



Phosphate fertilizer impacts on glyphosate sorption by soil



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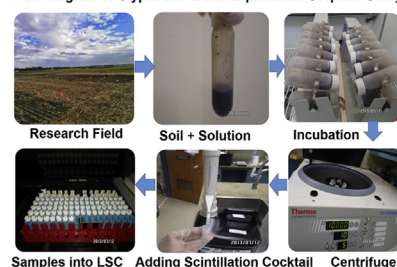
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HIGHLIGHTS

- Long-term additions of phosphate fertilizers to soil reduced glyphosate sorption.
- Co-application of phosphate with glyphosate reduced glyphosate sorption.
- Sorption was the same for analytical-grade glyphosate and Roundup Ultra 2.
- Cd impurities in phosphate fertilizers had no impact on glyphosate sorption.

GRAPHICAL ABSTRACT

Flow Diagram of Glyphosate Batch Equilibrium Sorption Study



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ABSTRACT

This research examined the impact of field-aged phosphate and cadmium (Cd) concentrations, and fresh phosphate co-applications, on glyphosate sorption by soil. Soil samples were collected in 2013 from research plots that had received, from 2002 to 2009, annual applications of mono ammonium phosphate (MAP) at 20, 40 and 80 kg P ha⁻¹ and from products containing 0.4, 70 or 210 mg Cd kg⁻¹ as an impurity. A series of batch equilibrium experiments were carried out to quantify the glyphosate sorption distribution constant, K_d. Extractable Cd concentrations in soil had no significant effect on glyphosate sorption. Glyphosate K_d values significantly decreased with increasing Olsen-P concentrations in soil, regardless of the pH conditions studied. Experiments repeated with a commercially available glyphosate formulation showed statistically similar results as the experiments performed with analytical-grade glyphosate. Co-applications of MAP with glyphosate also reduced the available sorption sites to retain glyphosate, but less so when soils already contain large amounts of phosphate. Glyphosate K_d values in soils ranged from 173 to 939 L kg⁻¹ under very strong to strongly acidic condition but the K_d was always <100 L kg⁻¹ under moderately acidic to slightly alkaline conditions. The highest Olsen-P concentrations in soil reduced K_d values by 25–44% relative to control soils suggesting that, under moderately acidic to slightly alkaline conditions, glyphosate may become mobile by water in soils with high phosphate levels. Otherwise, glyphosate residues in agricultural soils are more likely to be transported off-site by wind and water-eroded sediments than by leaching or runoff.

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1. Introduction

Glyphosate [*N*-(phosphonomethyl) glycine] is a broad spectrum, non-selective systemic, post-emergent herbicide introduced for agricultural use in the 1970s. Glyphosate-tolerant crops were

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commercialized in 1996 (Benbrook, 2012) and glyphosate now accounts for about one-fourth of global herbicide sales (GSBR, 2011). The glyphosate molecule contains a phosphonic acid moiety that facilitates molecule sorption by soil (Sprankle et al., 1975). With more than 40 million tons of phosphate fertilizers applied on agricultural land around the world (FAO, 2012), a wide range of studies have focused on phosphate and glyphosate interactions in soil (de Jonge et al., 2001; Gimsing and Borggaard, 2007; Gimsing et al., 2007; Kanissery et al., 2015). Glyphosate and inorganic phosphate have shown to compete for the same sorption sites (de Jonge and de Jonge, 1999) and hence phosphorus applications may influence the bioavailability and transport potential of glyphosate in soil (Gimsing and Borggaard, 2002a). Phosphate fertilizers contain cadmium (Cd) as an impurity and repeated application of phosphate fertilizer results in Cd accumulation in soil (Lambert et al., 2007). The addition of Cd to glyphosate solutions in the laboratory has been shown to enhance glyphosate sorption by soil because Cd and glyphosate form complexes that are retained on the surface of negatively-charged soil colloids (Zhou et al., 2004). The effect of impurities of Cd in phosphate fertilizers on glyphosate sorption in soil is not known.

The batch equilibrium procedure is a common method to examine the effect of phosphate additions on glyphosate sorption. All researchers, except de Jonge et al. (2001), added phosphate to soil in the laboratory prior to or during the batch equilibrium experiment, and hence exclusively studied the effect of “fresh” phosphate on glyphosate sorption. Phosphate did compete with glyphosate sorption in a wide range of soils (Sprankle et al., 1975; de Jonge and de Jonge, 1999; de Jonge et al., 2001; Kanissery et al., 2015), but not in other soils (Gimsing et al., 2004). Some research demonstrates that phosphate is preferentially sorbed by clay minerals (Gimsing and Borggaard, 2001), for example because the phosphate molecule (0.25 nm) is smaller than the glyphosate molecule (0.43 nm) (Cáceres-jensen et al., 2009). The impact of phosphate on reducing glyphosate sorption was stronger for synthesized Fe and Al-oxides than for pure clay-minerals (Gimsing and Borggaard, 2002b). All batch equilibrium studies utilized analytical-grade glyphosate, while herbicide products applied on agricultural land contain other ingredients that could impact the sorption of the active ingredient by soil (Farenhorst and Bowman, 1998). Hence, in evaluations of the impact of phosphate additions on glyphosate sorption, it is desired to include a commercially available glyphosate formulation in the study.

