Metaldehyde: A Brief on Biodegradation and Detection Methods

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ABSTRACT

Metaldehyde has been used to eradicate plant pests such as golden apple snails and slugs. This toxic pesticide has adverse effects on the environment including human health and animals. The biodegradation process naturally reduces the toxicity of this harmful pesticide, but in a long term. Many studies have been conducted in analysing the biodegradation process in water and soil. In addition, various methods have been proposed in detecting this pesticide, especially in the aquatic environment. Both biodegradation and detection are essential fields in ensuring the sustainability of our environment. This paper gives a brief idea on both concepts, and works as a platform for young researchers in the environmental pollution.

Key Words: Biodegradation, Sustainable Environment, Pesticide Detection

INTRODUCTION

Metaldehyde is one of thousands active ingredients used commercially to protect plant from pest likes snails and slugs (TOXNET, 2010). This pesticide is sold under various commercial name including Siputox in Malaysia (ACM, 2017) and Ariotox in Australia (EXTOXNET, 1996). This pesticide is believed to be very efficient as compared to other chemicals in killing golden apple snails in paddy farming area (TNAU, 2017). Generally, golden apple snail can cause huge losses to farmers.

This is because the pest can destroy up to one square meter of young paddy plant area in overnight. For this reason, the use of metaldehyde can prevent farmers from experiencing losses and able to increase their agricultural yield such as vegetable and rice as it is free from plant threats (RKB, 2017). However, once this pesticide enters the environment it invites problems such as pollution and toxicity to the environment, humans, and animals (Damalas & Eleftherohorinos, 2011).

Metaldehyde is believed to be very persistent in the aquatic environment because it has low vapour pressure feature that made it remain in the environmental compartments such as top soil and aquatic surface, rather than evaporate to the atmosphere (Saito et al., 2008; US EPA, 2006). Furthermore, metaldehyde is considered as a very tough chemical to be removed from water because of it extreme persistency characteristic (Fawellwz, 2014).

Metaldehyde was investigated to be less influenced by hydrolysis and photolysis abiotic degradation mechanisms. This stable compound was vastly mobile in sediment and dissipates into water sources (US EPA, 2006). The persistency of metaldehyde in environment had activated a warning alarm to human all over the world. This toxic compound has been found in significant level of concentration in water sources, especially in water surface due to its solubility characteristic in water. This pesticide can be detected in various types of soil and groundwater because of the mobility potential of this compound (Stuart et al., 2012).

BIODEGRADATION OF METALDEHYDE

There is a natural mechanism called biodegradation process that eradicates this pesticide from the environment on a gradual basis. As soon as the pesticide enters the surrounding environment system, it will undergo a biodegradation process that decreases its concentration. The biodegradation process is caused by microorganisms that are present abundantly especially in soil and water (Porto et al., 2011; Arbeli & Fuentes, 2007). On the other hand, cyanobacteria for example, is believed to degrade the organic pesticides in water (Kuritz & Wolk, 1995).

This natural degradation process of metaldehyde in soil happens in two conditions either with presence of oxygen or without oxygen. The half-life of this pesticide is longer under anaerobic condition (without oxygen) as compared to aerobic condition (presence of oxygen). The difference of half-life is about five months for both conditions (EFSA, 2014). In soil compartment, metaldehyde is absorbed by clay and organic matter due to its chemical properties. Besides that, the presence of this molluscicide in the soil and water sources affects human and animals that might consume aquatic animals, drink the water, or live in the polluted area (US NLoM, 1995).

In EFSA (2010), the degradation of metaldehyde in laboratory soil samples has been investigated in both conditions; aerobic and anaerobic. At first, no degradation process was detected in the dark with presence of oxygen. However, after a lag phase achieved in three weeks, it showed very low persistence and forming no main derivatives of metaldehyde. On the other hand, metaldehyde condition was investigated to be essentially stable beneath anaerobic state especially in the dark.

In another case, microorganisms were found to be very important agent to biodegrade metaldehyde in the soil. The examples of the potential biodegradable agents of metaldehyde are bacteria namely *Acinetobacter* and *Variovorax* as highlighted in Thomas et al. (2017). It was proofed that microorganisms are important in biodegrading metaldehyde. A number of soil samples were introduced into the enrichment culture containing known bacteria together with 100 mg/L of metaldehyde. The bacteria in the cultures used metaldehyde as their sole carbon source to grow. They metabolised metaldehyde and indirectly degraded the concentration of metaldehyde in the cultures samples.

