

FATE OF ALLELOCHEMICALS IN THE SOIL

DESTINO DE ALELOQUÍMICOS NO SOLO

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- REVIEW -

SUMMARY

Allelochemicals are compounds released by one plant or plant residues that may have a negative or positive effect on other plant. The importance of allelopathy was extensively explored during the past three decades, with the work concentrating in the extraction and identification of the chemicals, and demonstration of activity in petry dish experiments. These compounds interact in the soil environment similarly as herbicides and are subject to processes of degradation such as microbial degradation, oxidation, and photolysis, and processes of removal or transfer, such as volatilization and adsorption. The objective of this review was to access the fate of allelochemicals in the soil environment to help to find strategies to increase its activity. The activity of allelochemical is limited in time (because of slow release from the donor material) and in space (because of the interaction with the environment). Demonstration of allelopathy should include the fate of the proposed chemical in the soil environment, presenting studies of degradation and removal processes.

Key words: *adsorption, allelopathy, oxidation, photolysis, plant interference, volatilization.*

RESUMO

Aleloquímicos são compostos liberados por plantas ou seus resíduos e que podem ter efeito negativo ou positivo em outra planta. A importância da alelopatia foi estudada

intensamente nas últimas três décadas, sendo que a maioria dos trabalhos abordou a extração e identificação dos compostos e, demonstração de seus efeitos em experimentos realizados em placas de petri. Estes químicos interagem no ambiente assim como os herbicidas e estão sujeitos aos processos de degradação por decomposição microbiana, fotólise e oxidação e, processos de remoção ou transferência como volatilização e adsorção. O objetivo desta revisão de literatura foi estudar o destino de aleloquímicos no ambiente para auxiliar na definição de estratégias para aumentar sua atividade. A atividade dos aleloquímicos é limitada pelo tempo (devido a liberação lenta do material doador) e pelo espaço (devido à interação com o ambiente). Demonstrações de alelopatia devem incluir o destino no ambiente do composto envolvido, apresentando estudos de processos de degradação e remoção.

Palavras-chave: *adsorção, alelopatia, fotólise, interferência entre plantas, oxidação, volatilização.*

INTRODUCTION

Allelopathy is one of the types of interaction among plants, whereas competition, parasitism, commensalism, symbiosis, and proto-cooperation are the others (GLAUNINGER & HOLZNER, 1982; RADOSEVISH & HOLT, 1984). The term allelopathy

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was proposed by Molisch in 1937 and implied the production of chemicals by living or decaying plant tissue which interfered with a neighboring plant (PUTNAM, 1985).

PUTNAM & TANG (1986) and HARPER (1977) suggested that to provide evidence for allelopathy it would be necessary to adapt Koch's postulates (usually used to demonstrate microorganism effect in a plant disease) to allelopathy studies in the following procedures or experiments: a) describe symptoms; b) isolate, characterize, and assay the chemicals against the plant species previously affected, and identify the chemical; c) obtain similar symptoms when the chemical is added back to the system; d) monitor the release and transfer of the chemical from the donor plant or residues to the recipient plant.

In the last three decades several papers documented the first two steps of Koch's postulates for allelopathy demonstration (DUKE, 1986). However, the importance of allelopathy in the ecosystem, specially in the agro-ecosystem, is controversial, because there are few evidence for the last two steps of Koch's postulates (HARPER, 1977; NUMATA 1982).

Chemicals in the soil solution can be absorbed by plants, but are also subject to processes of degradation such as photolysis, oxidation, and microbial degradation, and to processes of removal or transfer, such as volatilization and adsorption (WEBER & MILLER, 1989; WEBER et al. 1986). The objective of this literature review was to access the fate of allelochemicals in the soil environment to help to find strategies to increase its activity.

