Ecological Engineering 120 (2018) 260–265

Contents lists available at ScienceDirect

Ecological Engineering

journal homepage: www.elsevier.com/locate/ecoleng

Herbicide accumulations in the Xingkai lake area and the use of restored wetland for agricultural drainage treatment

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ARTICLE INFO

Keywords:
Cross-border lake
Acetochlor
Butachlor
Wetland treatment
Removal efficiency

ABSTRACT

The plains surrounding the northern part of the Xingkai Lake (XL) straddling the border between China and Russia, is a major agricultural production area in Northeastern China. The accumulations from herbicides applied in the farmlands are an important factor affecting the local wetland environment. In this paper, top soil and surface water samples collected from different land use types were analysed. The sediments of a restored wetland receiving agricultural drainage (RW) were tested for two representative herbicides (butachlor and acetochlor). The removal of the two herbicides were calculated based on the herbicide differences between the sediments of RWO and RWI. Results showed that herbicides were widespread in XL area. The highest acetochlor concentration was detected in soybean dry land soil (117.1 ng g^{-1}) and RWI water (402.2 ng L^{-1}) compared with the average concentration of 121.63 ng g^{-1} in the riparian soils of the Sanjiang Plain, and the highest butachlor concentration was in the paddy field soil (140.55 ng g^{-1}), which mainly originated from human activities related to agricultural production. Due to the selective removal of RW and the differences in the characteristics of the herbicides, removal by RW was higher for butachlor than acetochlor, with removal efficiencies of 29.9% and 0.5%, respectively, indicating that butachlor was more degradable in wetlands. We conclude that herbicide accumulations do exist in the Lake Xingkai area, while RW used for agricultural drainage treatment can remove limited parts of herbicides. More rigid pesticide controls are recommended for the protection and management of the wetland environment in XL area.

1. Introduction

As a class of chemicals used to kill weeds to protect and promote the growth of plants and crops during in agricultural production, herbicides have accounted for the largest portion of production and use among total pesticides (Stokstad and Grullón, 2013). Currently, the use of persistent herbicides (e.g. atrazine, metsulfuron-methyl) have gradually been replaced by the use of low-toxicity and easily degradable herbicides (e.g. acetochlor, butachlor).

Acetochlor [2-Chloro-*N*-(ethoxymethyl)-*N*-(2-ethyl-6-methylphenyl)acetamide] is a pre-emergent herbicide widely used to control broad-leaved competing weeds in farmlands of soybean and corn etc. Since it was developed by Monsanto Company and Zeneca in 1971 and given conditional registration in the United States in 1994, acetochlor has been widely used all over the world (Kolpin et al., 1996; Zheng and Ye, 2002; Lengyel and Földényi, 2003). Butachlor [*N*-(butoxymethyl)-2-chloro-*N*-(2,6-diethylphenyl)acetamide] is another widely used herbicide to control a range of annual weeds emerging in rice etc. farmlands as well as submerged macrophytes in freshwater fishponds (Abigail et al., 2015). Acetochlor and butachlor were the two of the most widely used herbicides in China around the 2000s (Zheng and Ye, 2002). Their widely use have aroused increasing scientific concerns for their environmental behaviors and ecological consequences (e.g. Ye, 2003; Yu et al., 2003; Kulikova et al., 2005; Oliveira et al., 2013; Yang et al., 2013; Abigail et al., 2015; Bedmar et al., 2017), especially when their gene damage and joint toxicity were confirmed (Crump et al., 2002; Dwivedi et al., 2012; He et al., 2013).

It is generally believed that the ecosystem and human health risks of the two herbicides are determined by their toxicity and persistence in environment that is reflected by their degradation kinetics, which depends on their molecular structures and is affected by key environmental factors such as usage, physical, chemical and biological conditions. Due to the low adsorption coefficient (K_{oc}) and relative mobility coefficient (R_f) of acetochlor while high K_{oc} and R_f of butachlor, the

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https://doi.org/10.1016/j.ecoleng.2018.06.009
Received 23 March 2018; Received in revised form 5 June 2018; Accepted 7 June 2018
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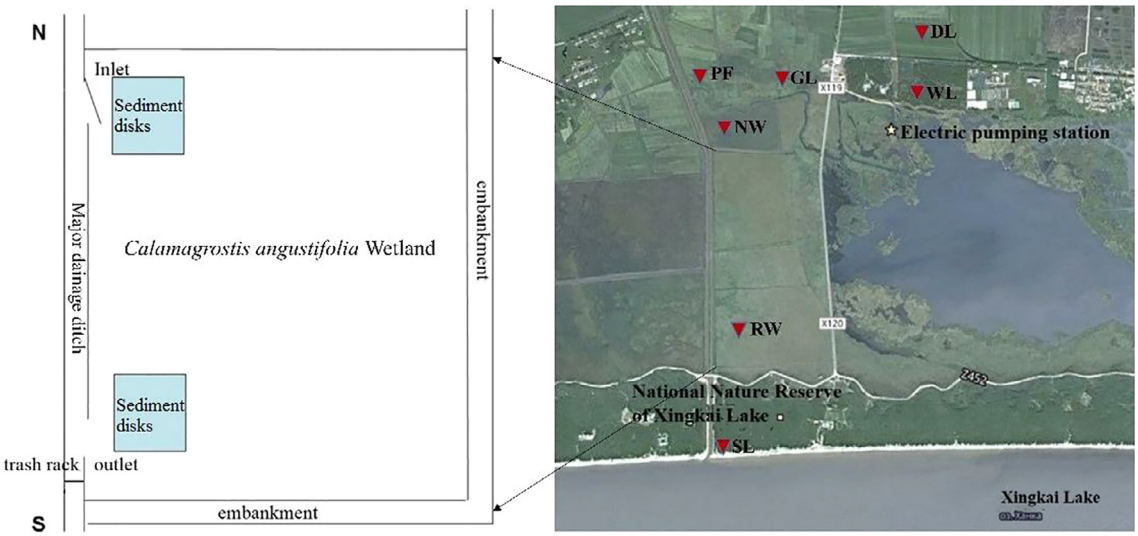

