilizer application contributed 65.1 % of the total P input. Cultivated land account for 27.7 % of the watershed, as the crop farming ecosystems, the P budget strongly relied on P fertilizer use [19]. Therefore, it is the key countermeasure to control the fertilization intensity in the watershed to keep the lake from further eutrophication, which can be done by improving fertilizer use efficiency or changing land use from planting nutrient exhaustive crops to poor resistance crops.

Atmospheric deposition is also a P input pathway which cannot be ignored. The Yangzonghai Lake watershed was 44.7 t yr^{-1}, accounting for 6.2 % of the total in 2010. In the studies of the Lake Simcoe in Canada, Winter et al. [20] found that P from atmospheric deposition accounted for 23–56 %. Yang et al. [21] studied the P contributed by atmospheric deposition in Tai Lake, China, and found that the P load from atmospheric deposition accounted for 46.2 % of the total load.

The P deposition in the Yangzonghai Lake watershed was in the medium level compared with other watersheds which had been reported in China. The monthly average P deposition load in the Yangzonghai Lake watershed in 2010 was 0.192 kg ha^{-1} $month^{-1}$, which was lower than that in Xingyun Lake watershed (0.370 kg ha^{-1} $month^{-1}$, [22]) and in the Tai Lake watershed, East China, in 2007 (0.230 kg ha^{-1} $month^{-1}$, [21]), but it was three times higher than that in the Lake Fuxian watershed (0.066 kg ha^{-1} $month^{-1}$, [22]), a nearby lake in the same province. The coal-fired power plant beside Yangzonghai Lake perhaps accounted for the significant difference.

The P from lake sediment release was 1.9 t yr^{-1}, contributing only 3.9 % to the P input (49.3 t yr^{-1}). This indicates that sediment dredging would not solve lake internal problems related to eutrophication, the results are similar to Shahe Reservoir's [1]. Input flux of P from water diversion was 1.4 t yr^{-1} in 2010, much smaller than other inflow fluxes. But taking consideration of its high P concentration (between 0.135 - 0.331 mg L^{-1}), the influence of water diversion should not be neglected. The imported P load could result in lake P concentration increasing 0.0028 mg L^{-1}. Thus, it is quite necessary to purify the channeled water before it enters into the lake.

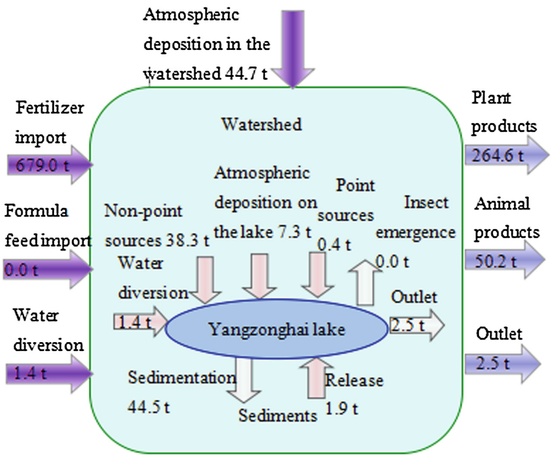

Fig. 7. The amount of P flux for each way in watershed-scale and lake-scale of Yangzonghai Lake

5 Conclusions

The amount of P flux for each way in watershed-scale and lake-scale of Yangzonghai Lake see Fig. 7, and P fluxes between inputs and outputs in Yangzonghai Lake watershed were imbalanced; 56.2 % (407.8 t) of input P was retained in the watershed. Such a high retention rate implies that the lake, which was on mesotrophic level, has a great pressure of further eutrophication. Among all the countermeasures to control the eutrophication of the lake, reducing the fertilizer application should be put first.

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Conflicts of Interest

State any potential conflicts of interest here or “The authors declare no conflict of interest”.

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