IDENTIFICATION AND CHARACTERIZATION OF ALLELOCHEMICALS

During the past three decades many scientists, such as RICE (1974, 1984), PUTNAM & TANG (1986), and WALLER (1989), have documented extensively the effects of allelopathy in reducing plant germination and emergence. RICE (1974) reviewed several examples of allelopathy in nature. *Salvia* (*Salvia leucophylla*) was reported to release inhibitory volatile terpenes thus eliminating grass competition in the California deserts. Sealed-culture-dish bioassays using several local grass seeds and salvia plant parts confirmed the possibility of allelopathy. Several terpenes were identified with chromatography of extracts from salvia parts, and further air analysis of the atmosphere within salvia shrubs demonstrated the presence of these terpenoids (MULLER and coworkers cited by RICE, 1974).

RICE (1984) also reported several studies of the allelopathic effects of black walnut (*Juglans nigra*). Allelopathy was attributed to juglone (5-hydroxy-a-naphthaquinone), and black walnut was reported to injury plants as diverse as alfalfa (*Medicago sativa*), potato (*Solanum tuberosum*), apple (*Malus domestica*), and white pine (*Pinus strobus*) (RICE, 1984).

BARNES *et al.* (1986) isolated the hydroxamic acids DIBOA (2,4-dihydroxy-1,4(2H)-benzoxazin-3-one) and BOA (2(3H)-benzoxazolinone) from fall-planted rye (*Secale cereale*) residues. Either DIBOA or BOA at 49 kg/ha reduced 81 and 39 % barnyardgrass emergence, respectively, suggesting that these compounds are the allelochemicals responsible for the reduction of barnyardgrass in rye fields (BARNES *et al.*, 1986).

Many of the chemicals reported as allelochemicals have important roles in plant protection. Plant pathologists have recognized two types of anti-infectious defense chemicals in plants: those that are present constitutively (fungistatics) and those synthesized after induction by the infection (phytoalexins) (WATERMAN & MOLE, 1994). Fungistatics include catechol found in onion (*Allium cepa*) bulbs (HARBORNE, 1987), whereas phytoalexins includes pisatin found in peas (*Pisum sativum*) (SCHAFFER *et al.*, 1989) and apigeninidin found in sorghum (*Sorghum bicolor*) (SNYDER & NICHOLSON, 1990).

Allelochemicals were reported to increase the severity of plant diseases such as root rots caused by *Fusarium spp* in beans (*Phaseolus vulgaris*) (TOUSSOUN & PATRICK, 1963) and asparagus (*Asparagus officinalis*) (HARTUNG & STEPHENS, 1983) or caused by *Pythium spp* in wheat (*Triticum aestivum*) and sorghum (PATRICK & TOUSSOUN, 1964). The suggested mechanism is that the allelochemicals inhibit the growth of other organisms thus favoring the pathogen development (HARTUNG & STEPHENS, 1983).

Some allelochemicals stimulate germination of parasitic weeds while inhibiting germination of several species (NETZLY *et al.*, 1986). CHANG *et al.* (1986) reported sorghum releases sorgoleones which in the reduced hydroquinone form are germination stimulants of witchweed (*Striga spp*). Later work indicated these compounds are water insoluble and readily oxidized, and do not account for witchweed germination in field conditions (HESS *et al.*, 1992). Recent work has identified witchweed germination stimulants are strigol produced by corn (*Zea mays*), proso millet (*Panicum miliaceum*), and

sorghum (SIAME *et al.*, 1993), and strigol analogs, such as sorgolactone in sorghum (HAUCK *et al.*, 1992) and alectrol in cowpea (*Vigna unguiculata*) (MULLER *et al.*, 1992). Strigol increased witchweed germination by inducing ethylene synthesis in the seeds (BABIKER *et al.*, 1993).

Many allelochemicals are believed to have evolved for herbivory protection in plants. Tannins are general feeding deterrents for most mammalian herbivores (MOLE & WATERMAN, 1987), and rotenone-like compounds are effective feeding deterrents for root feeding beetles (LANE *et al.*, 1985). Besides feeding deterrents some phenolics (p-coumaric and ferulic acids) also inhibit mammalian reproduction (HARBORNE, 1988).

