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Overcoming Hard Water Antagonistic to Glyphosate or Imazethapyr with Water Conditioners

Akbar ALIVERDI[1]*, Ali GANBARI[1], Mohammad-Hassan RASHED MOHASSEL[1], Mehdi NASSIRI-MAHALLATI[1], Eskandar ZAND[2]

[1] *Department of Agronomy and Plant Breeding, College of Agriculture, Ferdowsi University of Mashhad, Mashhad, Iran*
[2] *Department of Weed Research, Plant Protection Research Institute, Tehran, Iran; akbar.aliverdi@stu.um.ac.ir n (*corresponding author)*

Abstract

Carrier water quality may affect the activity of weak acid herbicides when concentrations of some cations are high. A dose-response experiment on glyphosate and imazethapyr activity, which were carried by the carrier types of distilled water and hard water, against jimsonweed were conducted to compare the water conditioning chemicals ammonium sulfate, ammonium nitrate, citric acid and potassium phosphate, with magnetized carrier as a new method. A magnetic field of 0.7 Tesla was applied to prepare the magnetized carrier. With the exception of potassium phosphate with imazethapyr, the activity of glyphosate and imazethapyr was significantly increased in the presence of the water conditioning methods when distilled water was used as the carrier. Ammonium sulfate was the most effective method. The activity of both herbicides was decreased when applied with hard water carrier. Potassium phosphate was not effective at reducing the antagonism of cations in the hard water carrier. In glyphosate, the performance of water conditioning methods in softening hard water carrier could be ranked as follows: ammonium sulfate (2.52-fold) > magnetized carrier (2.12-fold) ≥ citric acid (1.64-fold) ≥ ammonium nitrate (1.39-fold) > potassium phosphate (0.96-fold). In imazethapyr, this order was as follows: ammonium sulfate (2.99-fold) > ammonium nitrate (2.66-fold) > magnetized carrier (1.81-fold) ≥ citric acid (1.64-fold) > potassium phosphate (1.10-fold).

Keywords: adjuvant, herbicide efficacy, jimsonweed, magnetized carrier

Introduction

Water is the most frequently used carrier for herbicide applications. Thus, its physicochemical properties in spray mixture can affect the absorption and/or activity of herbicides (Hoffmann *et al*., 2008). A high concentration of cations (e.g. Na^+, K^+, Ca^{2+}, Mg^{2+}, and Fe^{3+}) in water can decrease herbicide efficacy. Weak acid herbicides that have been antagonized by one or more of the above cations include sethoxydim (Matysiak & Nalewaja, 1999; Nalewaja *et al*., 1989), glyphosate (Nalewaja and Matysiak, 1991; Bernards *et al*., 2005; Bailey *et al*., 2002), 2,4-D (Roskamp *et al*., 2013), clethodim (Nandula *et al*., 2007), imazethapyr (Gronwald *et al*., 1993), tralkoxydim (DeVilliers *et al*., 2001), and glufosinate (Pline *et al*., 2000). This antagonism is due to the formation of herbicide salts with a low solubility, resulting in a reduction in retention on the leaves (Hoffmann *et al*., 2008) and/or a reduction in absorption into the plant (Nalewaja *et al*., 1996). As a result, a decrease in herbicidal performance is observed. When 2,4-D amine is mixed with hard water, a sediment may form in lines, filters, nozzles or the bottom of the spray tank (Nalewaja *et al*., 1991). Approaches to minimize hard water antagonism have included decreasing the spray carrier volume (Wills *et al*., 1998) and using water-conditioning additives (Thelen *et al*., 1995a) which have proven effective at ameliorating cation-caused antagonism include ammonium sulfate (Soltani *et al*., 2011), ammonium nitrate, potassium phosphate (Wills *et al*., 1998), and citric acid (Thelen *et al*., 1995b).

Passing hard water containing Ca^{2+} (Kobe *et al*., 2001; Kobe *et al*., 2002; Plavic *et al*., 1999; Cho and Choi, 1998; Madsen, 2004; Coey and Cass, 2000), Mg^{2+} or Na^+ (Bin *et al*., 2011) through an external magnetic device results in the nucleation and crystallization of the respective carbonates. As a result, hard water can be conditioned for a period known as the 'magnetic memory of water' (Colic and Morse, 1999). Magnetized water may induce a change in water physicochemical properties such as conductivity (Sueda *et al*., 2007), surface tension (Cho and Lee, 2005), viscosity (Quinn *et al*., 1997), vaporization rate (Toledo *et al*., 2008), and pH (Fathi *et al*., 2006). All the aforementioned physicochemical factors can affect the performance of post-emergence herbicides (Rao, 2000).

