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Composition and flux of nutrients transport to the Changjiang Estuary

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Based on the results of water sample measurements of nutrient concentrations at the Datong Station of the Changjiang River from 1998 to 1999, combined with historical data of water quality, seasonal variations of nutrient concentrations and nutrient transports are discussed.The following results have been obtained: (1) the fluxes of the nitrate nitrogen, ammonium nitrogen and nitrite nitrogen increased by time-series from 1962 to 1990, even if runoff volume had a little variation; (2) the concentrations and fluxes of the dissolved inorganic nitrogen (DIN) and dissolved inorganic phosphorus (DIP) increased notably with time, but those of the dissolved silicon (DSI) decreased pronouncedly; and (3) the concentrations and fluxes changed synchronously with time between the Datong Station and the Changjiang Estuary.

Composition and flux of nutrients transport to the Changjiang Estuary ZHANG Shen, JI Hongbing, YAN Weijin, DUAN Shuiwang (Inst. of Geographic Sciences and Natural Resources Research, Beijing 100101, China) Abstract: Key words: CLC number: 1 Introduction Rivers play an important role in transport of nutrients (e.g., N and P) and their biogeochemical cycling in land-ocean interaction system. For example, human activities have resulted in about the same amount of nitrogen fixation (140×106 t) on continents as that of nature through fertilizer production, energy production and cultivation of crops, or 30% of the total amount transported to oceans by rivers (Galloway et al., 1995). The fluxes of dissolved phosphorus from land to sea surface carried by river runoff as orthophosphate is about 2 Tg P year-1 (1 Tg = 1×1012 g) (Chameides and Perdue, 1997). At the same time, rivers are hydrologically linked to estuaries and therefore an understanding of the effects of human intervention on nutrient fluxes in the landscape is important for improved understanding and management of nutrient loads on estuaries (Correll et al., 1992). The Changjiang River is the largest one in China, and the third largest one in the world, which carries large amount of water (924×103 km3), sediment (453×109 kg) and nutrients to its estuary and the seas. In the beginning of the 1980s, nutrient levels were still low in waters of the Changjiang River (Meybeck, 1982), but they have increased drastically in recent years (Zhang et al., 1995; Chen et al., 1998; Duan et al., 2000). And the algal booming had become a serious problem in the Changjiang estuary and coastal waters (Jin et al., 1996; Su, 2001). So, the studies on nutrient transports from the Changjiang River to estuary provide useful information not only for global nutrient cycling and budget, but also for the nutrient management of coastal waters of China. According to the nitrogen budgets, the dominant outputs of nitrogen include nitrogen denitrification, ammonium volatilization, storage nitrogen and river runoff export nitrogen (McKee and Eyre, 2000). The riverine exported nitrogen takes an important part in the nitrogen budgets. For example, the river runoff export nitrogen occupies 25% of the total nitrogen inputs to the Changjiang River during the 1983-1997 period (Yan et al., 2001). And the total phosphorus flux to the seas occurs largely via river transport and to a small extent via Station of the Changjiang River during the 1998-1999 period. Moreover, the comparison to the fluxes and concentrations of nutrients in the mouth of the Changjiang River was given, and some knowledge on the fluxes into sea and the distribution of nutrients transport in the Changjiang River was obtained. 2 Methodology 2.1 Sampling and analytical methods 2.1.1 Sampling site The Changjiang River, one of the largest rivers in China (about 6,300 km long), is one of the most developed regions and an important agricultural base of the country (The output of corn accounts for 44% of the country´s total). Warm and wet sub-tropical monsoon climate prevails most of the region, and the concentrated precipitation in summer leads to uneven distribution of water and sediment discharge of the river. Water discharge in high-flow period (May to October) accounts for 70%-80% of the year´s total. In order to calculate transport of nutrient s as accurate as possible in the Changjiang River, the sampling site was chosen at the Datong Hydrologic Monitoring Station in Anhui Province (Figure 1), which is located on the lower reaches of the river, free from tidal effects or direct industrial pollution of towns and cities. The Datong Station, one of the key stations of the national hydrological network, has taken hydrological and hydrochemical measurements of the Changjiang River