In another study, Rolph et al., (2014) described that microbes boosted the biodegradation process of metaldehyde in acclimated sand samples to treat raw water. There were numerous species of microbes that can stand high concentration level of metaldehyde and removed this compound from water. However, highly presence of metaldehyde in water altered the community of microbes.

There is another biological method used in eliminating metaldehyde from the soil. This method is quite interesting as they used earthworms as the samples. In the study, Gavin et al. (2012) reported that earthworms played an important role in removing metaldehyde applied on land. A quick vanishing of the metaldehyde pellet was monitored. The pellets were taken by earthworm for the food supply. These annelida consumed the smaller pellets not more than three days. It was very fast elimination work. The consumption of metaldehyde by earthworms can reduce the concentration levels of metaldehyde and also reduces the contamination rate of this compound. The earthworm has been investigated to tolerate with high concentration of metaldehyde applied in soil.

Kidd & James (1991) explained that acetaldehyde is the main degradation product of metaldehyde in the water. Later, Bieri, (2003) discovered the fate of metaldehyde in aquatic ambient. In details, metaldehyde has been hydrolysed into its monomer, namely acetaldehyde followed by oxidation process into acetic acid and finally to carbon dioxide and water. Figure 1 depicts the environmental fate of this pesticide in the water.

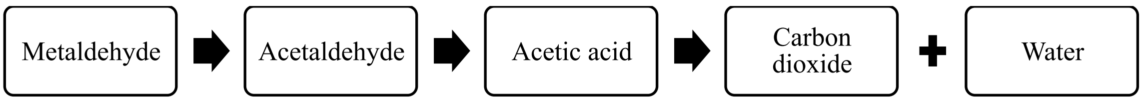

Figure 1: Pathway of Metaldehyde in the Environment
(Source: Bieri, 2003)

DETECTION OF METALDEHYDE

Early detection of this pesticide in the water resources can save millions of lives. Furthermore, at certain temperature, metaldehyde with an aqueous solubility of 220 mg/L will easily percolate from site applied and contaminate the watercourse and groundwater source. Consequently, several analytical methods have been practiced by researchers to detect metaldehyde substance existence in water sources (Autin et al., 2012). There are four steps involved in detecting metaldehyde. This analytical analysis starts with sampling (collecting samples), followed by extraction procedure (using solvents, extractors or kit), samples clean-up and ended with detection using high-end equipment (Rodrigues et al., 2013). Figure 2 shows the procedures involved in metaldehyde detection.

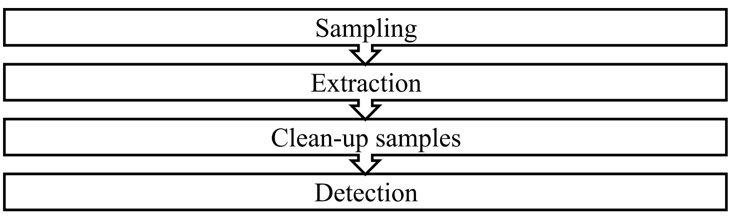

Figure 2: Procedures in metaldehyde analysis
(Source: Rodrigues et al., 2013)

Generally, chromatographic methods can be applied to detect metaldehyde compound in the water samples. These methods were proven to be very significant and fast, including high-performance liquid chromatography (HPLC), high performance liquid chromatography-coupled fluorescence, ultra-performance liquid chromatography-electrospray tandem mass spectrometry (UPLC-ESI-MS) (UK EA, 2009; Li et al., 2010). Other than that, gas chromatography (GC) (Bonansea et. al., 2013) and gas chromatography-mass spectrometric (GC-MS) (UK EA, 2009; Rolph et al., 2014) were also used to quantify metaldehyde in water samples. Table 1 tabulated the methods.

