

# Photostability of methidathion in wet soil amended with biosolid and a surfactant under solar irradiation

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## Abstract

Photodegradation studies of the organophosphorous insecticide methidathion in thin layers of wet soil samples have been carried out under solar irradiation. Soil samples consisted of an agricultural soil added with two amendments: a municipal biosolid and the cationic surfactant TDTMA (tetradecyl trimethyl ammonium bromide). Dark controls of the different soil treatments were also considered. Soil and biosolid samples were previously autoclaved to eliminate biotic degradation. In this study we investigated the role of these amendments in methidathion photodegradation which is a rapid (<7 days) and indirect process. Although scarce differences were found between non-amended and amended samples, methidathion from soil exposed under sunlight is degraded more quickly than in dark conditions. Photodegradation products (methidathion oxon GS 13007 and GS 12956) were not detected.

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## 1. Introduction

Methidathion is a non-systemic organophosphorus insecticide and acaricide, used since 1966 to control sucking and chewing insects and spiders on many crops, which has led to detect its presence in surface and groundwater of South-Eastern Spain (Hernández et al., 1996; Garrido Frenich et al., 2001). The use of this and other pesticides in plant protection may produce soil and water contamination, which can be attenuated by abiotic transformations. Due to this, the photochemistry of various pesticides has been widely investigated in

liquid solutions (Dejonckheere and Kips, 1974; Doong and Chang, 1997; Caselli et al., 2001; Vulliet et al., 2002) though information on the photolysis of pesticides in soil under environmental conditions is relatively scarce (Romero et al., 1998; Sukul and Spiteller, 2001; Bavcon et al., 2003). This process, dominant in the first millimetres of the soil layer because of the low penetration of UV radiation, takes place just after pesticide application and can also be caused by agricultural practices or by capillary phenomena (Gong et al., 2001). In arid and semiarid regions, such as the Mediterranean zone, sunlight photolysis together with evaporation of pesticide can play an important role in the decay of pesticides in the environment.

The photodegradation processes can be direct and/or indirect, the former taking place through sunlight absorption directly by the pesticide and the latter, indirectly,

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mediated by intermediate radicals or other species, such as oxides and components of the dissolved organic matter (Herber and Miller, 1990; Torrents et al., 1997).

Recent strategies for the prevention of soil and water contamination by pesticides have focused on the use of exogenous organic amendments, such as peat, manure, biosolids (Celis et al., 1998; Romero et al., 1998, 2001) or surfactants, because they can increase pesticide sorption and retard their migration (Abu-Zreig et al., 2000; Sánchez-Camazano et al., 2000; Sánchez et al., 2003a,b). Surfactants have shown their ability to modify the relationship between contaminants in the sorbed and liquid phases and may interfere in contaminants photodissipation (Chu and Jafvert, 1994; Huang and Hong, 2000). Besides, some components of exogenous OM, such as humic substances, apart from enhancing pesticides sorption, may affect their photodissipation, because they absorb sunlight and can react with water to form hydroxyl radicals on soil surfaces (Zepp, 1988; Konstantinou et al., 2001). Nevertheless, the effect of dissolved organic matter is not clear since it has been reported to enhance (Sakkas et al., 2001) or inhibit (Dimou et al., 2004) pesticide photodegradation.

Photolytic studies can be undertaken under natural sunlight or with artificial solar sources which simulate the natural sunlight spectral distribution. Although laboratory studies are interesting, they are not a good approach to irradiation in the field, due to the natural variations of environmental conditions (Höllrigl-Rosta et al., 1999).

It has been previously observed (Sánchez et al., 2003a) that the addition of biosolid and tetradecyl trimethyl ammonium bromide (TDTMA) increases methidathion sorption onto soil but their effect on the photolysis of this insecticide is still unknown. The aim of this work is to establish the photostability of methidathion in soil in the presence of TDTMA and a municipal biosolid. The study was carried out outdoors, under natural solar irradiation, on sterilized wet soil thin layers in the presence or not of the surfactant and/or the biosolid. The formation of the photoproducts GS 13007 and GS 12956, which can be considered the final degradation product, was also investigated.

## 2. Material and methods

### 2.1. Chemicals

Methidathion (*S*-[(5-methoxy-2-oxo-1,3,4-thiadiazol-3(2H)-yl)methyl] *O,O*-dimethyl phosphorodithioate) and the photoproducts GS 13007 (thiophosphoric acid *S*-(5-methoxy-2-oxo-[1,3,4] thiadiazol-3-ylmethyl)ester *O,O'*-dimethylester) and GS 12956 (5-methoxy-3H-[1,3,4] thiadiazol-2-one), all 99% purity (Novartis, Münchenwilen, Switzerland), as well as the internal stan-

dard bromophos-methyl, *O*-(4-bromo-2,5-dichlorophenyl) *O,O*-dimethyl phosphorothioate (99.9%, Labor Dr. Ehrenstorfer, Germany) (Fig. 1) were used.

The cationic surfactant tetradecyl trimethyl ammonium bromide (TDTMA), 99% purity (Aldrich Chem., Madrid, Spain), with a critical micellar concentration (cmc) of  $0.1 \text{ g l}^{-1}$ , and a municipal biosolid were selected as amendments. This dewatered biosolid, from a municipal wastewater facility in Granada, has 40% of OM (1.6% humic acids, 0.47% fulvic acids), pH of 7.2 and its elemental composition complies with the 86/278/EEC Directive, relative to the use of sewage sludge in agriculture.

### 2.2. Soils

The soil used in the experiments corresponds to a cultivated area of Granada (SE Spain) and has been characterized as silt loam soil (typic xerofluvent). Soil samples from the upper layer were air dried and sieved ( $<0.63 \text{ mm}$ ). The main surface soil properties analyzed according to official methods (MAPA, 1986) were: 31% sand; 58% silt; 11% clay; 34%  $\text{CaCO}_3$ , pH of 8.5; 0.92% organic C and 27% water holding capacity (WHC) (1/3 bar).

### 2.3. Photolysis experiment

To investigate the effect that the biosolid and the surfactant had on the photolysis process, different soil samples were prepared: unamended soil samples (S), and samples amended with biosolid (SB), surfactant (SS), or both amendments (SBS). In all cases, soil samples (10 g), amended or not with the biosolid at 4% (w/w), placed on Petri dishes (9.7 cm i.d.), were added with 10 ml of water and air dried for 48 h, in order to get an uniform soil layer of  $0.89 \pm 0.09 \text{ mm}$  thickness. Three milliliter of  $33 \text{ mg l}^{-1}$  solution of methidathion prepared either in water (S and SB) or in a surfactant solution at 10 cmc (SS and SBS) was dripped uniformly over these dry soil layers. To suppress the microbial activity biosolid and soil material were autoclaved previously, by heating under pressure for 1 h each day, for three consecutive days.

All Petri dishes were covered with a glass disk, sealed with silicon to minimize losses outdoors and exposed upside down to guarantee that the samples surface was equally exposed to natural irradiation, avoiding the shading produced by the borders or the silicon. For comparison purposes, another series of Petri dishes for each treatment were covered with aluminum foil and used as dark controls. At different time intervals, two Petri dishes of each type of soil (S, SS, SB and SBS) (0, 1, 3, 7, 10, 14 and 21 days) and their corresponding dark controls (0, 3, 10, 14 and 21 days) were analyzed. The insecticide concentration and the soil water content

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