since the early 1960s. (Figure 1) 2.1.2 Sampling Water and sediment samples were taken by boat from certain cross-sections on three occasions and five times respectively, i.e., high flow (June 15 and August 19, 1998), mean flow (November 16, 1998 and January 21, 1999), and low flow (March 28, 1999). The first sampling was made just before the flood in summer of 1998, and the second one during the flood. In each cruise, the surface, middle and deep samples were collected from center and both sides of the river at the location. Unfiltered water (raw water) samples were preserved in clean polythene bottles for analyzing total N and P. Filtered water (through 0.45 um filtration membranes) was preserved in clean polythene bottles for analyzing the dissolved fraction of N and P. Suspended matter samples were taken from the membranes directly and were immediately transported to laboratories. 2.1.3 Analysis and data collection The analytical nitrogen and phosphorus forms in water samples were analyzed according to the separation scheme:Unfiltered water sample Digestion total nitrogen (TN), total phosphorus (TP); Filtered water sample Digestion dissolved nitrogen (DN), dissolved phosphorus (DP); Filtered water sample No Digestion NO3--N, NO2--N, NH4＋-N, PO43--P; Particulate Nitrogen (PN) =TN-DN; Particulate Phosphorus (PP) =TP-DP; Dissolved Organic Nitrogen (DON) = DN-NO3--N-NO2--N-NH4＋-N; Dissolved Organic Phosphorus (DOP) =DP-PO43--P. The filtrates were measured by using ultraviolet spectrophotometer for nitrate (NO3-), salicylic acid-spectrophotometer method for NH4+, and nitrogen-(1-naphthalene)- ethylenediamine-spectrophotometer method for nitrite (NO2-). The unfiltered and filtered water samples were all digested and then analyzed with the same method as NO3- for TN and DN, determined using ammonium molybdate/potassium antimony tartrate (KSbOC4H4O6 · 1/2 H2O) /ascorbic acid-spectrophotometer method for PO43- or as PO43- for TP and DP. The contents of DON, PN, DOP and PP were calculated by subtracting (see separation scheme) (Committee of Analytical Method of Water and Wastewater, 1989). The Datong Station, supplied the water sample data from 1962 to 1990, others were collected from the hydrologic data of the Changjiang Valley in the Annual Hydrologic Roports of China (1968-1997). 2.2 Calculations of the Changjiang River fluxes Annual fluxes (Fa in g/yr.) of nutrients and runoff volume (W in m3) were calculated using the following equations: Fa = ΣCi(j) × Qi × △ti (1) W = ΣQi × △ti (2) where Ci(j) and Qi are concentrations of the jth form of nutrients (in mg/L) and water discharge (in m3/s) at time of sampling in a certain year, and △ti is the time interval (in s). 3 Results and discussion 3.1 Composition of nutrients and seasonal variation The results of nutrient concentrations are summarized in Table 1 and showed in Figure 2. It can be seen that the mean concentration of total nitrogen (TN) was 1.71 mg/L, being relatively stable, and the standard variation was only 0.11 mg/L. The NO3--N is the major form of dissolved inorganic nitrogen (DIN = NO3- + NH4+ + NO2-) at the Datong Station of the Changjiang River, accounting for about 66% of the total nitrogen (TN). The concentration of NO3--N was as stable as that of TN, and it increased and decreased gently with low and high water discharge, respectively. Generally, many scientists believed that in the flood season (June and August), the nitrogen concentrations of river water were lower because of dilution of higher flow in the dry season (December, January, March), and the nitrogen concentrations were higher because of lower flow. This phenomenon can be explained as the influence of discharge of sewage and point source contamination, because river flow is relatively high and so concentrations after mixing of contaminant and the sewage discharge, on the river flow will be relatively low in flood season. But the flood event in 1998 was the most serious one in China´s history, the "dilution effect" may not be answered correctly for the change of nitrogen concentrations. The concentrations of ammonium nitrogen (NH4+-N) and nitrite nitrogen (NO2--N) were all very low in flood period, but they could increase by about 20 to 30 folds in the dry season and became important components. The successive occurrence of the highest points of NH4+-N (in winter) and NO2--N (in spring) seemed to show a nitrification process, which may take a part in the transformation of NH4+-N to NO2--N. The dissolved organic nitrogen (DON) and particulate nitrogen (PN), the other two important components of nitrogen, accounted for 18% and 7%, respectively. In general, they were high in summer and low in winter. DON concentration increased drastically in flood season, from 0.096 mg/L to 0.686 mg/L, while PN increased comparatively