The objectives of this research were (i) to investigate the effect of hard water on glyphosate and imazethapyr efficacy, and (ii) to compare chemical water conditioners (ammonium sulfate, ammonium nitrate, citric acid and potassium phosphate) with a physical method that consist spray carrier passage through a magnetic field for ameliorating the antagonistic effect of hard water on glyphosate and imazethapyr activity against jimsonweed.

Materials and methods

Plant growth

Seeds of jimsonweed (*Datura stramonium* L.) were collected from plants in the fields of Qazvin city, Iran, during November 2012. They were stored in the dark at room temperature until use. Bioassays were conducted between June 2013 and September 2013 in a greenhouse located on the Ferdowsi University of Mashhad, Iran. To increase seed germination before starting the experiment, the seeds were washed every 1 hour for 7 days to remove seed germination inhibitors, according to Andersen (1968). Twenty-five jimsonweed seeds were sown at 0.5 cm depth in 2 L plastic pots filled with a mixture of sand, clay loam soil, and peat (1:1:1 v/v). Up to 88% of the seeds were emerged. At the one- and two-leaf stages, the seedlings were thinned to a final population of four uniform plants per pot. After each thinning, plants were fertilized twice with 20 mL of a water-soluble N:P:K (20:20:20) fertilizer, at a concentration of 3 g of fertilizer per liter of tap water. The pots were irrigated every three days with tap water. Treatments were sprayed at the four-leaf stage.

Treatments and chemicals

The treatments were six doses of glyphosate (0, 12.81, 25.62, 51.25, 102.5 and 205 g a.i. ha^{-1}) or imazethapyr (0, 4.69, 9.38, 18.75, 37.5 and 75 g a.i. of imazethapyr ha^{-1}) which were carried by the carrier types included of: (i) distilled water and (ii) hard water with and without the water conditioning methods included of: (i) ammonium sulfate ($(NH_4)_2SO_4$) at 20 g L^{-1}; (ii) ammonium nitrate (NH_4NO_3) at 20 g L^{-1}; (iii) potassium phosphate ($KHPO_4$) at 20 g L^{-1}; (iv) citric acid ($C_6H_8O_7$) at 20 g L^{-1}; and (v) passing 10 times through a magnetic field. Glyphosate isopropylamine (41% SL) and imazethapyr (10% SL) were used. The concentrations of the water conditioning chemicals were selected based on recommendations of Somervaille *et al.* (2012) and Wills *et al.* (1998). After checking the water samples of Mashhad region, Iran, the Sand-Washing Well in Tangal-shor village located at 36.20° North latitude and 59.35° East longitude was chosen to obtain hard water. The chemical composition of hard water was: Na^+ = 3722 ppm, K^+ = 14 ppm, Ca^{2+} = 501 ppm, Mg^{2+} = 573 ppm, SO_4^- = 56.1 meq. L^{-1}, CO_3^- = 0.5 meq. L^{-1}, HCO_3^- = 3.1 meq. L^{-1}, Cl^- = 165.1 meq. L^{-1}, EC = 23.0 dS. m^{-1}, and pH = 8.4. Treatments were applied at 180 L ha^{-1} at 200 kPa using a calibrated moving boom sprayer with an 8002 flat-fan nozzle (Spraying Systems Co., Wheaton, IL). A magnetic treatment device was devised and used for magnetizing the carriers. The arrangement of the permanent magnetic field and the dimensions of its different parts are shown in Fig. 1, which was involved a 20 cm copper tube with a rectangular channel (21 mm length and 1 mm width) flowed the solution between two pieces of magnet placed on either side of the copper tube. The maximum strength of the magnetic field was measured to be up to 0.7 Tesla by using a magnetometer (Model 1-ST; AlphaLab, Salt Lake City, Utah, USA). Gabrielli *et al.* (2001) indicated that the influence depend upon the pathway length in the magnetic field and the flow rate. Therefore, the carriers were passed 10 times through the magnetic device to magnetize effectively. Shoots were harvested four weeks after treatment, dried for 48 h at 70°C, and dry weight was determined. The experiment was performed again under the same conditions.

