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Effects of water-saving irrigation on the residues and risk of polycyclic aromatic hydrocarbon in paddy field

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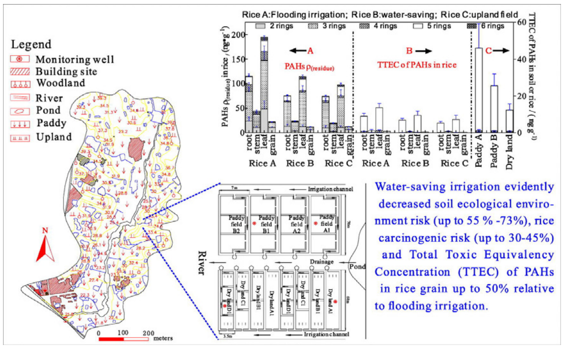
HIGHLIGHTS

- Water-saving effectively reduced the leaching quantity of PAHs into groundwater.
- Soil risk of PAHs was reduced up to 55%–73% by water-saving *vs.* flooding irrigation.
- Rice carcinogenic risk of PAHs was reduced up to 30–45% by water-saving.
- Total Toxic Equivalency Concentration of PAHs in rice grain was reduced about 50%.
- This is related to the change of paddy field environment due to moisture control.

GRAPHICAL ABSTRACT



Water-saving irrigation evidently decreased soil ecological environment risk (up to 55 % -73%), rice carcinogenic risk (up to 30-45%) and Total Toxic Equivalency Concentration (TTEC) of PAHs in rice grain up to 50% relative to flooding irrigation.

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ABSTRACT

The effects of different water-saving modes on PAHs residue and risk, field environment conditions and enzyme activities in paddy field were investigated in a field experiment plot in Laoyaba, Nanjing, China. Results showed that (1) water-saving treatment affected greatly the ΣPAHs in water and soil. The order of ΣPAHs residue in surface water and groundwater in farmland is as follows: dry fields < water-saving paddy field < flooding irrigation paddy field. The ΣPAHs in water during rice tillering stage were obviously higher than that in rice booting stage and milky stage, and the percentage of high-ring PAHs gradually reduced in water. (2) The residue of ΣPAHs in soil in flooding irrigation paddy field (534.4 ± 186.7 ng/g) were more than water-saving irrigation (454.3 ± 128.1 ng/g) and dry cultivation paddy field (430.2 ± 143.4 ng/g), and the ΣPAHs in dry field gradually decreased with the increase of water furrow number in farm. (3) When compared with flooding irrigation (337.87 ng/g), water-saving (228.39 ng/g) and dry cultivation (206.62 ng/g) could obviously decrease the residue of ΣPAHs in rice tissues (35%–55%), generally the concentration of ΣPAHs in leaf > root > stem > rice grain. (4) Water-saving irrigation evidently decreased soil ecological risk (up to 55%–73%) and rice carcinogenic risk (up to 30%–45%) caused by PAHs compared with flooding irrigation. Water-saving irrigation could also reduce the Total Toxic Equivalency Concentration of PAHs in rice grain up to 50% relative to flooding irrigation. (5) The significant negative correlations were observed between the residual PAHs and the activities of laccase and dioxygenase ($p < 0.019$), and the physical and chemical indexes (temperature, redox potential and dissolved oxygen of field, $p < 0.041$). The changes of field environment conditions and enzyme activities induced by moisture control may be the main key factors affecting PAHs residue in water, soil and rice.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are the most widely spread pollutants in environment. Their carcinogenic nature causes them to be named target pollutants (ATSDR, 1999). PAHs in soil pose a significant threat to ecological environment and human health, and are a main concern when seeking remedies for contamination (Jones et al., 1989). In recent years, there has been reports of PAHs toxicity causing human neural tube defects (Perera, 1997; Ren et al., 2011; Sanderson, 2011), birth defects (Sanderson, 2011) and cancers (Perera, 1997). Moreover, due to their bioaccumulation, semi-volatility, and persistence in environment, PAHs have become a serious concern of governments and scientific community (Nethery et al., 2012; Ren et al., 2011; Sanderson, 2011; Simonich and Hites, 1994; Zhang et al., 2009). Some carcinogenic PAHs have been listed in the environmental priority control pollutants blacklist by America, European Union, and China. Statistics show hundreds of thousands of tons of PAHs were discharged into atmosphere every year, and 99% of them transferred into soil through dry and wet deposition, which made soil become the repository for PAHs (Shannigrahi et al., 2005; Shen et al., 2011; Wang et al., 2009b). By now, farmland, especially paddy soil, has been widely polluted by PAHs, and the residues generally varied from ng/g to ng/mg. The concentration of PAHs in rice grain reached a dangerous level in some areas (13.2–85.3 ng/g, eight kinds of carcinogenic PAHs accounted for 3.9%, and the limited standard value of Benzo[*a*]pyrene in rice grain is 5 ng/g in GB7104094) (Ding et al., 2012; Ding et al., 2013; Jiao et al., 2010a; Jiao et al., 2010b; Tao et al., 2006). PAHs pollution has threatened the quality and safety of agricultural products. Diet is the primary source of human exposure to PAHs, with the major dietary contributors being cereals and vegetables (de Vos et al., 1990; Dennis et al., 1983; Phillips, 1999). Although PAH levels in cereals are often low, the large amounts consumed make grains a significant exposure source of PAHs for human beings (de Vos et al., 1990; Dennis et al., 1983; Menzie et al., 1992; Tao et al., 2006).

