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ScienceDirect

Journal of Environmental Sciences 2011, 23(10) 1684–1690

JOURNAL OF
ENVIRONMENTAL
SCIENCES
ISSN 1001-0742
CN 11-2629/X
www.jesc.ac.cn

Spatial and temporal distribution of acetochlor in sediments and riparian soils of the Songhua River Basin in northeastern China

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Received 25 November 2010; revised 13 January 2011; accepted 22 March 2011

Abstract

The Songhua River Basin is a burgeoning agricultural area in the modern times in China. Particularly in recent years, increasing chemical fertilizers and pesticides have been applied with the development of agricultural production. However, the situation of non-point source pollution (NSP) from agricultural production in this basin is still obscure. In order to solve the problem, the occurrence and distribution of acetochlor in sediments and riparian soils of the Songhua River Basin before rain season and after rain season were investigated. In addition, total organic carbon was analyzed. The result showed that the concentration of acetochlor ranged from 0.47 to 11.76 μg/kg in sediments and 0.03 to 709.37 μg/kg in riparian soils. During the high flow period in 2009, the mean concentration was 4.79 μg/kg in sediments and 0.75 μg/kg in riparian soils, respectively. Similarly, the mean concentration was 2.53 μg/kg in sediments and 61.36 μg/kg in riparian soils, during the average flow period in 2010. There was a significant correlation between the concentration of acetochlor and total organic carbon in surface sediments. Moreover, the distribution of acetochlor in sediments of the Songhua River was significantly correlated to land use and topography of the watershed. The investigated data suggested that the concentration of acetochlor in the Songnen Plain and the Sanjiang Plain was higher than that in the other areas of the basin, and riparian buffering zones in these areas had been destroyed by human activities. The optimal agricultural measures to alleviate the contamination of pesticides should be adopted, including controlling agricultural application of acetochlor and ecological restoration of riparian buffering strips.

Key words: acetochlor; Songhua River Basin; non-point source pollution; optimal agricultural measure

DOI: 10.1016/S1001-0742(10)60595-5

Citation: Sun X Y, Zhou Q X, Ren W J, Li X H, Ren L P, 2011. Spatial and temporal distribution of acetochlor in sediments and riparian soils of the Songhua River Basin in northeastern China. Journal of Environmental Sciences, 23(10): 1684–1690

Introduction

The widespread use of chemical fertilizers and pesticides leads to water pollution throughout the world, and many studies have shown their presence in rivers, streams, and groundwater (Kolpin et al., 1996; Konda and Pásztor, 2001; Thurman et al., 1996). Herbicide pollution in river water is of increasing public concern because herbicides can result in immediate and long-term risks for ecosystems and for human health (Blann et al., 2009; DeLorenzo et al., 2001; García, 2003). The occurrence of herbicides in waterbodies originates mainly from non-point sources such as surface runoff and artificial drainage from agricultural fields or spray drift. The monitoring and control of herbicides in rivers and streams are an important practical problem to be solved in many areas and countries, and the implementation of effective measures that prevent and/or reduce the losses of herbicides from non-point sources is a challenge for agricultural and water management.

The Songhua River is the third biggest river in China. Located at the joint section of the temperate zone and the cold-temperate zone and affected by the terrestrial seasonal wind climate, the region has a long cold winter, a rainy torrid summer and a windy dry spring. The watershed traverses three provinces including Heilongjiang Province, Jilin Province and Inner Mongolia Municipality. The Songhua River is the major source of freshwater for drinking and daily life, and water source used for industrial and agricultural production in the area. Moreover, fertile and productive soils were distributed in the Songhua River Basin as one of the most important commodity grain bases in China. For example, the annual output of corn in this area was up to 4346 tons in 2006, about 29.7% of output in the whole country (Agriculture Ministry of People's Republic of China, 2007). With the development

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of agriculture, various pesticides, especially herbicides had been increasingly applied in this area and the applied amount in this area greatly exceeded other areas of China (Su, 2004). In the Songhua River Basin, the annual proportion of herbicides was about 76.4%–83.4% of the total pesticides applied, in particular, the proportion of the herbicides applied to dry farmland was above 70%. Acetochlor or mixture of acetochlor and other herbicides was the main mode of herbicide application (Ma, 2009).