Borggaard (2011) reported that the interaction of phosphate and glyphosate sorption remains unclear, even though this interaction has been studied for four decades. de Jonge et al. (2001) is the only study in these four decades to have examined the effect of “aged” phosphate on glyphosate sorption. Utilizing soil from field plots that had received various amounts of phosphate and lime fertilizers for more than 60 years, batch equilibrium studies indicated that less glyphosate was sorbed in soils with greater Olsen-P concentrations. To the best of our knowledge, the combined effect of fresh and aged phosphate on glyphosate sorption in soil has not been investigated.

The objective of this study was to examine under a range of pH conditions, the impact of field-aged phosphate and cadmium concentrations on glyphosate sorption by soil, with and without phosphate co-applications in the laboratory. The pH conditions were chosen to include the range of possible glyphosate speciations, as described by Wang et al. (2004). In general, studies have reported that glyphosate sorption generally decreases with increasing soil pH (McConnell and Hossner, 1985; de Jonge and de Jonge, 1999; Gimsing et al., 2004).

2. Materials and methods

2.1. Experimental design and soil characteristics

Soil samples (0–15 cm) with a sandy clay loam texture were collected in the spring 2013 from research plots situated under a durum wheat and flax rotation near Carman (49° 29.7' N, 98° 2.4' W), Manitoba, Canada. The soil is classified as an Orthic Black Chernozem based on the Canadian System of Soil Classification, which is approximately equivalent to the Udic Boroll subgroup in the U.S. Soil Taxonomy (CSCC, 1998). The experimental plot was a randomized complete block design with 10 treatments and 4 replicates per treatment. In each of the forty plots, the composite soil sample consisted of ten samples collected in the plot using a Dutch augur. Treatments were a control (neither phosphate nor Cd applications), and plots receiving from 2002 to 2009 annual applications of mono ammonium phosphate (MAP) fertilizers that originated from three different phosphate rock sources containing 0.4, 70 or 210 mg Cd kg⁻¹, or low, medium and high Cd, respectively (Grant et al., 2013). MAP from these three sources was applied to plots at 20, 40 and 80 kg P ha⁻¹, or 20P, 40P and 80P, respectively. For all plots that received MAP, 20 kg P ha⁻¹ was placed near the seed to enhance fertilizer use efficiency, a common practice in Canadian Prairie agriculture. For the 40 and 80 kg P ha⁻¹ treatments, to avoid seedling toxicity, the additional MAP was broadcasted and then incorporated in soil. From 2010 to 2013, the rotation was continued but no phosphate or Cd was applied. Nitrogen fertilizer varied by year to optimize yields. The typical rate of N applied was 90 kg N ha⁻¹ in durum wheat and 50 kg N ha⁻¹ in flax.

Soil samples were air-dried and sieved (<2 mm) prior to soil properties analysis and sorption studies. Soil was digested with nitric acid and total Cd was determined by *inductively coupled plasma* (ICP) (U.S. EPA, 1996). Extractable Cd was extracted with *diethylene triamine pentaacetic acid* (DTPA) following procedures as recommended by Lindsay and Norvell (1978), and extracts were analyzed by ICP (Whitney, 2011). Various factors have been shown to influence the efficiency of micronutrient extraction by DTPA, including extraction temperature and shaking time (Lindsay and Norvell, 1978).

Available phosphate was extracted using Olsen (NaHCO₃) phosphorus test (Frank et al., 2011). Soil physical and chemical properties that are known to influence glyphosate and phosphate sorption by soil, but did not significantly vary across the plots by treatment, were also determined. Soil organic carbon content was determined using combustion technique with a high temperature induction furnace (Nelson and Sommers, 1996). Extractable Fe₂O₃ and Al₂O₃ were extracted with DTPA (Whitney, 2011) and 0.01 M CaCl₂ (Barnhisel and Bertsch, 1982), respectively, and extracts were analyzed by ICP. Extractable Ca was also measured by ICP using ammonium acetate as an extractant (Warncke and Brown, 2011). Results were soil organic carbon content: 2.80% (mean) ± 0.04 (standard error) (n = 16, number of plots analyzed); extractable Fe₂O₃: 246 ± 5 mg kg⁻¹ (n = 40); extractable Al₂O₃: 6.4 ± 0.65 mg kg⁻¹ (n = 16); and extractable Ca: 2252 ± 40.57 mg kg⁻¹ (n = 16). Given that the study focused on Cd and P applications as treatments, the concentrations of extractable and total Cd, as well as Olsen-P in all plots were determined. We did not expect to see treatment differences for the other parameters that were measured (i.e., extractable Fe₂O₃, Al₂O₃, and Ca). Fe₂O₃ was also measured in all plots as previous studies have demonstrated that there is a strong positive association between Fe₂O₃ concentrations and phosphate or glyphosate sorption in soils. Since our results indicated no treatment differences induced by Cd and P applications on Fe₂O₃ concentrations extractable Al₂O₃, and Ca

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