Statistical analysis

Data of shoot dry weight were subjected to a non-linear regression analysis for determination of ED_{10}, ED_{50}, and ED_{90} values (herbicide dose needed to obtain 10%, 50%, 90% reduction in dry weight, respectively) using the following logarithmic logistic dose-response model (Ritz & Streibig, 2005):

$$Y = C + \{D - C / 1 + \exp[B(\log X - \log E)]\}$$

Where Y is the response (shoot dry weight), C is the lower limit, D is the upper limit, B is the slope of the curve, E is the required dose of herbicide to give 50% control; and X is the herbicide dose. The validity of the model 1 and the comparisons between the parameters were made using an F-test for lack-of-fit at the 0.05% level of significance. If no significant lack of fit was detected when model 1 was tested, the model was acceptable. The relative potency (R) was the horizontal displacement between the two curves and was calculated using the ratio of doses producing the same response, as follows:

$$R = ED_{50a} / ED_{50b}$$

where ED_{50a} is for the treatment of 'a' and ED_{50b} is for the treatment of 'b'. If R < 1, treatment 'a' is more potent than treatment 'b' and if R > 1, the reverse is correct. Data were analyzed to determinate the parameters by open-source statistical software, $R_{2.6.2}$, utilizing the *drc* statistical addition package. By using PROC PROBIT in SAS software, data of ED_{50} were subjected to analysis of each experiment in confidence interval of 95% to determine whether the two runs of the experiments differed from each other or not. The results of one run will be discussed, as these were similar both runs.

Results

The ED_{10}, ED_{50}, and ED_{90} values in distilled water for jimsonweed were 11.40, 35.47, and 109.14 g a.i. of glyphosate ha^{-1}, respectively (Tab. 1). These values were 6.02, 13.95, and 32.34 g a.i. of imazethapyr ha^{-1}, respectively. The glyphosate ED_{90} rate under greenhouse conditions was about 53% of the recommended rate (205 g a.i. ha^{-1}) while the imazethapyr ED_{90} rate was about 57% of the recommended rate (75 g a.i. ha^{-1}) when distilled water was used as carrier. As judged by the relative potency values given in Tab. 1, the values were considerably higher than 1, indicating the enhanced glyphosate activity with addition/application of water conditioning methods when distilled water was used as carrier. With the exception of potassium phosphate with imazethapyr, the activity of both herbicides against jimsonweed was significantly increased in the presence of the water conditioning methods when distilled water was used as the carrier. They were ranked based on their performance to improve glyphosate activity as follows: ammonium sulfate (4.03-fold) > citric acid (2.81-fold) ≥ ammonium nitrate (2.35-fold) ≥ magnetized carrier (2.10-fold) ≥ potassium phosphate (1.97-fold). Moreover, they were ranked based on their performance to improve imazethapyr activity as follows: ammonium sulfate (3.52-fold) > ammonium nitrate = citric acid (2.58-fold) > magnetized carrier (1.83-fold) > potassium phosphate (1.28-fold).

Activity of both herbicides was decreased when applied

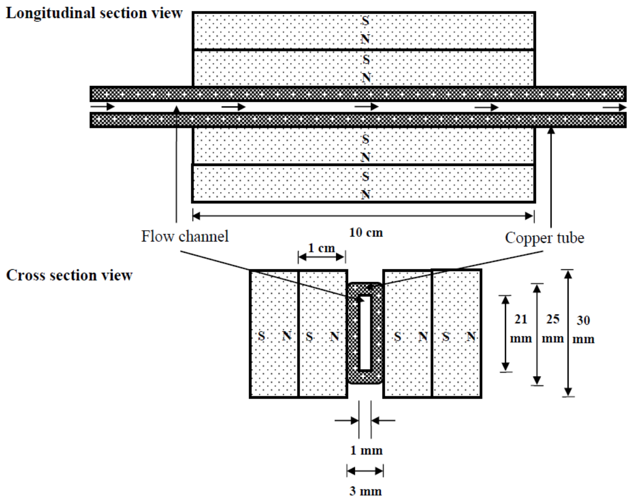

Fig. 1. Magnetic treatment device; N-North pole, S-South pole

Tab. 1. Estimated parameters for the response curves of jimsonweed treated with glyphosate or imazethapyr applied with the different carriers alone or in the presence of water conditioners