With a wide use of acetochlor to agricultural lands, it had undoubtedly resulted in pollution in adjacent surface waters and thus posed a potential risk to a range of aquatic organisms. For example, it had been shown that acetochlor could induce metamorphosis of ranid species and accelerated T-3-induced metamorphosis in amphibians (Crump et al., 2002; Li et al., 2009). So far, the U.S. Environmental Protection Agency (EPA) has classified acetochlor as a B-2 carcinogen and acetochlor registration can be canceled if its concentration exceeds 0.10 µg/L in groundwater or 2.0 µg/L as an annual average in surface water. Thus, investigations on the environmental distribution of acetochlor are important for both ecological risk assessment and agro-environmental protection.

Despite of its high ecological risk and wide application, there are very little data available for the distribution of acetochlor in surface water and bottom sediments in rivers and streams. Previous studies based on sorption and degradation experiments have shown that acetochlor presents a risk of soil contamination, especially in phaeozem (Chao et al., 2007; Xiao et al., 2005; Zhou et al., 2006), whereas there is no report about its presence in surface water and sediments in the Songhua River Basin. Thus, the primary objectives of this study include: (1) to investigate the distribution and spatial trends of acetochlor and nutrients in sediments and riparian soils of the Songhua River Basin; (2) to analyze main correlative factors affecting acetochlor pollution in sediments and riparian soils; and (3) to put forward the best management practices (BMPs) for alleviation of the herbicide pollution in the Songhua River Basin.

1 Materials and methods

1.1 Studying area

The Songhua River locates between 119°52′–132°31′E (Fig. 1), and flows about 1927 km from the Changbai Mountain through Jilin Province and Heilongjiang Province. The river drains 212,000 m^2 of land. It joins Amur at the town of Tongjiang. The Songhua River Basin is one of the most important agricultural area in China. In recent years, increasing chemical fertilizers and pesticides are being applied with the development of agricultural production in this country.

1.2 Sampling sites

The sampling sites of sediments and riparian soils were chosen on the basis of the national and provincial monitoring network for the Songhua River Basin after having considered the geomorphology of the main channels, the hydrological regime, the localization of the urban and industrial discharges, and the land use of riversides. All sampling and pretreatment of samples accorded with the Chinese National Standards (Environmental Protection Administration of China, 2002). In each transversal section of the river, the top sediment samples (a mixture of sediments from the upper 20 cm) were collected by a box-like grab sampler manufactured by the Harbin Station of Environmental Monitoring, Heilongjiang Province, China.

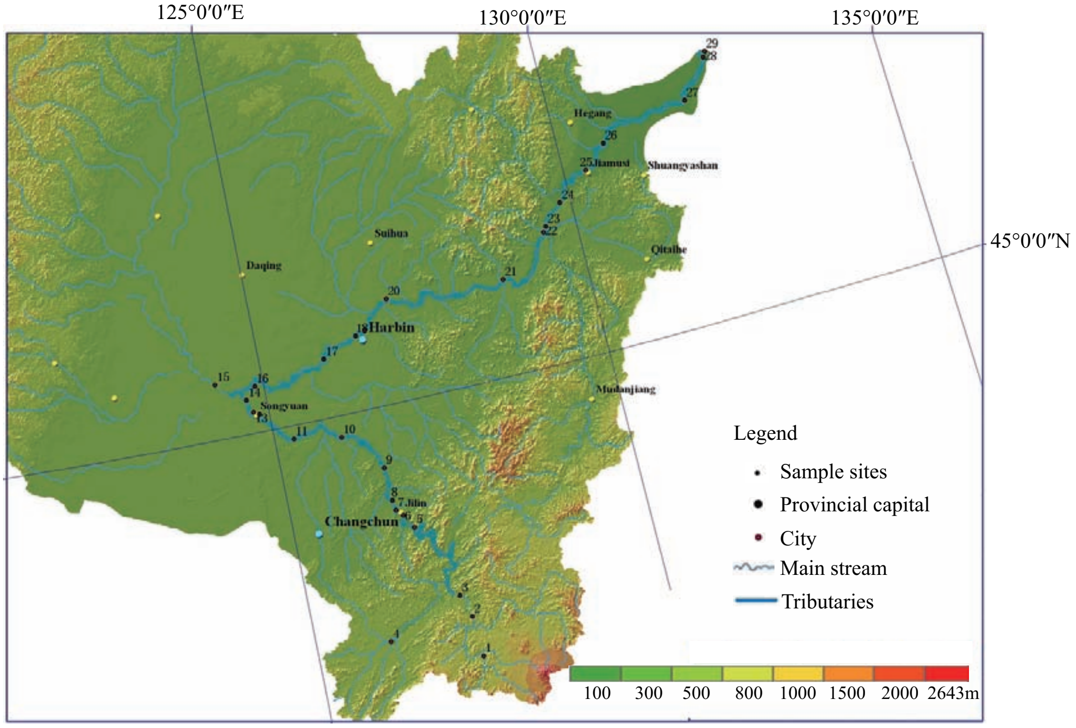

Fig. 1 Sampling sites in the Songhua River Basin.

