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Field appraisement of olive mills solid waste application in olive crops: Effect on herbicide retention

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ABSTRACT

The aim of this work was to study the effect of soil application of a solid waste from olive oil production (alperujo) on diuron and terbuthylazine retention in the field. Laboratory sorption experiments of the herbicides in unamended soil and soil amended with alperujo (A) or composted alperujo (CA) were performed, and a field study was conducted for 2 consecutive years. Three subplots composed of four olive trees were randomly selected from a 50-ha olive grove (SE Spain) for three different treatments: soil without amendment (*S* plots), soil amended with uncomposted alperujo (*S* + *A* plots) and soil amended with composted alperujo (*S* + CA plots). Uncomposted alperujo was applied to *S* + *A* plots during 4 consecutive years at the rate of 18 Mg ha^{-1} and composted alperujo was applied to *S* + CA plots for the first time on April 2005 at the rate of 8 Mg ha^{-1}. On April 2006, 15 Mg ha^{-1} of *A* was applied to *S* + *A* plots, and 13 Mg ha^{-1} of CA was applied to *S* + CA plots. Manual application of both *A* and CA was followed by soil tillage to a 5 cm depth. A commercial herbicide formulation (28.5% diuron and 28.5% terbuthylazine) was applied to *S*, *S* + *A* and *S* + CA subplots and soils were sampled periodically at 0–10, 10–20 and 20–30 cm depth and extracted for their herbicide content. Results from laboratory studies indicated higher herbicide sorption upon amendment with both *A* and CA when compared to unamended soil. Field studies revealed higher amounts of diuron and terbuthylazine in every plot and almost all sampling dates in the first 10 cm of soil. Soil amendment with alperujo gave rise to higher amounts of herbicide in the soil when compared to unamended soil, and this is specially significant in the case of the herbicide diuron and plots amended with uncomposted alperujo.

1. Introduction

Agricultural practices greatly influence conservation of soil and water resources. Soil amendment with organic residues is a common practise which, beside improving physical and nutritional soil properties, can be considered an "ecological" disposal of these wastes (Crohn, 1996). This agricultural practise is widely used in Spain and other Mediterranean countries, where soils have, in general, low organic matter contents.

In the case of olive oil production, the olive oil mill waste water has been applied to soils as an alternative to its uncontrolled disposal into the environment. Its effect on soil properties has been extensively investigated (Sierra et al., 2001; Paredes et al., 2005). The new technology for olive oil extraction is a continuous centrifuge two-phase process that generates a liquid phase (olive oil) and an organic slurry (olive mill watery husk, known as *alperujo* in Spanish). This system generates approximately 4,000,000 Mg/year usually from November to January, and represents a major environmental problem. Soil amendment with *alperujo* could be beneficial since most of the olive oil producer countries are exposed to desertification. Hence, soil incorporation of these residues with high organic matter content could increase fertility and control erosion (Brunetti et al., 2005; López-Piñeiro et al., 2007). On the contrary as with other organic residues, alperujo is free of heavy metals and pathogens.

There are few studies concerning the effect of direct application (without composting) of *alperujo* as an organic amendment on soil physicochemical properties, although there are some publications which point out that this material increases soil aggregation, restoring in this way the productivity of degraded soils (Piotrowska et al., 2006; López-Piñeiro et al., 2007). López-Piñeiro et al. (2006) also observed in a greenhouse study with winter wheat crop that yield increased up to 198% with soil amendment with *alperujo*. In a recent study with olive crops, these authors found an increase in production up to 40% (unpublished data). However, other studies have reported negative effect of alperujo on soil structural stability, seed germination, plant growth and microbial activity. In fact, several studies have reported the phytotoxic and

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antimicrobial effects of olive mill wastes due to the phenol, organic and fatty acid contents (González et al., 1990; Riffaldi et al., 1993). Composting of *alperujo* has been proposed as a valorisation method for this waste (Roig et al., 2006). Alburquerque et al. (2006) have reported detoxification of *alperujo* with composting due to a decrease in the fat and water soluble phenol content.

Soil amendment with olive oil mill wastes increases soil organic matter affecting soil physicochemical and biological environments and, consequently, affecting immobilization and degradation of herbicides. The increase in soil organic matter can increase sorption potential and reduce pesticide contamination of ground water (Barriuso et al., 1997; Arias-Estévez et al., 2008). However, in many cases, this increase in sorption is the reason for higher herbicide doses required. Consequently, although soil amendment with organic residues can be viewed as beneficial as far as erosion and ground water contamination are concerned, the increase in herbicide use could increase the risk of pesticide contamination. The mobilization and transport of organomineral soil particles with herbicide sorbed is another way of surface water pollution (Pateiro-Moure et al., 2008). Under controlled laboratory conditions, an increase in sorption and a decrease in leaching of simazine were observed in soils amended with alperujo (Albarrán et al., 2003). These authors also found that in soils enriched with alperujo biodegradation of simazine is reduced, which has been attributed in part to microbial preference to use the exogenous organic carbon and nitrogen rather than the herbicide. Atrazine and terbuthylazine mineralization by *Pseudomonas* sp. strain ADP has shown to be much lower in soils amended with alperujo than in unamended soils (Cabrera et al., 2008a). These results suggest that in some cases herbicide persistence can be increased upon organic amendment, affecting the risk of soil and associated water resources contamination. However, additional research is needed to investigate the effect of these residues on herbicide fate in soils under field conditions, since these studies are scarce, due to the high cost and difficulty. Barriuso et al. (1995) found a reduction in leaching and dissipation rate of atrazine in plots amended with different kinds of sewage sludges. On the contrary, Graber et al. (2001) observed a sustained enhanced effect on atrazine and terbuthylazine transport in a field amended with sewage sludge, whereas bromacil was preferentially accumulated in the upper soil layers of the sludge amended soil. These authors attribute the enhanced leaching of atrazine and terbuthylazine to dissolved organic matter–pesticide interactions in solution.