Table 1: Metaldehyde Detection Techniques

Detection	References
High performance liquid chromatography (HPLC)	UK EA (2009)
High performance liquid chromatography-coupled fluorescence (HPLC-FLD)	Li et al. (2010)
Ultra-performance liquid chromatography-electrospray tandem mass spectrometry (UPLC-ESI-MS)	Li et al. (2010)
Gas chromatography (GC)	Bonansea et al. (2013)
Gas chromatography-mass spectrometric (GC-MS)	UK EA (2009) Rolph et al. (2014) Autin et al. (2012)
Liquid chromatography-tandem mass spectrometry (LC–MS/MS)	Schumacher et al. (2016)

To our knowledge, GC-MS is the best and suitable technique to detect metaldehyde in water. Before GC-MS detection, the samples were extracted and cleaned-up using applied method. In details, the water samples were taken from raw water, ground waters and drinking water. The water samples then were extracted using solid phase extraction (SPE) cartridge in the laboratory and at a room temperature. After that, mixture of ethyl acetate, acetone and *iso*-octane were eluted. The extracted samples were analysed using gas chromatography with mass spectrometric (GC-MS) detection in selective ion monitoring (SIM) mode and under split-less injection (UK EA, 2009; Autin et al., 2012).

Rolph et al. (2014) and Pahadia et al. (2005) also detected metaldehyde compound in water sample using SPE method, followed by GC-MS analysis. In all the above studies, 10 mL of methanol and 2 mL of distilled water were used to condition the SPE catridge comprise of styrene-divinylbenzene, strata, and 200 mg/ 3mL according to the procedure given. Metaldehyde d16 was used as an internal standard together with spiked aqueous sample. The spiked samples were examined by GC-MS. The limit of detection (LOD) for GC-MS was determined at 0.05 µg/L.

In another study, ultra-performance liquid chromatography-electrospray tandem mass spectrometry (UPLC-ESI-MS) was used to detect and quantify metaldehyde concentration in the water samples. There were 10 water samples collected from different sampling locations represented different area of environment including river and stream. The samples were extracted using SPE procedure. A conditioned C18 cartridge was used to purify water samples. After that, 2 mL of methanol and water with ratio of 85:15 v/v were used to elute metaldehyde. Subsequently, UPLC-MS-MS was applied to analyse metaldehyde in the extracted samples (Li et al., 2010).

Furthermore, external standard calibration curve technique was implemented to measure the concentration levels of metaldehyde in the samples. This extraction and detection methods were appropriate for the determination of metaldehyde concentration in the water samples because exhibited good linearity at a concentration of 0.05 to 5 ng/mL. The value of 0.99 linear relative coefficient was obtained. Besides that, high recoveries were detected which range from 96.1 to 106.3%. The low limit of detection of metaldehyde 0.003 ng/mL was found by using this technique (Li et al., 2010).

CONCLUSION

As for the conclusion, both biodegradation and detection of toxicity pollutants are essential fields in ensuring the sustainability of our environment. However, more researchers on biodegradation of metaldehyde in the environmental compartments such as water and soil should be explored to provide better understanding on the environmental fate of this toxic pesticide. On the other hand, the development of more advance and quick detection method is very important in the present and future to provide early alarming system of this metaldehyde contamination.

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Nine Treatments of Metaldehyde in Water

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ABSTRACT

Metaldehyde is widely applied in the agricultural sector, largely in the removal of snails and slugs. Its water-soluble characteristic causes the compound to end up in our watery system. This is very dangerous as it is able to pollute drinking water and food source; thus adversely affect our health. Hence, removing metaldehyde is an important task for those involved with the treatment of water. For this reason, this paper gathers nine various methods which are proposed and used in treating water contaminated with metaldehyde. The simple but significant explanation will be able to assist those who are new in the fields of pesticide removal and water treatment. In parallel, it will be the basis for more advanced researches in the future.

Key Words: Metaldehyde, Water Treatment, Environmental Engineering

INTRODUCTION

Nearly 75% of the causes of infectious diseases are associated with water borne, as water is the main source of life especially in terms of drink and self-cleaning (Khan et al., 2017). However, contaminated sources of water, especially from chemical substances, are much disruptive. The pesticides will either dissolve or integrate with aquatic organism, plant and sediment (Salvestrini et al., 2017). Some of these pesticides are water soluble and can last long in the medium. Metaldehyde is one of them.

Metaldehyde is an effective molluscicide in overcoming the problem caused by snails and slugs (Brice et al., 2017). However, the metaldehyde fate will end in the water system through various ways such as leaching, spillage, run-off, erosion, absorption, adsorption and spray drift (BCMA, 2017). Metaldehyde taken through ingestion system can harm the stomach and intestines. In the long run, it causes failure of human vital organs such as liver and kidneys (Kidd & James, 1991; Sax & Bruce, 1975). This harmful effects triggered panic reactions of certain parties especially those related to water management.