The soil samples were collected from surface riparian soils (0–20 cm) at the corresponding sediment-sampling sites. Each site was orientated using a global positioning system (GPS). Sites were tabbed from sites 1 (upstream) to 29 (downstream) (Fig. 1). After the collected samples were homogenized on site in clean glass containers, they were immediately sealed and stored at –20°C in pre-cleaned glass jars until analysis. Samples were collected after rain season during the high flow period in 2009 and before rain season during the average flow period in 2010. The description of sampling sites are given in Table 1.

1.3 Chemicals

Analytical reagents including acetone and *n*-hexane were bought from the Yuwang Chemical Plant in China and used for sample processing and extraction. Anhydrous sodium sulfate was supplied by the Bodi Chemical Plant in China and treated at 600°C for 3 hr before use. Florisil siliceous earth (100–200 mesh) and neutral alumina (100–200 mesh) for column chromatography were obtained by the Sinopharm Chemical Reagent Co., Ltd. (China) and treated at 550°C for 3 hr before use. Acetochlor standard was purchased from the J&K Chemical Ltd. (China) with purity greater than 95%.

1.4 Chemical analyses and determination

Sediment and soil samples were freeze-dried by a vacuum freeze dryer (FD-1; Boyikang Experimental Instrument Ltd., Beijing, China). After having removed pieces of stones and plants, the samples were ground and sieved through 0.15 mm mesh. Then they were placed in brown glass jars and stored at –20°C before use. The samples were solvent extracted using ultrasonic oscillator following the Standards Method of Liaoning Province, China (Wang et al., 2010). Twenty grams of the samples were weighted into a glass vial and sonicated for 30 min with 40 mL of acetone/*n*-hexane (4/1, V/V) in an ultrasonic bath (KQ250B, Kunshan Ultrasonic Bath Plant, China). In this case, sediment samples were sonically extracted thrice with acetone/*n*-hexane and centrifuged. After extracts were transferred into glass flasks by careful decantation, they were evaporated under vacuum with a rotavapor (EYEL4 rotary vacuum evaporator N-N series, Electric Co., Ltd. Zibo Sanyuan, Japan) and a water bath (about 35°C). The organic phase was evaporated to near dryness. The residues were cleaned up by a column (8 mm i.d. × 150 mm) containing 0.5 g Florosil and 0.5 g neutral alumina. Anhydrous sodium sulfate (1 cm) was added to the top and

Table 1 Distribution of acetochlor in sediments and riparian soils (unit: μg/kg)