The aim of the present study was to investigate the effect that soil amendment with alperujo has on immobilization of diuron and terbuthylazine in the soil, and for this purpose laboratory and field experiments were performed. A preliminary field study with the herbicide terbuthylazine was performed and revealed, in some cases, higher persistence of the herbicide in soils amended with *alperujo* (Cabrera et al., 2008b). Both herbicides are widely used in olive tree cultures in Spain. Diuron is a substituted urea herbicide used to control a wide variety of annual and perennial weeds and it is relatively persistent in soil with half-lives from 1 month to 1 year (Field et al., 2003). Microbial degradation is considered to be the primary mechanism for its dissipation from soil (Giacomazzi and Cochet, 2004). Terbuthylazine is a chloro-*S*-triazine herbicide used for preemergence and postemergence weed control. The main degradation process is microbial (Guzella et al., 2003). The half-life of terbuthylazine in the soil has been reported to vary between 5 and 116 days depending on the soil characteristics and temperature (Sahid and Teoh, 1994; Dousset et al., 1997). In previous laboratory studies (Cabrera et al., 2007), we have shown that sorption of both herbicides increases and dissipation decreases upon amendment with *alperujo*. However, despite the increase in sorption, leaching can be favoured in some soils due to dissolved organic matter-herbicide interactions (Cox et al., 2007; Cabrera et al., 2007).

2. Materials and methods

2.1. Herbicides and organic residues

High purity standards were used for laboratory studies. Diuron (3,3,4-dichlorophenyl-1,1-dimethylurea) (purity 99%) is a crystal colorless solid, which has solubility in water of 36.4 mg L^{-1} at 25 °C and log K_{ow} (25 °C) of 2.85 ± 0.03 (Tomlin, 2006–07). Terbuthylazine (N^2-tert-butyl-6-chloro-N^4-ethyl-1,3,5-triazine-2,4-diamine) (99% purity) is a colorless powder with a water solubility of 8.5 mg L^{-1} at 20 °C and log K_{ow} of 3.21 (Tomlin, 2006–07). For field studies, the commercial formulation ANIBAL (28.5% diuron and 28.5% terbuthylazine) was purchased from Aragonesas Agro (Madrid, Spain). The main metabolite of diuron, DCPMU (3-(3,4-dichlorophenil-1-methylurea) (99.5% purity) and high purity standards of diuron and terbuthylazine were all purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany).

Two organic residues were used in this study: fresh uncomposted alperujo (*A*) from Mengibar (Jaen, SE Spain) and composted alperujo (CA), which was composted letting the fresh residue (*A*) air drying in a tank. Olive prune remains and leaves were added later to the alperujo to make easier aeration. Physicochemical properties of *A* and CA are given in Table 1. Organic carbon was determined in a Total Organic Carbon analyzer.

2.2. Field condition and soil characteristics

Field experiment was carried out in a 50-ha olive grove located in an experimental farm at Mengibar (Jaen, Southern Spain). The soil is a calcic cambisol (FAO, 1998). The climate of the area is typically Mediterranean. The olive trees (64 years old on 2008) were irrigated under drip irrigation during the drier months (from the middle of April to the middle of October). The total amount of water received during these months was 8544 L per olive tree in 2005 and 8400 L per olive tree in 2006. There was no irrigation during field study in 2007, which ended on April 2007.

For laboratory experiments, soil samples were taken from an area of about 1 ha within the plot. Before herbicide application, the top 10 cm soil layer was sampled at four randomly distributed points. Soil samples were air-dried, sieved through a 2 mm mesh and stored at 4 °C. The soil is a silty-clay soil. Physicochemical properties are: pH 8.32 (1:2 w/v ratio), 1.07% OC content, 42% silt and 52% clay. Soil texture was determined by sedimentation. In the laboratory, this soil (*S*) was amended with *A* and CA at the rate of 10% (w/w) $(S + A)_L$ and $(S + CA)_L$ through manuscript. This rate of alperujo is higher than the realistic amount that growers add (equivalent to 100 Mg ha^{-1}), and falls within normal application rates for wastes such as sewage sludges (Sloan et al., 1998). The

Table 1
Physicochemical properties of alperujo (*A*) and composted alperujo (CA) used in 2005/2006 and 2006/2007 field experiments and organic carbon content (%) of the amended soils in the field.

Amendments	Year	Organic C (%)	C/N
Alperujo	2005/2006	54	49
	2006/2007	53	39
Composted alperujo	2005/2006	39	26
	2006/2007	41	37
Amended soils			
S + *A*	2005/2006	1.64	–
	2006/2007	1.56	–
S + CA	2005/2006	1.38	–
	2006/2007	1.48	–