Herbicide	Carrier	Water conditioner	B	ED_{10} (g a.i. ha^{-1})	ED_{50} (g a.i. ha^{-1})	ED_{90} (g a.i. ha^{-1})	Relative potency	Relative potency
Glyphosate	Distilled water	Non	1.94 (0.37)	11.40 (2.41)	35.47 (4.31)	109.14 (9.47)	1.00 c	
		Ammonium sulfate	1.47 (0.60)	2.01 (1.42)	8.81 (1.81)	39.76 (8.12)	4.03 (0.93) g	
		Ammonium nitrate	2.34 (0.68)	5.88 (1.52)	14.98 (1.28)	38.19 (10.32)	2.35 (0.35) f	
		Citric acid	1.43 (0.54)	2.69 (1.51)	12.53 (1.77)	58.31 (3.42)	2.81 (0.52) f	
		Potassium phosphate	2.11 (0.69)	6.31 (1.61)	17.83 (1.78)	50.42 (5.17)	1.97 (0.31) e	
		Magnetized carrier	2.01 (0.58)	5.61 (1.63)	16.76 (1.76)	50.00 (6.44)	2.10 (0.33) ef	
	Hard water	Non	2.41 (0.55)	27.34 (5.07)	67.79 (8.63)	168.08 (8.90)	0.52 (0.09) a	1.00 a
		Ammonium sulfate	2.03 (0.45)	9.11 (2.24)	26.82 (3.06)	78.94 (12.71)	1.31 (0.23) de	2.52 (0.43) d
		Ammonium nitrate	2.12 (0.41)	17.24 (3.57)	48.53 (5.96)	136.61 (5.73)	0.72 (0.13) b	1.39 (0.24) b
		Citric acid	1.99 (0.37)	13.73 (2.80)	41.29 (5.79)	124.15 (5.65)	0.85 (0.16) bc	1.64 (0.31) bc
		Potassium phosphate	1.64 (0.40)	19.75 (4.79)	75.14 (7.52)	285.89 (7.61)	0.46 (0.12) a	0.96 (0.23) a
		Magnetized carrier	2.01 (0.39)	10.67 (2.41)	31.86 (3.98)	95.07 (5.74)	1.10 (0.20) c	2.12 (0.37) c
Imazethapyr	Distilled water	Non	2.61 (0.48)	6.02 (0.91)	13.95 (1.22)	32.34 (5.90)	1.00 d	
		Ammonium sulfate	1.93 (0.60)	1.27 (0.46)	3.96 (0.41)	12.33 (3.16)	3.52 (0.46) h	
		Ammonium nitrate	2.56 (0.72)	2.28 (0.47)	5.39 (0.39)	12.70 (2.74)	2.58 (0.27) g	
		Citric acid	2.27 (0.65)	2.05 (0.49)	5.39 (0.42)	14.15 (3.46)	2.58 (0.28) g	
		Potassium phosphate	2.64 (0.59)	4.10 (0.67)	9.41 (0.68)	21.58 (3.79)	1.28 (0.25) d	
		Magnetized carrier	2.13 (0.46)	2.73 (0.51)	7.63 (0.66)	21.36 (4.68)	1.83 (0.21) f	
	Hard water	Non	3.75 (0.69)	15.78 (1.89)	28.33 (2.20)	50.87 (7.44)	0.49 (0.06) a	1.00 a
		Ammonium sulfate	2.72 (0.61)	4.22 (0.83)	9.45 (0.79)	21.16 (4.23)	1.47 (0.18) e	2.99 (0.34) d
		Ammonium nitrate	3.09 (0.67)	5.23 (0.68)	10.63 (0.99)	21.61 (4.73)	1.41 (0.17) de	2.66 (0.32) d
		Citric acid	3.60 (0.84)	9.34 (1.56)	17.18 (1.15)	31.61 (4.65)	0.81 (0.09) c	1.64 (0.16) c
		Potassium phosphate	3.03 (0.71)	11.72 (1.85)	24.16 (2.30)	49.77 (1.30)	0.57 (0.04) b	1.10 (0.14) b
		Magnetized carrier	2.73 (0.60)	6.99 (1.26)	15.61 (1.42)	34.86 (7.61)	0.89 (0.11) c	1.81 (0.21) c

