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Study of photocatalytic Degradation of Mancozeb and chlorpyrifos methyl under intense Ultraviolet Light over different time period

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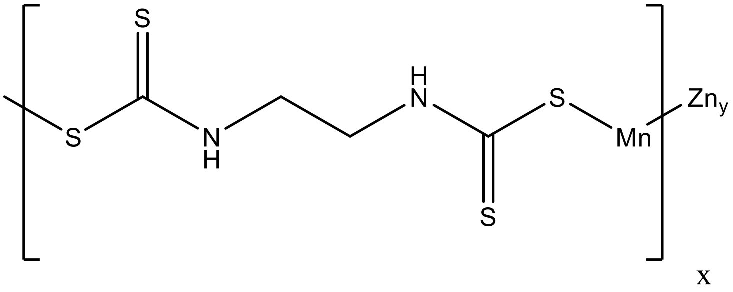
Introduction

Mancozeb, an ethylene bis-dithiocarbamate (EBDC) fungicide, has been used for the management of fungal pathogens all over the world in major agricultural crops for over five decades. Its popularity is mainly associated to its multi-site action mechanism, its low cost and its capacity to prevent resistance of pathogenic fungi to its use [1]. Despite the continued use of this class, the increased concern with the environmental persistence, degradation products and toxicity of this class has stimulated a great deal of scientific and regulatory interest. All this understanding is important in realizing the agronomic value balanced to the possible ecological and human health risks. The chemical degradation patterns of mancozeb are of special interest because of the production of ethylenethiourea (ETU), a toxic metabolite which is well known for its carcinogenic and teratogenic properties [2], [9]. Oxidation reaction Studies investigating oxidation reactions, such as oxidation in the presence of ozone and chlorine dioxide, have found several oxidation degradation products - which raise additional concerns in toxicology [2]. Environmental variables also influence the stability of mancozeb; for example, formulation type and adjuvants have a significant effect on rainfastness on the crop foliage and influence residue levels and resulting environmental contamination [3]. Soil exposure studies also provide strong evidence of a shift in the mineralization of carbon and nitrogen indicating disturbances of their important microbial processes [6]. Analytical detection of mancozeb residues has improved a lot and nowadays the efforts are based on developing sensitive and portable technologies. Novel approaches are e.g. sensors based on sulfur quantum dots in electrochemical schemes [4], fluorescent test papers based on europium complexes combined with smartphone-based read-out [7] and nanozyme-based sensing platforms [13]. These kinds of developments are not only important to monitor environmental and food residues, but also accordingly meaningful to assess the exposure risks in real time. Colorimetric and fluorimetric nanosensors also add to the power of on-site rapid testing [10], [14]. From a toxicological point of view, mancozeb has different hazards to different biological systems. Aquatic studies have been able to render information on the developmental toxicity of zebrafish embryos at even an environmentally relevant concentration [8]. In mammal, oxidative stress produced reactical oxygen species, and apoptosis have been implicated in chronic exposure responses regarding liver injury, oxidative stress and apoptosis throught mechanism [11], [16], [20]. Reproductive toxicity is well documented too, as experimental models indicate a long-lasting adverse effect in the cells of the outer cell layer forming the maturating follicles (granulosa cells), in the morphology of the follicles and in female fertility parameters [15], [17]. Long-term carcinogenicity studies done in rodents confirm the concern over ETU-related tumorigenesis [9]. In addition, mancozeb also impacts non-target terrestrial organisms. Earthworm avoidance and survival tests suggest that there is adverse soil ecotoxicity, which suggestions that the soil ecosystem health is disrupted [19]. Plants subjected to mancozeb make changes to its metabolism that might have a response regarding the nutritional quality and stress response, recorded in lettuce leaves [12]. These results all together reveal multi-systemic and multi-species effects of mancozeb at different ecological compartments. Given the increased pressure from regulators and fear for our public health it is increasingly interesting to find out about safe, alternative fungicides.