slowly with the rise of water discharge and suspended sediment (SS) concentration. The concentration of total phosphorus (TP) varied relatively greatly, from 0.053 mg/L in dry season to 0.20 mg/L in flood season at the Datong Station of the Changjiang River (Figure 2). Seasonal pattern of particulate phosphorus (PP) was as same as that of TP, but the content of dissolved inorganic phosphorus (phosphate, PO43--P) varied oppositely to TP, with low level in summer and high in winter. No obvious seasonal change for dissolved organic phosphorus was found. Contrary to PN, PP accounted for a high proportion (about 80%), especially in summer (83%), which was closely related to SS concentration in waters (R2 = 0.98). In winter DIP (phosphate) became the predominant form of phosphorus (about

54%). Compared with average levels of the world river water, the concentration of each form DIN in the Changjiang River was almost ten folds those in the background, but the DON and DIP contents were almost at the same level to the background, and the PP concentration was even much lower than that of the background value. It is suggested that the anthropogenic disturbances have greatly altered N and P cycle in the watershed. Some factors, such as the extensive application of chemical fertilizers in the drainage basin and the soil erosion induced by agricultural production and deforestation may contribute to it. The lower PP concentrations might be due to lower SS concentration in the waters (Table 1), the latter was decreased by human activities in the watershed, such as the construction of dams and/or reservoirs (Zhang et al., 1999). Compared with historical data, the concentrations of DIN and DIP were changed largely, such as during the three periods of 1962-1977, 1978-1987 and 1988-1990, the concentrations of NO3--N had almost doubled in every intermediate period, and the concentrations of NH4+-N were also increased substantially, but it had a little variation in the first two periods. 3.2 Temporal and spatial changes in nutrients transport 3.2.1 Forms of nutrients export from the Datong Station of the Changjiang River Based on the annual fluxes (Fa) calculated by equation 1, the forms of nutrients transport from the Changjiang River were obtained as follows: the proportion for annual nitrogen transport in the form of nitrate nitrogen was the highest, about 61.5%; next to the dissolved organic nitrogen (DON), occupying 25.5%; the particulate nitrogen and ammonium contributing 8.7% and 3.8. respectively; and the least for nitrite nitrogen, only occupying 0.5%. The total dissolved nitrogen (TDN) export occupied about 91% of the total nitrogen outputs and predominated the dissolved inorganic nitrogen (DIN), and among them the dissolved organic nitrogen (DON) had also a very high proportion. The characteristics of the latter, might be because the highest flood occurred in the Changjiang River during the 1998-1999 period, which led to a large number of organic matters input from industrial and domestic sources, or because people need to know how the effluent of organic nitrogen in streams is to be transported from land to sea (Van Breemen, 2001). The above-mentioned proportion characteristics for the Changjiang River were close to export from the Nakdong River in Korea (Kim Ki-Hyun, et al., 1998), which belongs to the medium level pollution rivers in the world. We can find that the predominant form of phosphorus export from the Changjiang River was the particulate phosphorus (PP), occupying about 87.2% and that the dissolved inorganic phosphorus (DIP) and organic phosphorus (DOP) accounted for about 6.5% each. 3.2.2 Temporal trends of nutrients transport We have just got to know the forms for the nutrients transport at the Datong Station of the Changjiang River, but what are the time-series characteristics for the nutrients transport and how about transport for them at the hydrological station? The runoff volume and fluxes for three forms of DIN as a function of time were shown in Figure 3. The annual average runoff volume displayed a little variation during the 1962-1990 period, and even had a decrease from 1962 to 1990 (Figure 3a). But a pronounced increase occurred along with time for the major form of the river export of nitrogen--nitrate nitrogen. With consistence changes in its concentration, the annually averaged fluxes for nitrate nitrogen were 157.0×103 t in the first intermediate period of 1962-1977, and the fluxes were 470.9×103 t and 900.2×103 t during the 1978-1987 and 1988-1990 periods, respectively (Table 2). And the fluxes had almost doubled in every intermediate period. The changes of fluxes of ammonium nitrogen and nitrite nitrogen with time were less evident than those of nitrate nitrogen (Figure 3b), but they showed on overall rise in fluxes in the