$(NH_4)_2SO_4$, NH_4NO_3, $C_6H_8O_7$, and $KHPO_4$ were used at 20 g L^{-1}. To magnetize the carrier, it was passed 10 times through a magnetic treatment device with a magnetic field of 0.7 Tesla. The parameter of b is the slope of the line. ED_{10}, ED_{50}, and ED_{90} are the required rate of herbicide to give 10%, 50%, and 95% control, respectively. The standard errors are in parentheses. In each column, relative potencies followed by same letter are not significantly different at the 0.05 probability level with the carrier of hard water (Fig. 2). Based on comparison to distilled water, the ED_{90}-values of glyphosate and imazethapyr increased about 35% and 37% when they were added to hard water, respectively. In both herbicides, the antagonisms caused by hard water were significantly ameliorated by the addition of ammonium sulfate, citric acid, and ammonium nitrate, and by the application of magnetizing the carrier (Tab. 1). Potassium phosphate was not effective at reducing the antagonism of cations in the hard water carrier. In glyphosate, the performance of water conditioning methods in softening hard water carrier could be ranked as follows: ammonium sulfate (2.52-fold) > magnetized carrier (2.12-fold) ≥ citric acid (1.64-fold) ≥ ammonium nitrate (1.39-fold) > potassium phosphate (0.96-fold). In imazethapyr, this order is partly changed as follows: ammonium sulfate (2.99-fold) > ammonium nitrate (2.66-fold) > magnetized carrier (1.81-fold) ≥ citric acid (1.64-fold) > potassium phosphate (1.10-fold).

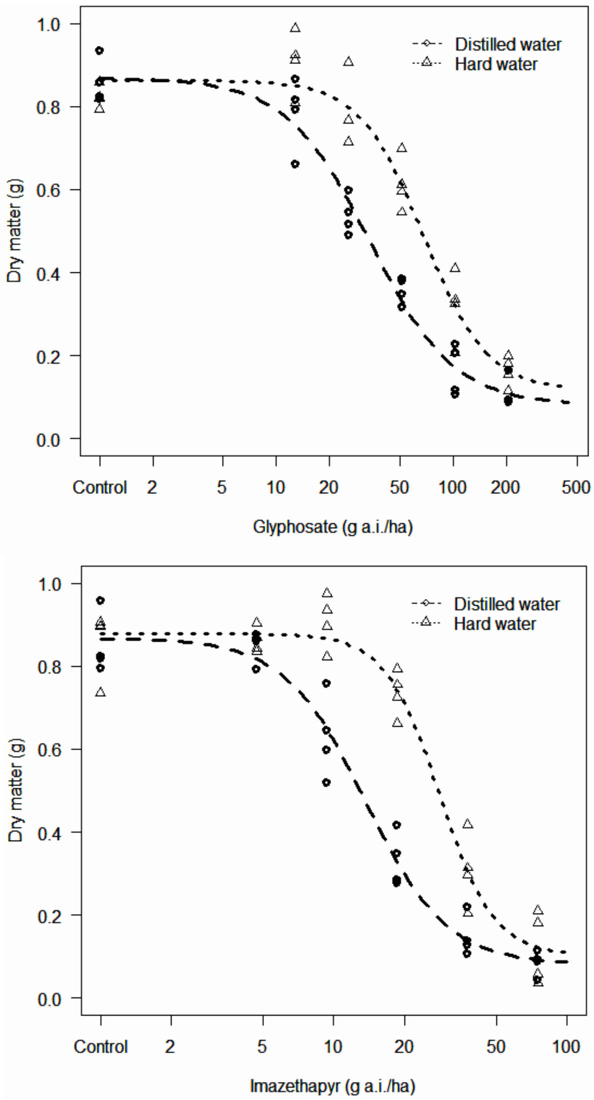

Fig. 2. Log-logistic dose-response for shoot dry weight of jimsonweed (*Datura stramonium* L.) to imazethapyr or glyphosate when they were applied with the carriers of distilled water (o) and hard water (Δ). The r^2 values for the regression model were 0.98 and 0.96 for distilled and hard water, respectively

Discussion

Enhancement of herbicidal activity by water conditioning chemicals in distilled water confirms the results by Nurse *et al.* (2008), Thelen *et al.* (1995a), Pline *et al.* (1999), and Molin and Hirase (2004). Although enhancement of glyphosate or imazethapyr activity by the application of magnetized carrier has not been reported, passing cycloxydim and clodinafop-propargyl solutions through a magnetic field did result in increased herbicide activity (Rashed-Mohassel *et al.*, 2009). They believed that the increased activity was related to a reduction in surface tension of the spray solution which then produces smaller droplets. Due to a lower level of energy existing in smaller droplets retention of the droplets is increased and the contact angle of the droplet on the leaf surface is reduced. These may increase absorption of active ingredient by the leaves (Penner, 2000). It is also reported that chemical water conditioning methods may adjust the spray solution pH so that more of the active ingredient is transported across the leaf surface and into the plant (Pline *et al.*, 1999).