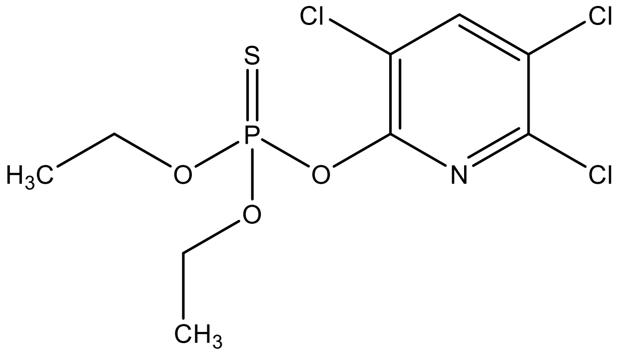
The recent studies are proposing effective replacement strategies by the disease control of late blight in potatoes with an emphasis on integrated disease controlled strategy and reduced dependency on EBDCs [18].



Mancozeb

Chlorpyrifos Methyl

Chlorpyrifos methyl has wide application due to its high efficiency against a range of insect pests, however, owing to the increasing evidence from the scientific community, significant concerns have been raised with regard to its persistence in the environment, its ecotoxicity and human health risks. Most of the activity of CPF is through inhibition of acetylcholinesterase which result in an accumulation of acetylcholine at synaptic joints leading to neurotoxicity of target and non-target organisms [22]. During the past 20 years, CPF contamination has become a major problem worldwide that requires more regulation and remediations [21]. The toxicological effects of CPF have been represented for the most part in mammalian and non-mammalian systems. In vitro studies have demonstrated that the pesticide induces both genotoxic and cytotoxic activity on human lymphocytes even at low concentrations [23]. Subacute exposure experiments further show oxidative stress, dysfunctions of the liver and kidneys and potential protective actions of natural antioxidants such as green tea extracts [24]. Similar toxicopathological manifestations have been reported, however, in poultry including lymphoid organ damage and physicochemical lesions throughout the body [29], [38]. Environmental impacts are also alarming: residues of CPF in food crops (okra, tomato) persist for a long time and have direct exposure risks to the consumer and farmer [30], [36]. The ecological consequences of contamination in CPF are not restricted to just land and aquatic ecosystem. Studies suggest that there are large changes in soil microbial diversity and enzyme activities with CPF application, which influence nutrient cycling and soil fertility [34], [35]. CPF residues in soil also have a negative effect on plant growth and productivity, demonstrated in the cultivation system of onion [28]. In aquatic environments, Hematological perturbations and oxidative stress, decreased antioxidant response to oxidative stress, and histopathological changes have been found in fishes under CPF exposure, thus representing major potential consequences to destabilise the aquatic food chain [40]. Another concern that is seen to be increasing greatly is the development of CPF resistance of different populations of the pests. Resistance mechanisms, e.g. modification of acetylcholinesterase genotypes have been verified in species such as Bemisia tabaci [37]. Similar resistance patterns have been found in ticks (Rhipicephalus microplus) [27], as well as in predatory mites (Kampimodromus aberrans, Typhlodromus pyri) [34,35] raising questions in the sustainability of CPF in pest management strategies over time [33,39].



Chlorpyrifos

Photocatalytic Degradation

Photodegradation is degradation, or a change in chemical structure resulting from an interaction of the energy of photons or light on the molecule or materials, normally visible light or internal and ultraviolet (UV) light. When a substance absorbs energy in the form of light, it may be electronically excited and this starts different chemical reactions that may alter the structure of the substance or may break the substance down [41], [43]. This process is important in many fields including polymer study, study of medicines, study of green chemistry, work about the stability of natural products. In polymers, photodegradation generally occurs via bond scission, oxidation, cross-linking or chain cleavage which involve excited-state reactions and formation of reactive species, such as a radical or singlet oxygen [42], [44], [48]. How things break down depends on things such as the type of polymer, the presence of impurities and the environment. When light strikes natural polymers such as cellulose and lignin they break it down. This could cause the color to change or strength of the material to go down [46]. Photodegradation can modify the extent to which a drug is strong, safe and long-lasting. When drug molecules are exposed to light, for example, it can lead to oxidation, isomerization and cleavage among other things. Testing for photostability is an important part of making new drugs. In order to be able to test the photochemical reactions correctly, the process setting and the treatment for kinetics have to be standardized. This includes the use of controlled sources of irradiation, appropriate wavelengths and also measurement of the degradation products [45], [49]. Photodegradation can occur directly, when a molecule absorbs light or indirectly, when another molecule absorbs light and in turn passes energy on to begin the degradation process. Photo-organocatalysis focuses on catalytic ways of using light to cause chemical reactions, which change the way things are made. This shows how useful the ideas of photodegradation can be [47]. It's also very important to know how molecules behave when in different circumstances. The photodecomposition of riboflavin has unique pathways for the higher pH levels: gas-phase photolysis investigations increase the understanding of the molecular behaviour in solution [50].