Paddy is an important land use scheme in world, playing a vital role in food security and the country's health (de Vos et al., 1990; Dennis et al., 1983; Menzie et al., 1992; Tao et al., 2006). In recent years, Studies have shown soil physical-chemical factors are important for the synthesis and reduction of PAHs. Soil moisture can change the PAHs retention form, soil nutrient status, physical and chemical characteristics (such as pH, redox potential), and microorganism activities (Li et al., 2005; Teng et al., 2010). Compared with flooding irrigation, water-saving irrigation can reoxygenate soil, raise redox potential, strengthen aerobic microbial activities, increase organic matter mineralization rate, increase the content of soil available nutrients due to regular draining and drying field (Li et al., 2005). In general, aerobic conditions are conducive to microbial activity increase and the remediation of soil contaminated by phenanthrene and pyrene (Teng et al., 2010). Wilson et al. found nitrate can continue degradation of aromatic hydrocarbons even if oxygen was depleted (Wilson and Bouwer, 1997). The long-term flooding of soil and plant material can significantly increase soil biosynthesis of 4–6 rings PAHs (139%–238%) under reducing conditions, and the content increases of 4-ring PAHs benzo[*a*]anthracene, chrysene and all 5- and 6-ring PAHs are more significant when only plant material was incubated (250%–674%) (Thiele and Brümmer, 2002). The environmental behavior of PAHs in soil is also closely related to the coexisting organic matter content and structure, clay minerals and their complexes, thereby facilitating PAHs desorption from soil (such as the adding of organic solvent, surfactant, humic acid, composting and organic materials), and efficient microbial activation can enhance the remediation of PAHs contaminated soil (Gao et al., 2011; Gao et al., 2010a; Gao et al., 2010b; Joner et al., 2001; Ma et al., 2012; Read et al., 2003).

For the low-ring PAHs, volatilization becomes a major pathway of its dissipation in soil. For example, Su et al. found the absorption contribution of rice and rhizosphere degradation contribution on naphthalene, phenanthrene and pyrene are not significant (0.24%–14%), after four months, the volatilization losses of three PAHs were 98%, 95%, and 30%, respectively (Su and Zhu, 2008). Du et al. also found gaseous loss is a main way to reduce naphthalene by isotope tracer method in wheat field. The mineralization losses accounted for about 84.41% of gaseous loss, the mineralization rate and evaporation rate were increased by 43.51% and 190.32%; crops significantly promoted the gaseous loss of naphthalene—this is related to the root exudate activity of the plants (Du et al., 2011). Sun et al. found that the abiotic loss of phenanthrene and pyrene in low organic soil (3.75 g/kg) were 83.4% and 57.2%, respectively. The root exudates strengthened their degradation of phenanthrene (15.5%) and pyrene (21.3%), and the contribution of root secretion grew more important with the increase of PAHs ring number (Sun et al., 2010). Rhizosphere effect is still a major acceleration mechanism of PAHs dissipation. Rice roots can promote PAHs degradation and rhizosphere microbial biomass, reduce inhibitory effects of PAHs on microbial activity (Su and Yang, 2009). Root exudates contribute to the desorption of PAHs and the formation of dissipation gradient in rhizosphere, the degradation rate of rhizosphere and root zones of PAHs compared with non-root zones (Gao et al., 2011; Gao et al., 2010a; Gao et al., 2010b; Joner et al., 2001; Ma et al., 2012; Read et al., 2003).

In recent years, there has been considerable interest by environmental scientists in the uptake of PAHs by plants (Jiao et al., 2010a; Jiao et al., 2007; Jiao et al., 2010b), both from field surveys and laboratory experiments, in order to develop better insight into the process of plant uptake of PAHs and to provide quantitative descriptions of different pathways of such accumulation (Tao et al., 2006). However, there is little research concerning the effects of different water-saving modes on the residual tendency of PAHs in soil, water and rice tissues during crop growth periods. The present investigation sought to determine the impact of moisture control (different water-saving modes: routine flooding irrigation, water-saving irrigation, and dry cultivation) on the levels and distribution patterns of PAHs in paddy field. Besides, to explore the possible mechanism of the change of field environment conditions affecting PAHs distribution and dissipation based on the moisture control and field practices, with the monitoring of PAHs residue, physical and chemical indexes, and the activities of laccase and dioxygenase.

2. Materials and methods

2.1. Experimental materials and study area

The rice species is Suyou-22 (*Oryza sativa* L.). The field experimental area (about 2×10^4 m^2) is located at Laoyaba irrigation area in Chahe agricultural small watershed, about 16 km from southeast of Lishui County in Nanjing, China (N 31°34′,E 119°10′). Average annual rainfall is 1087.4 mm, and approximately 52.6% of the rainfall is concentrated in rice growth stage from June to September. The regional natural soil belongs to yellow brown soil, and the anthropogenic soil is mainly hydragric paddy field soil, its physical and chemical properties are as follow: pH 5.5, clay content is 24.1%, organic matter content is 1.38%, and soil bulk density is 1.29 g/cm^3

The arrangement plan of field experiments was shown in Fig. 1. It included conventional flooding irrigation paddies A (A1 and A2), water-saving irrigation paddies B (B1 and B2), and four kinds of upland field (dry land A, B, C and D, with 0, 1, 2, 3 water furrows in the field, respectively). Every land treatment has three parallel fields. In the middle of each field, there is one groundwater monitoring well of 1 m depth, covered with plastic film to avoid the effect of rainfall and atmospheric deposition in non-sampling period. A pond is located in the east of the experimental fields, which used for farm land drainage and irrigation water source in dry period, and a small stream near the west of experimental fields. The irrigation water comes from a reservoir near the studied area.

The control parameters of field capacity for three kinds of paddy fields were shown in Table 1.

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