Activity of both herbicides was clearly antagonized in our bioassays by the presence of Na^+, Mg^{2+}, and Ca^{2+} in the hard water used. This confirms data reported by Pratt et al. (2003) and Nurse *et al.* (2008). The dose-response curves with distilled water and hard water were not parallel and suggested that the antagonism was greatest at lower glyphosate or imazethapyr doses. Because the glyphosate or imazethapyr molecule has a negative charge in alkali solution, they can react with the cations in spray solution; leading to the formation of glyphosate-salt or imazethapyr-salt complexes which having a greater difficulty being absorbed into the plant (Pratt *et al.*, 2003). In addition, the presence of the cations in spray solution increases the droplet size, allowing less retention and/or absorption of active ingredient by the leaves (Hoffmann *et al.*, 2008).

The antagonism caused by the cations existing in hard water carrier in the tested herbicides was significantly overcome by all water conditioning methods, except potassium phosphate. The ability of water conditioners to negate glyphosate incompatibilities with cations in water has been reported (Nalewaja and Matysiak, 1993). Ammonium sulfate was the most successful method of ameliorating decreased herbicidal activity due to antagonism with Na^+, Mg^{2+}, or Ca^{2+} in the spray solution. By adding ammonium sulfate, the sulfate ion (SO_4^-) conjugates with the hard water cations and removes free cations from solution by forming cation-SO_4 molecule, allowing ammonium ion (NH_4^+) to form herbicide-NH_4 molecule (Thelen *et al.*, 1995a). Similarly, by adding ammonium nitrate, the nitrate ion (NO_3^-) conjugates with the hard water cations to form cation-NO_3 molecule, thus allowing NH_4^+ to form herbicide-NH_4 molecule (Penner, 2006). A herbicide-NH_4 molecule diffuses across the cuticle easier and quicker than the herbicide-salt molecule (Thelen *et al.*, 1995a). Moreover, Macisaac *et al.* (1991) suggested that ammonium sulfate reduces the crystallization of glyphosate on the plant surface. By adding citric acid, a stronger conjugate base (negative portion) is provided (Thelen *et al.*, 1995b; Nalewaja *et al.*, 2007), allowing to the acid binds to and removes positively charged cations from solution (Pline *et al.*, 2000).

A mechanism for water conditioning by magnetizing water is facilitating the nucleation and crystallization. Herzog *et al.* (1989) reported that in the presence of an external magnetic field, the nucleation and crystallization of calcium carbonate in hard water can be greatly increased. Afterwards, the crystallization of calcium (Wang *et al.*, 1997; Kobe *et al.*, 2001; Kobe *et al.*, 2002; Plavic *et al.*, 1999; Cho and Choi, 1998; Madsen, 2004; Coey and Cass, 2000), magnesium and sodium carbonates (Bin *et al.*, 2011) in hard water was confirmed when it was passed through a magnetic field. Since dissolved ions in hard water have either negative charge, namely CO_3^-, HCO_3^-, SO_4^-, or Cl^- or positive charge, namely K^+, Na^+, Mg^{2+}, Ca^{2+}, or Fe^{3+}, they are agitated by passing a magnetic field. The positively charged ions are attracted by the N pole and the negatively charged ions were attracted by the S pole of the magnet. Therefore, the collision among the positive and negative ions increases, facilitating the nucleation and crystallization (Cho and Choi, 1998). The latter will allow removing free cations from solution, leaving the herbicidal molecule unrestrained, without forming herbicide-salt complex. On the other hand, previous studies has been discovered that the size in contact angles of magnetized water on the surface of silica gel decreases (Feng and Bo, 2008), thus the surface tension of magnetized water decreases relative to that of distilled water (Rashed-Mohassel *et al.*, 2009), producing smaller droplets which may lead to increased retention and/or absorption of active ingredient (Penner, 2000).

Conclusions

With the exception of potassium phosphate, all water conditioning methods overcame the antagonism that was observed, especially ammonium sulfate. Although the application of magnetized carrier as a new method was not an effective method as much as the addition of ammonium sulfate, from the point of view of environmental and agricultural, applying magnetized carrier will be benefit because it needs no chemical. A systematic research is thus required to improve water conditioning physical method via using paramagnetic substances to facilitate the nucleation and crystallization (Goncharuk *et al.*, 2009) or filtering after magnetization to remove the formed crystals (Fathi *et al.*, 2006).

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