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Short-term transport of glyphosate with erosion in Chinese loess soil — A flume experiment

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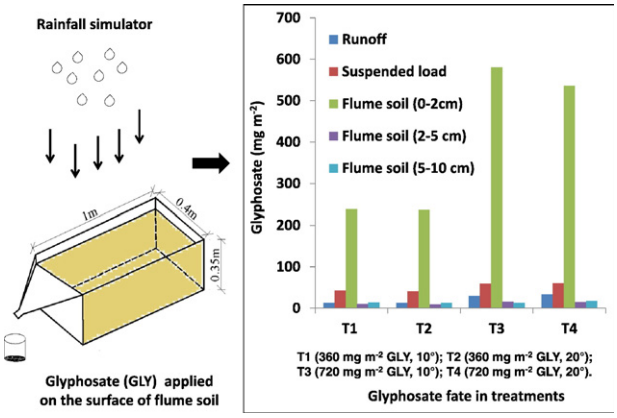
HIGHLIGHTS

- Glyphosate transport with erosion in Chinese loess soil was studied.
- Glyphosate and AMPA in runoff and suspened load decreased with rainfall duration.
- Particulate facilitated transport is the main mode for glyphosate transport.
- The risk of glyphosate retained in upper 2 cm soil is higher than in deeper soils.

GRAPHICAL ABSTRACT



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ABSTRACT

Repeated applications of glyphosate may contaminate the soil and water and threaten their quality both within the environmental system and beyond it through water erosion related processes and leaching. In this study, we focused on the transport of glyphosate and its metabolite aminomethylphosphonic acid (AMPA) related to soil erosion at two slope gradients (10 and 20°), two rates of pesticide with a formulation of glyphosate (*Roundup*®) application (360 and 720 mg m^{-2}), and a rain intensity of 1.0 mm min^{-1} for 1 h on bare soil in hydraulic flumes. Runoff and erosion rate were significantly different within slope gradients ($p < 0.05$) while suspended load concentration was relatively constant after 15 min of rainfall. The glyphosate and AMPA concentration in the runoff and suspended load gradually decreased. Significant power and exponent function relationship were observed between rainfall duration and the concentration of glyphosate and AMPA ($p < 0.01$) in runoff and suspended load, respectively. Meanwhile, glyphosate and AMPA content in the eroded material depended more on the initial rate of application than on the slope gradients. The transport rate of glyphosate by runoff and suspended load was approximately 14% of the applied amount, and the chemicals were mainly transported in the suspended load. The glyphosate and AMPA content in the flume soil at the end of the experiment decreased

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significantly with depth ($p < 0.05$), and approximately 72, 2, and 3% of the applied glyphosate (including AMPA) remained in the 0–2, 2–5, and 5–10 cm soil layers, respectively. The risk of contamination in deep soil and the groundwater was thus low, but 5% of the initial application did reach the 2–10 cm soil layer. The risk of contamination of surface water through runoff and sedimentation, however, can be considerable, especially in regions where rain-induced soil erosion is common.

1. Introduction

With the increasing use of agrochemicals, the threat to environments and human health is receiving more attention. Glyphosate (N-(phosphonomethyl)glycine, $C_3H_8NO_5P$), a highly effective broad-spectrum herbicide, is widely used around the world in agriculture, horticulture, parks, and domestic gardens, especially in the cultivation of genetically modified crops (Candela et al., 2007). As a systemic herbicide, glyphosate is intercepted and taken up by the foliage and then enters plant physiological processes that transport it or its principal metabolic product, aminomethylphosphonic acid (AMPA), to the root system, which releases them into the surrounding soils and waters (Laitinen et al., 2007). Applied to bare soil before or after sowing, glyphosate directly reaches the soil and underlies the highest risk of being transported with soil erosion and runoff (Todorovic et al., 2014). The elimination of the glyphosate in the soil mainly depends on microbial degradation through two pathways, one leading to the intermediate formation of sarcosine and glycine, and the other leading to the formation of AMPA (Rueppel et al., 1977). The half-life of a chemical is an important parameter for assessing environmental threats, but estimates of the half-life of glyphosate have ranged from days to months (Al-Rajab and Hakami, 2014; Bergström et al., 2011; Mamy and Barriuso, 2007; Rueppel et al., 1977). The rate of AMPA degradation is also controversial, half-life time ranging from 35 to 151 days (Bergström et al., 2011; Borggaard and Gimsing, 2008; De Jonge et al., 2000). The fate and quantification of glyphosate thus are required to evaluate their threats and to determine the specifics of glyphosate application and related pesticide management (Todorovic et al., 2014; Zablotowicz et al., 2009).