Photo degradation of Mancozeb

Photodegradation is important for figuring out what happens to mancozeb in the environment since this wants to-obtain how long it endure and how dangerous by-products form. Upon exposure to sunlight, mancozeb is degraded very quickly although it is mainly hydrolyzed and photoably degraded. Extensive kinetic modeling has demonstrated that photolysis is important in the mechanisms of conversion of mancozeb to a more stable and toxic metabolite, ethylenethiourea (ETU). The chemistry of the water as well as the brightness of the light has a big effect on the frequency of this. Studies of the photodegradation of sprays of broad-spectrum fungicides indicate that indirect processes mediated by reactive oxygen species, such as hydroxyl radicals and singlet oxygen, are able to promote higher rates of degradation in natural aquatic environments [57]. Similar results in pyrrole fungicides underlines the importance of indirect photoreactions in prediction of the formation of photoproducts in the environment [58]. Recent advances in the field of photocatalysis have shown that nanomaterials are able to increase the rate of decomposition of mancozeb under the action of sunlight. The exposure of natural sunlight for ZnO nanoparticles doped with nanoceria strongly improves the decomposition of pollutants and slows down the growth of ETU. Such an approach could be a means of reduction of pollution in the environment [53]. Nanoformulated mancozeb has improved degradation dynamics as a result of the stronger light absorption and reaction dynamics on surfaces [54]. Photodegradation, particularly the indirect and photocatalytic, is having a huge effect on the way mancozeb is modified in the environment, and the risks.

Photo degradation of Chlorpyrifos

Photodegradation is a major abiotic process that help get rid of chlorpyrifos (CPF) in the environment, especially in water systems that get a lot of sunlight. CPF absorbs UV energy at a moderate rate which makes the direct photolysis to occur slowly. However, this reaction takes place much faster in the presence of photocatalysts, reactive oxygen species (ROS) or photosensitized conditions [61], [65]. Advanced oxidation technique (AOPs) in particular photocatalysis in titanium dioxide (TiO2) was proven to be very effective in breaking down CPF. When TiO2 (titanium dioxide) nanoparticles are exposed to the ultraviolet or visible wavelengths of light they produce reactive oxygen species (ROS) such as hydroxyl radicals (OH) and superoxide radicals (O2-). These ROS then attack phosphate ester linkages, as well as pyridinol ring of chlorpyrifos (CPF) causing it to slow oxidize and break apart [62], [63]. Studies show that TiO2 has the capacity to break down CPF in a short time with exposure to UV or visible light. The rate of breakdown is dependent on the amount of catalyst, the pH and intensity of the light [62, 64]. Budarz at al. demonstrated that competitive processes between different types of organic matter have a large influence on degradation of CPF in actual multi-component systems. These interactions modify the availability of ROS and selectivity of routes [63]. 3,5,6-trichloro-2-pyridinol (TCP) is mostly the product of photodegradation of CPF. Thereafter, it experiences more photochemical and photocatalytic changes. TCP has greater longevity and water solubility than CPF. However, processes based on TiO2 degrade TCP into smaller and less dangerous byproducts, such as chlorinated acids and CO2 [67]. It has been shown that the UV-C irradiation accelerates the degradation of CPF and at the same time makes it more toxic in the short term with the formation of ROS-induced intermediates [66]. Solar photo Fenton and heterogeneous photocatalysis are two examples of process, which count on natural sunlight in to disrupt down CPF. Such processes are useful when it is needed to treat large quantities of water. Photodegradation through direct and catalyst mediated processes is mandatory for degradation of CPF and is a significant step in the rehabilitation methods of CPF in damaged settings [61], [68].