EVIDENCE AGAINST ALLELOPATHY

Some scientists, such as HARPER (1977), GLAUNINGER & HOLZNER (1982), and SCHMIDT (1988) strongly advocates against the importance of allelopathy in agroecosystems. To prove the efficacy of allelopathy is almost impossible (HARPER, 1977; NUMATA, 1982; PATTERSON, 1986; WILLIAMSON, 1990), because the adaptation of Koch's postulates from microbiology to allelopathic studies (PUTNAM & TANG, 1986) is inappropriate due to: (i) chemicals do not reproduce, therefore the compounds would require constant application simulating natural release conditions; (ii) chemicals degrade; (iii) among microbes there is a one-to-one correspondence between disease and organism, while on allelopathy, complex interaction between chemicals may exist (WILLIAMSON, 1990).

The 60m grass-free zone around salvia shrubs in the California desert, initially supposed to occur due to allelopathy, was later suggested to be created by grazing and trampling activities of animals. Several experiments involving feeding stations, fenced shrubs, and shelter placement demonstrated rabbits could account for grass seedling reduction around salvia shrubs (BARTHOLOMEW cited by HARPER, 1977).

PATTERSON (1986), WILLIAMSON (1990), and VIDAL (1995) recognized allelopathic effects in petry dish studies, but noted differential results between petry dish and soil experiments. Possible causes of these differences included rate limitations, and degradation and removal processes, which also affects other chemicals in the environment (DALTON, 1989; GUENZI & MCCALLA, 1966a; PATTERSON, 1986; WALLER, 1989).

RATE LIMITATIONS OF ALLELOCHEMICALS

The role of DIBOA and BOA from rye residues in barnyardgrass (*Echinochloa crusgalli*) control seem weakened when considering that the rate of these compounds combined, if released all at once from a 5t/ha rye biomass, would be only around 10 to 16kg/ha, and it would be necessary 49kg/ha of DIBOA or 100kg/ha of BOA to obtain reasonable (> 68 %) barnyardgrass control (BARNES *et al.*, 1986). Besides that, 50% of BOA and DIBOA disappeared from rye residues 10 days after desiccation (YENISH *et al.*, 1995). MILLER & JORDAN (1994) reported the reduction of soil water by fall-planted rye may have reduced the growth and development of weeds (and soybeans (*Glycine max*)) in the soybean crop.

Likewise, the concentrations of phenolic acids from wheat residues were relatively low in the soil, compared with those required to inhibit germination and growth of plants (GUENZI & MCCALLA 1966b). For instance, the concentration of ferulic acid was 120mg/kg in wheat dry mass (GUENZI & MCCALLA 1966a), therefore 12t/ha of wheat residues in a no-tilled field would contribute 1.44kg/ha of ferulic acid if all the ferulic acid was immediately released to the soil. Assuming that after release the ferulic acid would accumulated in the upper 100µm (0.1mm) of the soil, the concentration of this phenolic acid would be 1100mg/kg soil (assuming soil density of 1.3g/cm³). This concentration would dilute linearly to 5mg/kg soil if ferulic acid diffused uniformly to 2cm depth in the soil. Petry dish experiments indicated ferulic acid at 900mg/ha soil inhibit giant foxtail germination by 50% (VIDAL, 1995). In the surroundings of the decomposing wheat residues in no-tilled fields the concentration of ferulic acid could be high enough to inhibit foxtail (*Setaria faberi*) germination by 75% only if all of the ferulic acid in the residue were immediately released and degradation and removal processes were prevented (VIDAL, 1995).

ALLELOCHEMICAL DEGRADATION PROCESSES

Allelochemicals can be decomposed by physical, chemical and microbial processes. HARTLEY & JONES (1975) and KATASE (1981) documented that ultraviolet light may inactivate several phenolics, among them coumaric and ferulic acids. An eloquent demonstration of chemical decomposition of allelochemicals is the oxidation of sorgoleones from the active hydroquinone to the inactive quinone form

(HESS *et al.*, 1992). These compounds were discarded as important witchweed germination stimulants because they are readily oxidized, reducing the concentration of the hydroquinone form in the soil solution (HESS *et al.*, 1992).

