



Mecoprop (MCP) removal in full-scale rapid sand filters at a groundwater-based waterworks

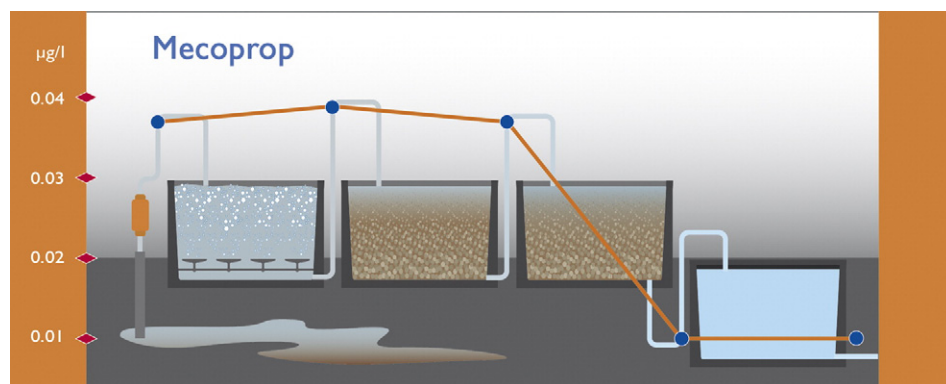
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HIGHLIGHTS

- A full-scale groundwater based waterworks was able to remove MCP.
- In the secondary rapid sand filters, MCP decreased from 0.037 µg/L to <0.010 µg/L.
- The filter sand removed MCP both by sorption and by microbial degradation.
- Microbial removal was unchanged while sorption decreased with depth of the filter.

GRAPHICAL ABSTRACT



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ABSTRACT

Contamination by the herbicide mecoprop (MCP) was detected in groundwater abstraction wells at Kerteminde Waterworks in concentrations up to 0.08 µg/L. MCP was removed to below detection limit in a simple treatment line where anaerobic groundwater was aerated and subsequently filtered by primary and secondary rapid sand filters. Water quality parameters were measured throughout the waterworks, and they behaved as designed for. MCP was removed in secondary rapid sand filters — removal was the greatest in the sand filters in the filter line with the highest contact time (63 min). In these secondary sand filters, MCP concentration decreased from 0.037 µg/L to below the detection limit of 0.01 µg/L. MCP was removed continuously at different filter depths (0.80 m). Additionally, biodegradation, mineralisation and adsorption were investigated in the laboratory in order to elucidate removal mechanisms in the full-scale system. Therefore, microcosms were set up with filter sand, water and ¹⁴C-labelled MCP at an initial concentration of 0.2 µg/L. After 24 h, 79–86% of the initial concentration of MCP was removed. Sorption removed 11–15%, while the remaining part was removed by microbial processes, leading to a complete mineralisation of 13–18%. Microbial removal in the filter sand was similar at different depths of the rapid sand filter, while the amount of MCP which adsorbed to the filter sand after 48 h decreased with depth from 21% of the initial MCP in the top layer to 7% in the bottom layer. It was concluded that MCP was removed in secondary rapid sand filters at Kerteminde Waterworks, to which both adsorption and microbial degradation contributed.

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1. Introduction

Groundwater is used as a source of drinking water all over the world (IWA, 2012). Concentrations of natural and anthropogenic inorganic

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and organic compounds, such as iron (II), manganese (II), ammonium, arsenic and pesticides, may exceed drinking water guidelines (European Parliament and Council, 1998) in abstracted groundwater which calls for treatment before distributing to consumers.

Pesticides are frequently detected in groundwater abstraction wells all over Europe (EEA, 2004). However, the concentration of pesticides in drinking water and groundwater should not exceed 0.1 µg/L for a single compound, or 0.5 µg/L for the sum of all pesticides (European Parliament and Council, 2006). Pesticides can be removed in drinking water treatment systems by applying advanced technologies such as advanced oxidation (e.g. Suty et al., 2004) and granular activated carbon filtration (GAC) (e.g. Heijman et al., 2002). However, the cost-effectiveness and environmental sustainability (Godskesen et al., 2011) of these processes still remain uncertain.

In Denmark, water treatment is generally simple with aeration of anaerobic groundwater. Aeration is followed by filtering in rapid sand filters designed for a contact time between 7.5 and 20 min. No disinfection is included in the treatment process (Winter et al., 2003). The purpose of aeration is to add oxygen (to a concentration of 8–10 mg/L) to the anaerobic inlet water, while volatile compounds such as methane and hydrogen sulphide are stripped off. Increased oxygen concentrations are necessary for the subsequent removal of iron (II), manganese (II) and ammonium.

Rapid sand filters are biological filters used in drinking water treatment plants globally (Rittmann et al., 2012; Zearley and Summers, 2012; Mouchet, 1992). They constitute a highly complex system of several simultaneous removal mechanisms whereby iron (II) and manganese (II) are removed by physico-chemical and biological oxidation processes and precipitation (Mouchet, 1992; Tekerekopoulou et al., 2013), while ammonium is oxidised biologically in the nitrification process into nitrite and then nitrate (Lytle et al., 2007).