Glyphosate strongly adsorbs on soil particles by ligand exchange through the phosphonic acid moiety (Sheals et al., 2002; Sorensen et al., 2006; Sprankle et al., 1975). Bonded residues of glyphosate are not considered to be bioavailable and therefore not harmful to the environment onsite (Barriuso et al., 2008), but if transported with soil erosion to surface water, the bond residues then enter into the aquatic food chain (Sihtmae et al., 2013). Soil properties and climate also influence the mobility and interactions of glyphosate (Gjettermann et al., 2009; Sorensen et al., 2006; Wang et al., 2005; Zhao et al., 2009; Zhou et al., 2010). Absorbed glyphosate and AMPA can be desorbed at the water–soil interface (Borggaard and Gimsing, 2008; Candela et al., 2007; Coupe et al., 2012; Donald, 2002; Passeport et al., 2014), and competition with phosphates for adsorption sites may lead to free glyphosate rather than the bound form in the soil matrix (Borggaard and Gimsing, 2008; Gimsing et al., 2004; Zhou et al., 2010). The free forms of glyphosate and AMPA are thus easily dispersed, especially in wet soils due to preferential flow (Vereecken, 2005), and heavy rains shortly after glyphosate application increase the entry of glyphosate to surface water bodies through transport with runoff and suspended load (Botta et al., 2009; Candela et al., 2010; Gjettermann et al., 2009; Peruzzo et al., 2008; Stone and Wilson, 2006; Vereecken, 2005).

Luijendijk et al. (2003) reported that up to 24% of the glyphosate sprayed on hard surface soil was transported in runoff to surrounding fields, and Todorovic et al. (2014) showed that approximately 47% of applied glyphosate was transported in the runoff associated with erosion and tillage managements (plough or not). The solubility of glyphosate contributes much to its contamination of surface water, but glyphosate and AMPA bound to particles suspended in water is another means of glyphosate and AMPA transport, known as particle-facilitated transport (Rügner et al., 2014; VandeVoort et al., 2013). Degenhardt et al. (2012) reported that 67% of added glyphosate was detected in wetland sediment monitoring within 77 days from June to September. Leaching with drainage is another method of glyphosate and AMPA transport that may lead to potential contamination of, and accumulation in, groundwater (Kjær et al., 2011; Ruiz-Toledo et al., 2014; Ulén et al., 2012). Bergström et al. (2011) reported that 0.009 and 0.019% of the glyphosate and AMPA, respectively, were found in leachate samples of a clay soil after 748 days of monitoring in fields. Landry et al. (2005) demonstrated that 0.02–0.06% of applied glyphosate was leached from a calcareous soil column, and Al-Rajab et al. (2008) reported that 0.28, 0.20, and 0.11% of an initial application of ^{14}C-glyphosate were found in leachates of clay loam, silty clay loam, and sandy loam soils, respectively. Leaching occurs after short heavy rains, attributed to an increase in the probability of leaching through soil macropores, especially in unstructured soils (Gjettermann et al., 2009; Kjaer et al., 2005; McGechan, 2002; Stone and Wilson, 2006; Styczen et al., 2011; Vinten et al., 1983). Based on 28 months of field monitoring, 99% of the lost glyphosate (5.12 g ha^{-1}) was found in the runoff, and glyphosate and AMPA accounted for 0.51 and 0.07%, respectively, of the applied glyphosate (Laitinen et al., 2009). Limited amounts of glyphosate and AMPA are found in leachates and runoff, but the potential risk of contamination in ground/surface water is often not considered, especially by suspended particles (Ruiz-Toledo et al., 2014).

China has become the largest glyphosate supplier in the world, and the thousands of tons of glyphosate-based herbicides are applied to agricultural land each year (Dill, 2005; Zhang et al., 2011). The repetitive use of glyphosate-based herbicides in the field, however, increases the possibility of glyphosate occurrence in, and threat to, soil, plants, surface/ground water (Borggaard and Gimsing, 2008), and animals (Lanctot et al., 2013; Muangphra et al., 2014; Yadav et al., 2013; Zaller et al., 2014). Meanwhile, the risk is particularly high when applied pesticide on bare soil directly before and after sowing. Intensive cultivation with concentrated precipitation releases substantial amounts of agrochemicals, especially on the Loess Plateau in China where soil erosion is common (Shi and Shao, 2000). Nutrients and pesticides in runoff and suspended particles have become the main factors determining water quality, especially in rural regions along rivers (Li et al., 2011). Many studies have focused on glyphosate adsorption, degradation, and leaching in environments, but little is known about glyphosate transport associated with soil erosion (Borggaard and Gimsing, 2008; Donald, 2002). Laitinen et al. (2009) and Todorovic et al. (2014) had experimented (plot observations) under field conditions but only reported glyphosate transport by runoff. Thus, the proposal of the present study was to quantify the transport of glyphosate and AMPA associated with runoff and erosion in Chinese loess soils. The proportion of glyphosate was estimated in runoff, suspended load and soils and the risks for offsite pollution was discussed due to the threats to neighbouring areas of glyphosate application followed directly by rain.

2. Materials and methods

2.1. Experimental design

2.1.1. Facilities and soils

The experiment was conducted in an artificial rain-simulation facility. Rain intensity was adjusted by nozzle size and water pressure and was calibrated prior to the experiment. The experiment was conducted