Fungi, such as *Poria*, *Gloeophyllum* (in aerobic conditions), *Chaetomium*, and *Preussia* (anaerobic conditions), and bacteria, like *Pseudomonas*, are able to degrade natural aromatic compounds (PAUL & CLARK, 1989). Natural aromatic compounds lack halogens thus are readily decomposed by soil microorganisms (BOLLAG & LIU, 1990). For example, HAIDER & MARTIN (1975) observed 55, 70 and 95% of $^{14}\text{CO}_2$ was released in less than one week after soil application of the allelochemicals ^{14}C -caffeic, ^{14}C -benzoic, and ^{14}C -p-hydroxybenzoic acids, respectively. Similarly, HAIDER & MARTIN (1967) reported *Epicoccum nigrum* decomposed several phenolics in the soil, whereas SCHMIDT (1988) verified *Pseudomonas spp* rapidly mineralized compounds, such as chlorogenic acid, ferulic acid, and gallic acid, among 15 other aromatics.

SCHMIDT (1988) found that *Pseudomonas spp* can grow rapidly using juglone as its sole source of carbon and energy. Around 90 % of juglone was mineralized in less than 7h after incubation. The high affinity of this bacterium with juglone and other phenolics led to rapid degradation of these compounds. SCHMIDT (1988) concluded that it is unlikely that allelochemicals like juglone have any role in natural conditions because the ubiquity of *Pseudomonas* in the soil. SCHMIDT (1988) speculated that anaerobiosis caused the phytotoxic effects earlier attributed to juglone.

ALLELOCHEMICAL REMOVAL PROCESSES

Among the main processes of removal of organic compounds from the soil environment are volatilization and adsorption. The probability that considerable amounts of volatile terpenoids from salvia accumulate in the soil is very low. Volatile pesticides can be removed from the soil or plant surfaces by volatilization at a rate of 80 to 90 %, in contrast to only 5 to 10 % loss by leaching and runoff (TAYLOR & SPENCER, 1990). Once the chemical crosses a layer of laminar flow near the surface, it is rapidly dispersed in the atmosphere through turbulent movement processes (TAYLOR & SPENCER, 1990).

Adsorption is the most important soil factor controlling the fate of chemicals in the environment because it controls the chemical concentration present

in the soil solution (see review by SHEA, 1989). The Freundlich equation is the most common mathematical description of adsorption:

$$x/m = K_f C^{1/n}$$

where x = amount adsorbed, m = amount of adsorbent, C = chemical concentration at equilibrium, K and n are empirical constants reflecting the degree of binding and the degree of linearity between the amount adsorbed and the concentration at equilibrium.

If $1/n > 1$, the adsorption isotherm has a concave shape (S-type isotherm). The S-type isotherm indicates that the chemicals have a relatively low adsorption initially, and adsorption increases as the number of molecules on the surface increases (also called cooperative sorption mechanism) (WEBER & MILLER, 1989). Three conditions are required for this kind of sorption to occur: the molecule has one point attachment, the molecule has moderate intermolecular attraction (orient vertically when sorbed), suffers strong competition for adsorption sites from water and other molecules (WEBER & MILLER, 1989). Herbicides that follow an S-type adsorption to montmorillonite are acetochlor, alachlor, and metolachlor (WEBER & MILLER, 1989; WEBER *et al.*, 1986).

If $1/n < 1$, the adsorption isotherm has a convex shape (L-type isotherm). L-type isotherms occur when specific bonding are involved, or sorbed molecules are not vertically oriented, or water molecules are not strong competitors for adsorption sites, or when a mixture of several adsorbents (organic matter plus clay for instance) is present (WEBER & MILLER, 1989). Herbicides that follow an L-type adsorption to soils are alachlor, atrazine, bromacil, diquat, diuron, fluridone, glyphosate, paraquat, and prometryn (WEBER & MILLER, 1989; WEBER *et al.*, 1986).

If $1/n = 1$, K_f becomes a partition coefficient (K_D), and the linear equation is called a C-type isotherm (for constant partition) (SHEA, 1989; WEBER & MILLER, 1989). Higher K_f values indicate a higher affinity between the phenolic and the adsorbents (SHEA, 1989; WEBER & MILLER, 1989).