corresponding time range. The decreasing of the annual averaged fluxes was found for ammonium nitrogen during the 1978-1987 period, while a negative relation was noticed between ammonium nitrogen and nitrite nitrogen in the period. At present, three hypotheses have been considered in the interpretation of the characteristics: first, a strong nitrification reaction occurred which is related to the transformation of ammonium and nitrite; second, the nitrogen fixation decreased with changes of land use; and third, industrial waste water and domestic sewage reduced though the agricultural runoff (higher nitrate nitrogen) increased. The monthly average DIN concentrations diminished substantially from 1962 to 1977, but rose sharply after 1977 at the Datong Station (Figure 4a). The above phenomenon was accompanied with monthly average discharge measured seldomly changed during the period of 1962-1990. The temporal trends of the DIN were marked by high concentrations during high-water periods from 1978 to 1990 (Figure 4a). This suggested that DIN concentrations in waters were related to the removal of DIN from the non-point source or the eroded soils. Since fertilizers represent the main source of DIN in this extensive farming area, it is the fertilizers that lead to the sharp increase in DIN concentrations as a result of rapid development of agricultural production in China since 1978. The dramatic change of the annual fluxes and concentrations of DIN in the waters with the elapse of time were demonstrated for the Changjiang River in Figure 4b. The annually averaged DIN concentration exceeded 1.6 mg/L in 1997 but only 0.2 mg/L in 1968, increased by nearly eight folds during the period 1968-1997. Compared to the three decadal stages from 1968 to 1997, no sharp rise of the DIN concentrations in the waters was observed in the first decade, but during the latter two decades the annually averaged DIN concentrations were doubled, especially during the 1988-1997 period for the Changjiang River. The DIN conce ntrations and fluxes were consistent with the increase in waters at the Datong Station of the Changjiang River. The amount of annually averaged DIN in surface runoff was approximately 140×107 kg, about 12% of the total quantities of nitrogen input in 1997; while in 1968, it was less than 20×107 kg, being only 4% (Yan et al., 2001). This means a similar increase between them. As for the dissolved inorganic phosphorus and dissolved silicon (with dissolved SiO2) characterized by distinct changes throughout the year (Figure 5), both concentrations and fluxes of dissolved inorganic phosphorus were 0.004 mg/L and 2.8×103 t during the 1966-1977 period, and they increased to 0.006 mg/L and 5.8×103 t during the 1978-1984 period, the annual average flux had increased two times (Table 2). The annually averaged flux and concentration of dissolved silicon dramatically decreased in consistency with the following years of 1962-1984, especially after 1968. The annual average concentrations were 9.45, 7.39 and 6.25 mg/L in the years 1963, 1973 and 1983, respectively. The concentration dropped by 3.2 mg/L during the 20 years (Li and Cheng, 2001). And the annual average fluxes (with dissolved Si) were 3,246.6×103 t and 2,700.8×103 t during the 1966-1977 and the 1978-1994 periods, respectively (Table 2). Many factors, including natural ones and human activities can attribute to the time-series increase in nutrients loads of the Changjiang River. Even if runoff volume and discharge may play minor roles, no uprisings were showed in them before 1990. However, impacts of human activities on nutrient transport in the river such as application of chemical fertilizers and disposal of wastewater cannot be ignored (Zhang et al., 1995; Chen et al., 1997; Duan et al., 1999). Meanwhile, for the reason that the dissolved silicon in river waters was almost provided from chemical weathering of rocks, the time-series decrease in dissolved silicon in the Changjiang River can be wholly ascribed to the intensive influence of human activities, such as the dam and/or reservoirs constructions. 