Some of the researchers suggest a total ban on metaldehyde in agricultural areas due to difficulty in treatment and high cost (Brockett, 2016). There is another proposal, on a buffer zone creation at least 10 meters from the nearest water source (Rush, 2017). The polemic continues as there is a contradiction between commercial use and food security. Hence, it is important for water-related researchers to seek for the safest, cheapest and most practical treatment. For this reason, this paper explores some of the methods that have been proposed and applied in treating this harmful chemical, namely metaldehyde.

METALDEHYDE TREATMENTS

The presence of metaldehyde in drinking water sources was detected by Bristol Water, a water supply company in the United Kingdom (UK) (Blake, 2008). It was concluded based on samplings of water taken at the Sharpness Canal in 2007. This discovery triggered anxiety when the rate beyond the standard limit for pesticides set in the UK and European countries (Marshall, 2013). Metaldehyde toxicity is closely related to the food chain. As humans are positioned at the top of the food chain, the negative consequence is multiplied as a result of the biomagnification effect (Favari et al., 2002).

With the aim to reduce or eliminate metaldehyde compound from the watercourses, researchers have come out with various methods of treatments. In general, these treatments can be categorized into two which are carbon-based treatment and non-carbon-based treatment. Treatments that use carbon as a base are powdered activated carbon, granular activated carbon and phenolic carbon tailored. On the other hand, catchment management, photodegradation method and bio-filtration process are among non-carbon-based treatment, as shown in Table 1.

Table 1 Metaldehyde Treatments

Category	Type
Carbon-based treatment	Powdered activated carbon
	Granular activated carbon
	Phenolic carbon tailored
Non-carbon-based treatment	Catchment management
	Bio-filtration process
	Coupled adsorption with electrochemical destruction
	Photodegradation method
	Polymeric sorbent
	Chlorination and Ozonation

Carbon-Based Treatments

Powdered Activated Carbon

High carbon content such as charcoal is the main input to the powdered activated carbon. It is widely applied in the United Kingdom for water treatment purpose (TrojanUV, 2016). This technique uses adsorption process to degrade pollutants from industrial and agricultural wastes in water. It is applied to eliminate unwanted odour and taste in water, caused by high levels of pesticides used in certain seasons (Yoon et al., 2003). Approximately 90% of metaldehyde in water can be removed using this treatment. Physical properties of this carbon material, especially its small particle size and high surface area are the factors (Li et al., 2017). In the same study, this method has an absorption rate almost similar to the granular activated carbon. In contrast, it offers economical investment cost than granular activated carbon (Knappe et al., 1998). However, its extensive disposal of waste creates concern to the public (Renou et al., 2008).

Granular Activated Carbon

Still applying the same source as in 2.1, but the output is in a bigger form. Using granular, a study by Salvestrini et al. (2017) shows its high capability of absorption and adsorption for metaldehyde treatment. This is due to higher specific surface area and high point of zero charge. Furthermore, absorption of metaldehyde on the surface of granular is due to electrostatic interactions and/or hydrogen bonding between molecular electronegative oxygen molecules and positive surfaces imposed by the adsorbents. In Tao & Fletcher (2013), the granular system adsorbs metaldehyde faster than a non-functionalised hyper-cross-linked polymer Macronet (MN200). For its weakness, more than 25% of adsorbed metaldehyde is leached due to the destruction of adsorbate molecules in the granular. In treating metaldehyde from water, this treatment faces shorten life span. In other word, the materials need to be replaced regularly and this this pulls a lot of money from water treatment companies (Franks, 2017).

Phenolic Carbon Tailored

This innovation is based on the preliminary idea, suggested by Ragan et al. (2012). It is stated that the elimination of pesticides is not merely dependent on the surface of a medium, but is also determined by the efficiency of porosity control. This statement is successfully supported by results recorded in Busquets et al. (2014). Tailored phenolic resin-derived carbon sealed up to three times the metaldehyde as compared to its own surface. At the same time, the conventional granular activated carbon method only sought to absorb metaldehyde twice over. In comparison, this method absorbs metaldehyde up to 63mg/g more rather than the granular method. Compared to other activated carbon sources, this activated carbon is in the form of fine beads and is derived from phenolic resins. This research also serves as the foundation for the comparison in a recent study, as written by Li et al. (2017).