Sampling number	Sampling site	Surface sediment		Riparian soil		Land use around sites	Type of riparian buffering strips
		High flow period	Average flow period	High flow period	Average flow period		
s1	Xijiang Park in Husong	ND	0.39	ND	11.38	Woodland	Artificial riverbank
s2	Baiyunshan Hydro-electric Dam	ND	ND	–	3.61	Dry land in mountainous region	Lawn
s3	Redstone Bridge	ND	ND	ND	7.88	Village	Artificial riverbank
s4	Huifa River Bridge	ND	0.44	–	–	Dry land in plain	No buffering strip
s5	Fengman Hydro-electric Dam	ND	1.51	ND	6.98	Village	Lawn
s6	Longtankou	ND	0.84	ND	0.94	Dry land in plain	No buffering strip
s7	Jiuzhan Ferry	ND	ND	–	–	Paddy field in plain	Bare land
s8	Shaokou Ferry	ND	ND	ND	16.21	Dry land in plain	Bare land
s9	Huojia Ferry	ND	ND	ND	111.32	Paddy field in plain	Bare land
s10	Songhuajiang Village	ND	ND	10.15	119.39	Woodland	Lawn
s11	Zhenjiangkou	ND	ND	ND	17.54	Paddy field in plain	Lawn
s12	Xumuchang	2.13	0.36	ND	1.5	Dry land in plain	Lawn
s13	Xidazui	0.96	2.23	–	–	Dry land in plain	No buffer strip
s14	Shiqiao	3.52	0.95	ND	47.09	Dry land in plain	No buffer strip
s15	Daanlaokanzi	0.86	ND	–	–	Dry land in plain	Artificial riverbank
s16	Liuyuanzi	–	ND	0.47	1.85	Grassland	Lawn
s17	Hatugangzi	–	ND	0.59	0.92	Grassland	Lawn
s18	Sifangtai	ND	ND	–	–	Dry land in hilly land	No buffer strip
s19	Harbin	ND	ND	ND	162.55	Town	Artificial riverbank
s20	Dadingzi	ND	–	ND	168.02	Woodland	Lawn
s21	Tonghe Ferry	2.20	0.58	0.58	1.82	Dry land in plain	No buffer strip
s22	Mudanjiangkou	ND	0.03	2.22	2.46	Woodland	Bare land
s23	Wokenhekou	4.51	0.03	ND	12.17	Dry land in plain	Lawn
s24	Hongkeli	ND	0.63	4.50	709.37	Dry land in plain	No buffer strip
s25	Jiamusi Water Quality Monitoring Station	11.76	–	3.58	0.87	Dry land in plain	Lawn
s26	Huachuan	9.03	ND	2.02	3.63	Dry land in plain	Lawn
s27	Fujing Port	7.63	ND	1.91	2.18	Dry land in plain	No buffer strip
s28	Water Quality Automatic Monitoring Station in Tongjiang	2.74	1.01	–	–	Dry land in plain	Lawn
s29	River estuary of Songhua River, Ussuri River and Amur River	ND	–	6.89	1.57	Dry land in plain	Artificial riverbank

ND: not detected. "–": no sample.

bottom of the column to remove water. The column was then eluted firstly with 8 mL of acetone/*n*-hexane (2/98, *V*/*V*) and the solution was discarded. Further 25 mL of acetone/*n*-hexane (4/96, *V*/*V*) was needed to elute to obtain acetochlor. After collection, the eluate was evaporated near dryness under a nitrogen flow. Subsequently, the sample was dissolved in 1 mL of *n*-hexane for GC determination.

Determination was done by a GC system with a micro cell electron capture detector (μ-ECD) and a split/splitless injector (Agilent Series 6890 plus, Agilent Technologies, USA), equipped with ChemStation Software. A fused silica capillary column (HP-5, 30 m × 0.25 mm i.d. and 0.25 μm film thickness) was used. The oven temperature was programmed from 60 to 150°C at a rate of 20°C/min, then from 150 to 230°C at a rate of 8°C/min and held for 5.5 min. Nitrogen (99.999%) was used as carrier gas and make up gas. A split/splitless injector was used in the splitless mode. The injector temperature and the electron capture detector temperature were set at 220 and 300°C, respectively. The injection volume was of 1 μL. The recovery was 70.2%–98.3% and RSD was 3.0%–7.1%. Limit detection was 0.01 μg/kg. The recovery was satisfactory for acetochlor and it was corrected for quantification. Total organic carbon (TOC) and total nitrogen of soils and sediments were detected by Vario EL III elemental analyzer (Elementar, Elementar Analysensysteme GmbH, Germany) besides acetochlor. Total phosphorus was detected by color comparison by spectrophotometer (Varnian Cary 50 Conc, Varnian, USA) after heating digestion by perchloric acid and sulphuric acid mixed solution (Lu, 2000).

1.5 Statistical analysis and data processing

All statistical analyses were performed with the SPSS (version 17.0; USA). The processing of the data was done using analysis of variance, followed by Bonferroni's Multiple Comparison Test. A probability of $p < 0.05$ was considered statistically significant.

2 Results and discussion

2.1 Occurrence and distribution of acetochlor during the high flow period and the average flow period

The distribution of acetochlor in sediments and riparian soils from upstream to downstream is listed in Table 1. The data revealed that there was an approximate detection rate of acetochlor in sediments between the high flow period in 2009 and the average flow period in 2010. Concretely speaking, the detection ratio of acetochlor was 34.6% in 26 samples collected in 2009 and 34.9% in 23 samples collected in 2010. However, there was a great change in the detection rate of acetochlor in riparian soils. Namely, 52.4% of 21 soil samples collected during the high flow period in 2009 contained acetochlor and 100% of 23 soil samples collected during the average flow period in 2010 could detect acetochlor. The concentration of acetochlor ranged from 0.03 to 11.76 μg/kg in sediments and 0.47 to 709.37 μg/kg in riparian soils (Table 1). During the high flow period in 2009, the mean concentration of acetochlor was 4.79 μg/kg in sediments and 0.75 μg/kg in riparian soils; similarly, the mean concentration during the average flow period in 2010 was 2.53 μg/kg in sediments and 61.36 μg/kg in riparian soils. Obviously, the detection rate and the concentration of acetochlor in riparian soils during the average flow period were higher than those during the high flow period. It may be mainly due to the serious soil erosion during the high flow period.