3.2.3 Spatial trends of nutrients transport We compared the average concentration of nutrients transport between that from the Datong Station into the estuary and that from the Changjiang Estuary into the sea (Table 3). The concentration of DIN was higher than that of the available documented data, especially that had changed greatly in the concentration of nitrate nitrogen, but without pronounced changes in the concentrations of the DIP (phosphate) and dissolving silicon. The following factors might be an influential of nutrients transport in spatial trend: firstly was the time factor. The concentration of nitrate nitrogen was notably changed with time as the preceding said, so our measured results in 1998-1999 were higher than those in the 1980s. At the same time, a characteristic increase in concentration with time had been noticed for data of the Changjiang Estuary (Table 3), and obviously it was unanimous with the time-series features for the Datong Station. Moreover, our data of nitrate nitrogen in 1998-1999 were close to the value of nitrate nitrogen measured in the Changjiang Estuary in September 1999, it is an important evidence for the concentration affected by time. Secondly was the distance factor. Some processes, such as, nitrification, denitrification, mixing etc., would occur in the course of nutrients transport within a distance of about 706 km from the Datong Station to the Changjiang Estuary. Thirdly was the estuary process factor. The estuary area had the possibility to be invaded by the sea water and result in a descend in the concentrations of most nutrients, even in those of few nutrients go up (such as phosphorus) (Shen et al., 2001), but the Datong Station was not affected by the sea water. Therefore, we thought that the estuary process was a dominant factor in changing the average concentration of output nutrients, and the distance might take a minor part in those along the range. And, we found that some nutrients were assumed the conservative characteristics in the course from the Datong Station to the Changjiang Estuary at the same time. The annually averaged flux of nutrients between the Datong Station and the Changjiang Estuary was contrasted (Table 2), about 1,847.4×103 t of N, 149.28×103 t of P and 2,427.5×103 t of Si were transported to its estuary from the Datong Station in the one year cycle. The fluxes of DIN at the Datong Station were higher than those in the Changjiang Estuary from various years, and the fluxes of DIP in the various locations varied greatly, but the fluxes of dissolved silicon in those locations did not change very much. The variations of the annually averaged fluxes were similar to that of average concentration, which was derived from the influence of time, distance and estuary process. We found the fluxes of nitrate nitrogen and DIN approached to be identical between the Datong Station and the Changjiang Estuary in the same time intervals. And some authors noticed that transformation ammonium nitrogen between dry and wet seasons can be more than tenfolds (Shen, 1997), or it might partly influence the fluxes of DIN. For the fluxes of DIP in the Changjiang Estuary were usually larger than those at the Datong Station at the same time, the estuary process might be responsible for the rise for those of DIP. 4 Conclusions (1) The nitrate nitrogen and particulate phosphorus were dominant components of nutrients in waters of the Changjiang River at the Datong Station. Among them, the mean concentrations of total nitrogen and nitrate nitrogen were 1.71 mg/L and 1.17 mg/L, respectively, being stable against the seasonal change. Those of ammonium nitrogen and nitrite nitrogen decreased in the flood season, grew higher in dry season and probably occurred to nitrification reaction; the DON and PN account for 18% and 7%, being generally high in summer and low in winter. It was pronounced in PP, DIP changed with season, and the former was high in summer and low i

n winter, and vice versa for the latter. The DOP had a little variation with season. (2) The concentrations of the DIN in waters of the Changjiang River in 1998-1999 were ten-folds higher than those of the average level of the world river waters, and the DOP and DIP contents were about the same level to the background, but the PP concentration was lower than the background. The concentrations of nitrate nitrogen were almost doubled in every intermediate period of 1962-1977, 1978-1987 and 1988-1990 from historical data of water quality. The forms of N and P export from the Changjiang River were predominance with DN, especially in NO3--N and DON, and PP. The DON occupied about 30% of the former, which need to be further studied. (3) The fluxes of three forms of DIN rose during the period 1962-1990, accompanying with a little change for the runoff volume, and particularly those of the nitrate nitrogen were the most obvious changes. At the same time, the concentrations and fluxes of DIN and DIP increased along with time, but appeared a negative trend for the dissolved silicon. The human activates could be the most important influencing factors. (4) The difference of the average concentration and annually averaged flux were subtle between the Datong Station and the Changjiang Estuary. The reasons for this phenomenon are the time, distance and estuary process. The estuary process was the predominant factors.

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关键词： Changjiang Estuary; nutrient; seasonal variation; forms; temporal and spatial trends