2. Materials and Methods

2.1 Material

Mancozeb and chlorpyrifos technical products were procured from United Phosphorus Ltd. (UPL) is a yellow to greyish powder fungicide which contains about 85% active mancozeb, manganese about 20% and zinc about 2-2.5% of active, moisture less than about 2%, pH 6-9 (1% suspension).

2.2 Experimental Method

Pure Technical grade Mancozeb sample weighted 100 mg and dissolved in 100ml distilled waterand exposed to ultraviolet chamber with continuously stirring with magnetic stirrer and sample are collected for different time period. Degradation of Mancozeb produce Ethylene thiourea (ETU). ETU by uv spectroscopy determined and degradation calculated.

2.3 Analysis by UV spectrometer

Prepared standard and sample solution in distilled water and scanned with UV spectrophotometer to find lmax and select 279 nm with 1 nm band pass and blanked instrument with solvent and equilibrated for 15 min, Then measured absorbance of standards (100 PPM) to get calibration curve and after sample of different hours with time lapse were collected and determined. Photo degradation carried under 254 nm ultra violet light to fall within linear range, measured absorbance, applied blank correction and calculated concentrations from calibration.

3.0 Characterization

3.1 X- ray Diffraction Analysis

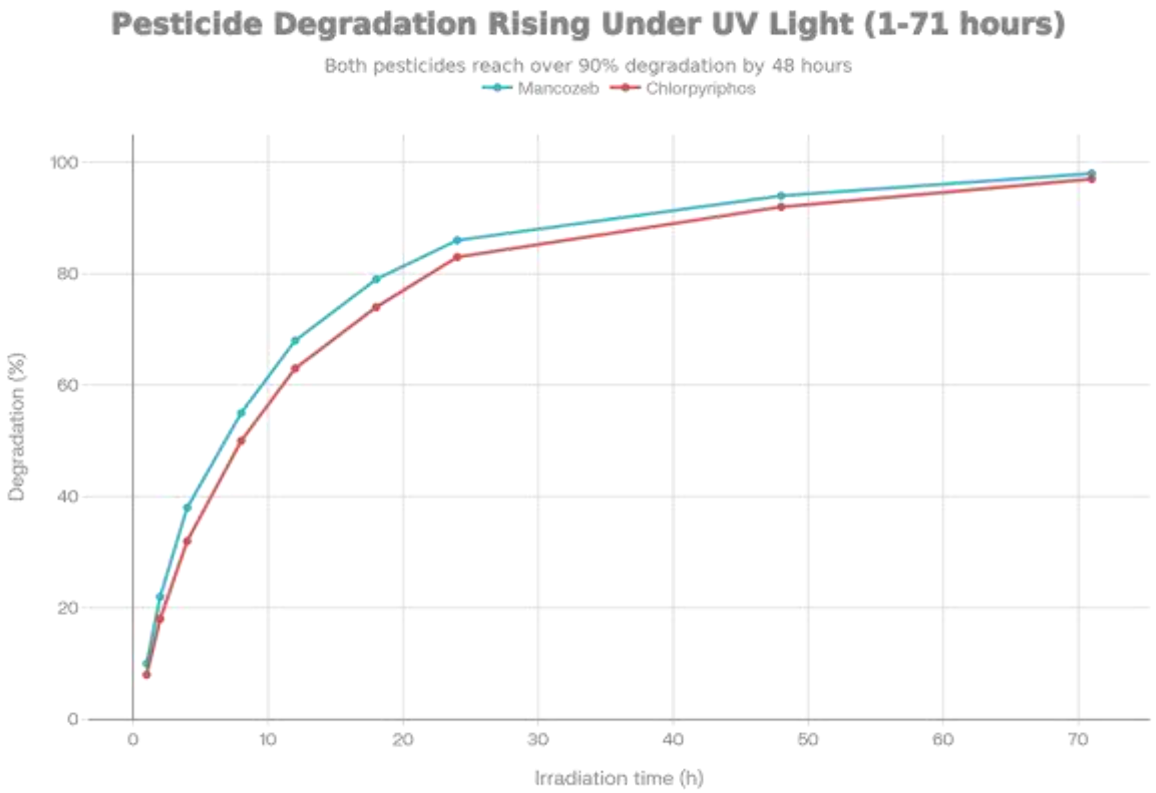
Structural analysis of the composite was done by Rigaku D/Max Miniflex -600 X-ray diffractometer (30 kV, 2 mA) and an SC-70 detector. Powder samples were analysed to check the composition of the phases.