2.2 Spatial trends of acetochlor in sediments and riparian soils

The spatial changing trends of acetochlor in sediments and riparian soils of the basin are depicted in Fig. 2. Because the sites from s1 to s9 distributed in the mountainous upstream area and its river velocity was high, acetochlor in sediments from the sites could not be detected in the high flow period. However, all the riparian soil samples from this upstream area contained acetochlor during the average flow period, because these sites border upon agricultural land. Noticeably, the concentration of acetochlor in the sites from s10 to s19 belonging to the Songnen Plain increased gradually because its river velocity became slow, abundant fluvial sediments gravitated and acetochlor entered into water bodies by soil erosion; on the contrary, the concentration of acetochlor in the sites from s20 to s22 belonging to middle hilly reaches of the Songhua River decreased to a certain extent during the high flow period because its river velocity increased. The concentration of acetochlor in the sites from s23 to s29 belonging to the Sanjiang Plain was at the highest the high flow period and the average flow period, because it is an alluvial plain, and its river velocity declined. In particular, the concentration of acetochlor in sediments went up evidently along the river. Compared with that in the upstream area (I), the concentration of acetochlor in sediments and riparian soils from the Sanjiang Plain (IV) in the downstream area of this river was higher (Table 2).

Compared with that in other watersheds, the concentration of acetochlor in sediments from the Songhua River Basin was not high even though it was serious in the

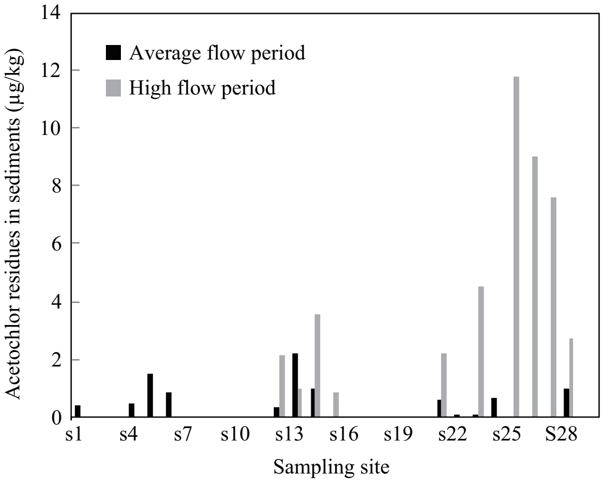

Fig. 2 Distribution of acetochlor in sediments of the Songhua River Basin.

Table 2 Concentration of acetochlor in sediments and riparian soils of four reaches

Reach	Sediment (μg/kg)		Riparian soil (μg/kg)		Topography
	High flow period in 2009	Average flow period in 2010	High flow period in 2009	Average flow period in 2010	
I	0.00	0.80	0.80	22.62	Changbai Mountainous area
II	1.87	0.53	1.18	50.12	Songnen Plain
III	2.20	1.40	0.31	57.43	Hilly area
IV	7.14	3.78	0.56	121.63	Sanjiang Plain

I: Reaches in upstream from sites s1 to s9; II: reaches in in the Songnen Plain from site s10 to s17; III: reaches from sites s18 to s21; IV: reach in the Sanjiang Plain from sites s22 to s26).

Table 3 Comparison of acetochlor concentrations between the Songhua River Basin and other watersheds

Area	Sample	Acetochlor concentration (μg/kg)	Reference
Beijing Miyun Watershed, China	Corn field	37 (24–64)	Zheng and Ye, 2001
Huaihe River, China	Sediment	4900 (3900–6600)	Wang et al., 2002
Beijing Guanting Reservoir, China	Sediment	0.0374 (0.05–0.188)	Xue et al., 2005
Belgrade, Serbia	Agricultural soil	13.3	Markovic et al., 2010
Northern Liaoning Province in China	Agricultural soil	41.1 (12.3–71.0)	Wang et al., 2010
Songhua River Basin	Sediment/riparian soils	2.47 (0.47–11.76)/43.76 (0.03–709.37)	This study

Data are expressed as average (range of acetochlor concentration).

corresponding riparian soils. In particular, the concentration of acetochlor in riparian soils before rain season was higher than concentration of acetochlor in soils of Beijing Miyun Watershed and Belgrade (Table 3). Similarly, the concentration of acetochlor in riparian soils was also higher than that (41.1 μg/kg) in agricultural soils from Liaoning Province in China (Wang et al., 2010).