Fig. 1. Sketch of the sampling sites. RW: the restored wetland used for agricultural drainage treatment; NW: the natural wetland; SL: the sandy land; GL: the grassland; WL: the wood land; DL: the dry land; PF: the paddy field.

former herbicide is a rather mobile pollutant in dry land soils, while butachlor is a persistent pollutant in paddy field soils (Swann et al. 1983; Zheng and Ye, 2002; Lengyel and Földényi, 2003). Consequently, both acetochlor and butachlor have potential threat to the aquatic ecosystem when soil flooding or runoff events occur.

As the transitional ecotone between terrestrial and aquatic ecosystems, wetlands can remove herbicides and nutrients through physical, chemical, and biological processes (Reddy and Delaune, 2008; Maillard et al., 2011), thereby significantly reducing herbicide pollution to rivers and lakes and reducing toxicity; however, herbicide removal efficiency in wetlands is species-dependent and affected by many processes and factors (Vymazal and Brezinova, 2015). It has been confirmed that acetochlor can be effectively degraded in constructed wetlands under nitrate and sulphate-reducing conditions, especially in the rhizosphere zone where plants can enhance the removal efficiency (Chen et al., 2017; Elsayed et al., 2015), and butachlor can be degraded and used by microorganisms for growth in constructed wetlands (Debnath et al., 2002). However, there are litter reports of the two herbicides in natural wetlands in field scale and their accumulations and fates are still unclear.

The Xingkai Lake (XL) is the largest and transboundary lake in Heilongjiang River Basin of China. The concentration of acetochlor in the Sanjiang Plain was reported as one of the two highest accumulation parts of the Heilongjiang River Basin, with the average acetochlor concentration of 121.63 ng g^{-1} in riparian soils in 2010 (Sun et al., 2011), while the data did not include the survey of XL area. In recent years, agricultural practices and land use conversions in the riparian zone of XL have led to degradation of wetlands (Huo et al., 2018). At present, the status of herbicides in XL area is not clear, and very few studies existed concerning the impact of herbicides on the wetland ecosystems. In this paper, the concentrations of two representative herbicides (acetochlor and butachlor) widely used in agricultural production in XL area were surveyed, and the removal efficiencies of a restored wetland for agricultural drainage treatment were investigated as well. The objectives are (1) to clarify the distribution and accumulation characteristics of acetochlor and butachlor in soils of different land use types and water, and (2) to analyse the removal effectiveness of the two herbicides in the wetland ecosystem.

2. Materials and methods

2.1. Study area

Located in Jixi City, Heilongjiang Province, China, XL has an elevation of 69 m and a total area of 4380 km^2 (131°58′30″E–133°07′30″E, 45°01′00″N–45°34′30″N); it is composed of the Daxingkai Lake (DXL) and the Xiaoxingkai Lake (XXL) and is the border lake between China and Russia. The west, south, and northeast sides of XL have hilly terrain, while the north side is developed into XL Plain. There are a large number of various wetlands distributed in the lake area. The soil in the lake area is dominated by marsh meadow soil and the vegetation is composed of wetland plants and crops. In the lake area, besides the wide open water, the woodlands and meadows are chimerically distributed, with their original states being intact; the lake area is extremely rich in biodiversity and was included in the Ramsar Site List of internationally important wetlands in 2002.

XL has four distinct seasons: its winter is long, dry, and extremely cold and has little snow; its summer is warm and humid and has abundant rainfall; its spring and autumn are windy and have little precipitation. The annual average precipitation is 654 mm, with the most monthly precipitation in August (average 119.7 mm) and the least in January (average 5.4 mm). The annual average temperature is 3.1 °C, with the highest monthly temperature in July (average 21.2 °C) and the lowest in January (average −19.2 °C) (Huo et al., 2018). The southwest wind prevails in the spring and summer, and the northwest wind dominates in the autumn and winter; the annual average wind speed is 4.0 m s^{-1}, and the annual average number of windy-days is 38 d. Surrounding the lake is well-developed farmlands in high hilly land and high plains, with many soybean and other crops in the past.

The major drainage system in XL including three drainage ditches has brought together the slope flood and upstream agricultural drainage within the watershed. During the flood season, a large amount of sediment, straw, and other debris entered DXL, which renders a certain degree of pollution to the local natural environment causing water quality decrease at the inlet of DXL. To address the agricultural drainage, in 2000, the drainage water of the major drainage ditch was firstly diverted by a diversion lock dam to the restored wetland, then is re-imported to the major drainage system and finally flows into DXL (Fig. 1). The total area of the restored wetland (RW) for agricultural drainage treatment surrounded by embankments and roads is 1.31 km^2, with the surface water depth of 1.2–1.5 m and the maximum water storage of 1.96 million m^3 (Zheng and Shi, 2012). In this way, by

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