3.2 UV Spectrophotometer

Characterization of mancozeb solution was done using Shimadzu UV 1800 UV-VIS spectrophotometer by recording the absorption spectrum in the appropriate wavelength range, determining the wavelength of maximum absorption (l max) and drawing a calibration curve of absorbance versus concentration 4 Result and discussion The degradation of mancozeb and chlorpyriphos by photocatalytic mechanism in the presence of UV irradiation was monitored for the period of 71 h at various time intervals (1, 2, 4, 8, 12, 18, 24, 48 and 71 h). Both pesticides showed a gradual increase in degradation efficiency with time of exposure, showing that the reaction was time dependent and depended on continuous UV irradiation. Overall, mancozeb degraded slightly faster than chlorpyriphos at all times, but had an effect that was more pronounced in the latter stages of the experiment.

4.0 Result and Discussion

At the initial stage (1-4 h), the degradation of both pesticides was relatively moderate with mancozeb reaching about 10-38% and chlorpyriphos about 8-32% removal, respectively, which demonstrates that the catalyst became active soon after exposure to UV. Between 4 and 12 h, degradation rate was increased more sharply and removal of mancozeb was increased up to about 68% while chlorpyriphos removal was about 63%, suggesting efficient generation of reactive species in measured period. In the period of intermediate to long irradiation (18-24 h), the degradation was still increasing, with mancozeb reaching around 79-86% and chlorpyriphos reaching about 74-83%, revealing that the majority of easily oxidized fractions were already attacked at this time.



Prolongation of the UV exposure to 48 and 71 h led to near complete degradation, which reached near total degradation for mancozeb (94-98%) and chlorpyriphos (92-97%), indicating that prolonged irradiation is effective for near complete removal of both pesticides. Throughout the experiment, mancozeb showed consistently slightly higher degradation percentages than chlorpyriphos at the same irradiation times suggesting that mancozeb was more susceptible to being attacked by photocatalytic processes under the conditions presented. The small difference between the two curves at longer times also suggested that although their degradation kinetics are slightly different, both the compounds can be efficiently removed from the aqueous solution under prolonged UV-assisted photocatalysis.

5.0 Conclusion

Experimental study carried out clearly showed that the ultraviolet photocatalytic degradation was a highly effective treatment for mancozeb removal from aqueous systems and almost complete degradation occurred at longer irradiation times. In environmental terms, that meant good possibilities for the application of UV-based photocatalytic treatment for pesticide-contaminated agricultural runoff and wastewater, where conventional treatment methods are often unable to fully remove such persistent chemicals. By destroying the mancozeb and not transferring it to the other phase (e.g. from water to sludge), the photocatalysis reduces the pesticide load reaching the natural surface and groundwater bodies directly. This reduction was extremely important as mancozeb was linked with toxicity to aquatic organisms, as well as potential long term risks to human health due to bioaccumulation and contamination of the drinking water supply. Effective degradation under UV also meant that, in areas of high solar radiation, solar assisted photocatalytic systems could be developed as low chemical, relatively low energy systems. Compared with other methods, such as activated carbon adsorption, or simple dilution, UV photocatalysis had the added advantage of converting mancozeb into less harmful products, reducing both acute and chronic ecological effects. Overall, the result support the ultraviolet photocatalytic degradation as a promising and environmentally friendly technology to mitigate pesticide pollution, particularly for mancozeb containing effluents.

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