2.3 Factors affecting distribution of acetochlor in sediments

There are many factors affecting the transference of herbicides in rivers and streams, for examples, soil and hydrologic properties and geomorphologic characteristics of the watershed, land use and herbicide management, climate factors particularly precipitation and temperature (Leu et al., 2004; Xu et al., 2009; Zhang et al., 2009).

The result in this study indicated that hydrologic properties, geomorphologic characteristics, riparian buffering zones and sediment properties of the watershed were the main factors. There was no detection of acetochlor in sediments from reaches I (s1–s9) and III (s18–s21) (Fig. 2, Table 2) during the high flow period, but it was obviously detected in riparian soils in these areas, which suggested that contamination of acetochlor did not take place in sediment because of rapid river flow among the mountainous area and a few sediments.

Another important factor was land use in the watershed (Tran et al., 2010). Reaches II (s10–s17) and IV (s22–s26) lie in the Songnen Plain and the Sanjiang Plain with intensive agricultural activities and herbicides were applied abundantly into agricultural fields. The result had suggested that the average concentration of acetochlor in riparian soils in these areas was obviously higher than that in other areas.

The Pearson correlation coefficient between the concentration of acetochlor and total organic carbon in surface sediments collected during the high flow period was 0.646 ($p < 0.05$), but the correlation during the average flow period was not significant. The total soil/sediment organic carbon content and its qualitative characteristics are the most important factors affecting sorption-desorption of herbicides in soil or sediments. The previous study indicated that the sorption capacity of acetochlor in soils and sediments increased with an increase in the content of soil organic carbon (Hiller et al., 2009). The relation during the average flow period was not significant maybe the transference of acetochlor from soils to sediments was impacted differently between sampling sites.

2.4 Riparian buffering strips and their influences

Many studies had suggested that land use had impacted on water quality and commonly occurred within a shorter distance of the receiving water body (Allan, 2004; Barling and Moore, 1994; Harding et al., 1998), particularly in streams, it affected surface water quality directly. Many buffering strips in the Songhua River Basin had been destroyed and became bare land, the investigation indicated that 27.6% of sampling sites had no riparian buffering zones and agricultural land was adjacent to river directly (Table 1).

Riparian buffering strips are effective in plugging up pollutants into the river, especially in reducing agricultural non-point sources by filtration, deposition, adsorption, and infiltration (Borin et al., 2010; Caron et al., 2010; Krutz et al., 2005; Lacas et al., 2005; Zhang et al., 2009). For example, Patty et al. (1997) found that riparian buffering strips could reduce 44%–100% of atrazine from agricultural runoff (Patty et al., 1997). Riparian buffering strips are one of the best management practices proposed for reducing transport of pesticides to surface water.

3 Conclusions

The Songhua River is an important freshwater body in northeastern China, and the loss of pesticides and excessive application of chemical fertilizers are the main pollution sources. This study suggested that the concentration of acetochlor in sediments was lower than that in agricultural soils, and there was a great difference in the concentration of acetochlor in riparian soils between the high flow period and the average flow period because of soil erosion. The topography, land use and sediment properties were the main determinants of acetochlor contamination in sediments. Riparian buffering zones in the Songhua River Basin had been destroyed by human activities. In this sense, the optimal agricultural measures to alleviate the contamination of pesticides should be adopted by controlling the applied amount of acetochlor and ecological restoration of riparian buffering strips.

Acknowledgments

This work was supported by the National Special Water Project in China (No. 2008ZX07526-002-01). We thank our river sampling participants from Agriculture University of Northeast China, Nankai University, and the Harbin Station of Environmental Monitoring in Heilongjiang Province, including Prof. Ying Zhang and her assistant Dr. Qingjuan Meng, and Dr. Zhigang Wang from Agriculture University of Northeast China; Prof. Hongbin Yu, Dr. Dr. Xingdong He, Dr. Tao Hua, Dr. Xin Yu, Dr. Wenlu Zeng, Dr. Zhiqiang Wang and their graduate students from Nankai University, and Director Mingyu Wang and his colleagues of monitoring station.

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