Non-Carbon-Based Treatments

Catchment Management

Catchment management is an effort run by several parties including governments, water supply companies and non-governmental organizations (NGOs). In this alternative system, farmers are introduced to alternative pesticides which are easier to degrade in the environment (WCMA, 2016). For those who still want to use the metaldehyde, they are exposed to the right way. This is to avoid negative effects of spillage and leaching (Castle et al., 2017). Another initiative undertaken is building up a shallow waterway. In absorbing this chemical compound from flowing into the main drinking water source, the grass is planted along the waterway (BCMA, 2017). However, this method has a minimal impact. This is due to the reluctance of the relevant parties, especially farmers. In addition, cost is one of the obstacles. Alternative pesticides are likely to involve high input costs, but provide a reverse return (Thames Water, 2017).

Bio-Filtration Process

Slow sand filter is a biological method to treat water. Basically, this method only requires a container filled with one layer of sand followed by a layer of gravel that serves as a filter to treat water (Logsdon, 2002). This cost saving system is not only easy to design but also works effectively in filtering water for residents up to 5,000 people (WHO, 2000). Also emphasized in this report, it is an eco-friendly water management as no by-products are generated from this technique. This is confirmed through a study conducted by Rolph et al. (2014). This filter positively removes metaldehyde from water in a full scale and lab scale experimental set-up. However, it must be adjusted under suitable conditions such as length time and flow rate. Sharing the same view is Gasperi et al. (2010). Compactness, modularity and intensiveness are the three main reasons why this method is preferred as compared to activated sludge tank.

Coupled Adsorption with Electrochemical Destruction

This method which combines two types of procedures; absorption and electrochemical destruction was applied by Ashgar et al. (2012). The study was further developed by Mohammed et al. (2012) and Nabeerasool et al. (2015). In Mohammed et al. (2012), focus is given to two important entities in this system, absorbent medium (low cost but capable) and electrochemical medium (expensive but less efficient).

In Nabeerasool et al. (2015), metaldehyde is the only focus in the use of this treatment. It is concluded that this combined technique removes metaldehyde from natural water effectively. At the same time, the results obtained comply with the standards set by the United Kingdom and other European countries. The key to this success is the complete oxidation process (Brown et al., 2004). This process is able to degrade and abolish chemical compounds. Furthermore, no poisonous spinoff is leaved out in the aquatic environment.

Photodegradation Method

In general, pollutants in water are mineralized to carbon dioxide and water by hydroxyl particles that are generated from ultraviolet radiation (Krishnan et al., 2017). In a laboratory scale, a combined method using photo-oxidation with advanced oxidation process and ultraviolet radiation with titanium dioxide (UV/TiO_2) and hydrogen peroxide (UV/H_2O_2) successfully removes metaldehyde from water (Autin et al., 2012). The degradation process happened due to the chemical characteristics of materials used. However, the situation is quite different in the actual water system. Presence of non-target organic matter such as other pesticide compounds reduces the degradation development of metaldehyde (Autin et al., 2013). At the same time, its high cost makes it more appropriate in treating industrial wastewater rather than be applied for sewage treatment in the agricultural sector, as highlighted in the same study.

Polymeric Sorbent

Ion-exchange resin is a material that acts as an ion exchange medium. It is an insoluble polymer and micro in size. The main ingredient of ion-exchange is resin, which is derived from plants (McNaught & McNaught, 1997). This method removes metaldehyde by replacing its ions with similarly charged ions (NHDES, 2009). Both chemical compounds with similar charges are eliminated in the next process. A research by Tao & Fletcher (2013), discovered that ion exchange resin S957 which comprised of macroporous with high phosphonic and sulfonic acid assemblage, excellently removes metaldehyde compound from raw water. It is also a strong acid cation and an efficient chelating agent. In the same study, no leaching of any other compound is observed. A fact given by Polysciences Inc. (2016) acknowledged that ion exchange method is the most economical and capable of treating high quality water including nuclear power plants.

Chlorination and Ozonation

Chlorination and ozonation are two common methods applied in treating contaminated water, and are still subjects of research until now (Khatun et al., 2017; Wei et al., 2017). Occasionally comparisons are made in finding the best solution between these two treatments (Chapdeline, 1993). If used in excessive doses, both treatments can risk end-