Several researchers have observed that binding of allelochemicals to soil would reduce their performance in nature (DALTON, 1989; DALTON *et al.*, 1989; GUENZI & MCCALLA, 1966a; HORRIE *et al.*, 1989). DALTON *et al.* (1989) reported 20 to 60% adsorption of ferulic acid one day after incubation

in several sterilized soils. Likewise, GUENZI & MCCALLA (1966b) observed the phenolic acids from wheat residues were highly adsorbed to soils, whereas WANG *et al.* (1971) documented 60 to 98% adsorption of syringic or ferulic acids only three hours after incubation in soil. VIDAL (1995) reported 20 to 38% of the phenolics ferulic and salicylic acids, and catechol were adsorbed to soil. The adsorption of salicylic acid followed an L-type isotherm, whereas the adsorption of catechol and ferulic acid followed an S-type isotherm (VIDAL, 1995).

Analysis of the effects of soil components on adsorption indicated highest adsorption in soils with highest organic matter (OM) levels (DALTON, 1989; DALTON *et al.*, 1989; HORRIE *et al.* 1989; WANG *et al.*, 1971). Allelochemicals from broom (*Cytisus scoparius*) accumulated in soils with low OM and inhibited lettuce seedling emergence. However, the allelochemicals did not accumulate and did not affect seedling emergence in high OM soils (HORRIE *et al.*, 1989). Sorgoleones also had limited activity because they are lipophilic and readily bound to OM in the soil (CHANG *et al.*, 1986; HESS *et al.*, 1992). Clay type may also affect adsorption. DALTON *et al.* (1989) detected 20 % adsorption of ferulic acid on kaolin or gibbsite, 70 % adsorption on goethite, and 100 % adsorption on histosol. They explained the increased adsorption in these two last soil components was in part due to increased surface area, and in part due to the presence of Fe and Al oxyhydroxides (DALTON *et al.*, 1989).

FINAL CONSIDERATIONS

In the soil environment there are many supposed allelochemicals. KIMBER (1973) indicated that in nature, the concentrations range from inhibitory for some allelochemicals to stimulatory for other allelochemicals, and the resultant net effect in plants may be lower inhibition or stimulation or no effect at all. Some authors have argued that allelochemicals act synergistically, thus magnifying their phytotoxic capabilities (EINHELLIG & RASMUSSEN, 1978). Few experiments were conducted to test this hypothesis. However, herbicide science indicates that synergism is a rare occurrence and usually antagonistic (GREEN, 1989; ZHANG *et al.*, 1995) or additive (GREEN, 1989) effects are the norm. As expected, DUKE *et al.* (1984) reported antagonism between p-coumaric and ferulic acids on lettuce seed germination, and BLUM *et al.* (1984) observed antagonism between

ferulic, caffeic, and vanillic acids on cucumber radicle growth.

It is possible to speculate that allelochemicals may have importance in plant interference mainly when there is contact between the source of the chemical and the target plant or tissue, avoiding the interactions between the chemicals and the soil environment. Glyphosate, a herbicide that strongly binds to soil, can move between one plant to another when their roots are in contact (RODRIGUES *et al.*, 1982). The stimuli of witchweed germination by strigol and strigol analogs is limited to a maximum 2 mm radius around the root of the donor plant and witchweed seeds (HAUCK *et al.*, 1992; MULLER *et al.*, 1992; SIAME *et al.*, 1993; BABIKER *et al.*, 1993).

CONCLUSION

The literature review presented above suggests that, in soils, allelochemicals have activity limited in time and space. Time limitation occurs because the allelochemicals are not released at once from the decaying straw residues, and because degradation and removal processes reduce the concentration available in the soil solution. Space limitations occur because, even if all the allelochemicals in the crop residues were released at once, and microbial degradation and adsorption were precluded, the performance of these natural occurring chemicals would be spatially limited to seeds placed near the donor plant or the decomposing residues, because of reduced concentration.

This review also indicates that effective demonstration of allelopathy should present the fate of the proposed chemical(s) in the soil environment, including studies of degradation and removal processes.

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