Besides removing inorganic components, different investigations have shown that biological filters can remove organic chemicals such as methyl *tert*-butyl ether (MTBE) (Arvin et al., 2004), 2-methylisoborneol (MIB) and geosmin (Ho et al., 2007). Though lab-scale studies have shown a biological removal potential of pharmaceuticals and pesticides (Zearley and Summers, 2012; Hedegaard and Albrechtsen, 2014), it is still uncertain if full-scale rapid sand filters have similar removal potentials, and the biological and chemical processes involved remain unknown (Benner et al., 2013).

Phenoxy acids, including mecoprop (MCPP), are some of the most frequently detected pesticides in groundwater, and they have been used extensively as herbicides in agriculture (Buss et al., 2006). MCPP is still used in some European countries, i.e. France, Italy and Austria (EU Pesticide Database, 2013), but its use has been restricted in Denmark (The Danish Ministry of the Environment, 2013; GEUS, 2013). However, in Denmark, MCPP was detected in 4.2% of the investigated groundwater samples in the period 1990–2012, and the guideline value of 0.1 µg/L was exceeded in 1.1% (GEUS, 2013). Usually, when the guideline is exceeded the normal practice is to close or dilute water from contaminated abstraction wells.

The water treatment at Kerteminde Waterworks, Denmark, consists of the same simple treatment line as other waterworks in Denmark. MCPP contamination has been detected below the guideline value of 0.1 µg/L in abstracted anaerobic groundwater for more than nine years (Fig. 1) (Jupiter, 2013), and it has been observed that the herbicide was removed during the treatment (Ferguson et al., 2009). However, it is not known where in the treatment line MCPP was removed, or which mechanisms governed the removal. These questions cannot be answered based on the international literature, so the purpose of this investigation is to:

- Investigate where in the waterworks treatment line MCPP was removed from the water phase.
- Investigate which mechanisms were responsible for the removal of MCPP.

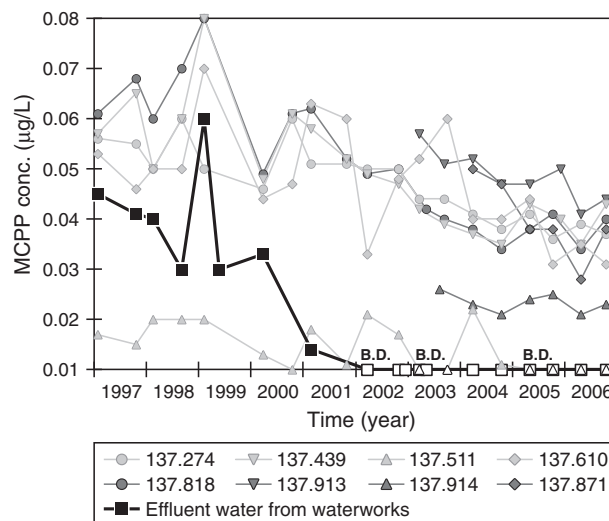


Fig. 1. MCPP concentrations at Kerteminde Waterworks from the initial MCPP detection up to the investigations, measured in abstraction wells (well numbers 137.274, 137.439, 137.511, 137.610, 137.818, 137.913, 137.914, 137.971) and in effluent water taken from the waterworks (Jupiter, 2013).

- Investigate whether compounds which the waterworks was designed to remove were actually removed from the water as expected.

2. Materials and methods

2.1. Investigations at Kerteminde Waterworks

Kerteminde Waterworks, Denmark, is a typical Danish municipal groundwater-based waterworks operating a simple treatment line (Fig. 2). The waterworks was expanded in 1979, and as a result it now has two parallel filter lines. In the treatment line anaerobic groundwater is aerated by diffused air aerators, and filtered in three parallel primary rapid sand filters. Afterwards, the water is filtered in parallel secondary sand filters — with a distribution of approximately 20% of the water to four filters in Filter line 1, which are part of the original waterworks, and 80% to two parallel filters in Filter line 2, which was added in 1979. Filter characteristics are described in Table 1. The water is finally stored in clean water tanks before being distributed to consumers. No disinfection is included in the treatment process. The water quality of the groundwater is characterised by reduced conditions in the aquifer, expressed by low nitrate concentrations, for instance (Table 2).

During the investigations Kerteminde Waterworks distributed approximately 600,000 m³ of water per year and received raw water from eight groundwater abstraction wells, seven of which were contaminated with varying concentrations of phenoxy acids, especially MCPP (from below 0.01 µg/L to 0.080 µg/L, Fig. 1). The waterworks was operated 24 h a day, with varying flow according to consumption. The inlet concentration of MCPP depended on the combination of abstraction wells and could be anything up to 0.08 µg/L. The investigations in this paper were carried out in two steps, as described in the following.

MCPP removal at Kerteminde Waterworks. It was investigated where in the treatment line MCPP was removed. Water quality parameters were measured throughout the waterworks in Filter line 1 (Fig. 2) (Table 2), where the secondary rapid filters had a contact time of 63 min (Table 1).

MCPP removal mechanisms at Kerteminde Waterworks. Four months after the investigations in Filter line 1 the removal of MCPP was investigated throughout Kerteminde Waterworks in Filter line 2, by focusing on the secondary rapid sand filters with a contact time of 8 min (Table 1). Reactions in the filter sand were investigated at

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