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Microbial degradation of pesticides in rapid sand filters for treatment of drinking water

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Abstract

In Denmark drinking water supply is based on groundwater which is treated by aeration followed by filtration in rapid sand filters. Unfortunately pesticide contamination of the groundwater poses a threat to the water supply, since the simple treatment process at the waterworks is not considered to remove pesticides from the water phase and pesticides are detected in 24% of the active Danish waterworks wells. This study aimed at investigating the potential of microbial pesticide removal in rapid sand filters for drinking water treatment. Removal of the pesticides MCPP, bentazone, glyphosate and the degradation compound p-nitrophenol was investigated in the rapid sand filters at Islevbro and Sjælsø waterworks plant I and II. Microcosms were set up with sand from rapid sand filters, water and an initial pesticide concentration of 0.03-0.38 µg/L. In all the investigated waterworks the concentration of pesticides in the water decreased – MCPP decreased to 42-85%, bentazone to 15-35%, glyphosate to 7-14% and *p*-nitrophenol 1-3% – from the initial concentration over a period of 6-13 days. The largest microbial removal was observed at Sjælsø waterworks Plant II, where the pesticides were partially mineralised – up to 43% of the initial glyphosate was found as CO_2 after 6 days. At Sjælsø waterworks Plant II the contact time in the primary rapid sand filter was 43 minutes. It was found that less than 20 minutes was needed to biologically remove more than 50% of the initial bentazone (concentration 0.1 µg/L). It is therefore certain that there is a potential for microbial removal of pesticides from contaminated groundwater in Danish waterworks.

Keywords: pesticides; degradation; rapid sand filters; groundwater; waterworks

Introduction

In Denmark generally all drinking water is based on groundwater. Unfortunately, large parts of the groundwater is affected by pesticides. In 2012, pesticides and metabolites were detected in 24% of the active waterworks abstraction wells (Figure 1) (GEUS, 2012). In Denmark the water treatment is simple consisting of aeration of anaerobic groundwater followed by filtration in primary and secondary rapid sand filters, subsequently the water is stored until it is distributed to the consumers. No disinfection is included in the treatment process. Pesticides pose a threat to the water supply since none of the water treatment steps are known to remove pesticides, hence, the normal strategy is to substitute contaminated wells or dilute the water to meet the guideline value on 0.1 µg/L.
Recent investigations have shown that the herbicide MCPP was removed in the secondary rapid sand filters at Kerteminde waterworks, Denmark, and microbial processes were involved in the removal (Ferguson et al., 2009). Due to the environmental and economic sustainability of rapid sand filters compared to advanced treatment methods it is of large interest to utilise this water treatment method to remove pesticides (Hedegaard and Albrechtsen, 2014). This study aimed at investigating the potential and kinetics of microbial pesticide removal in rapid sand filters for drinking water treatment. The removal of MCPP, bentazone, glyphosate and p-nitrophenol was investigated in the rapid sand filters at Islevbro and Sjælsø waterworks plant I and II.

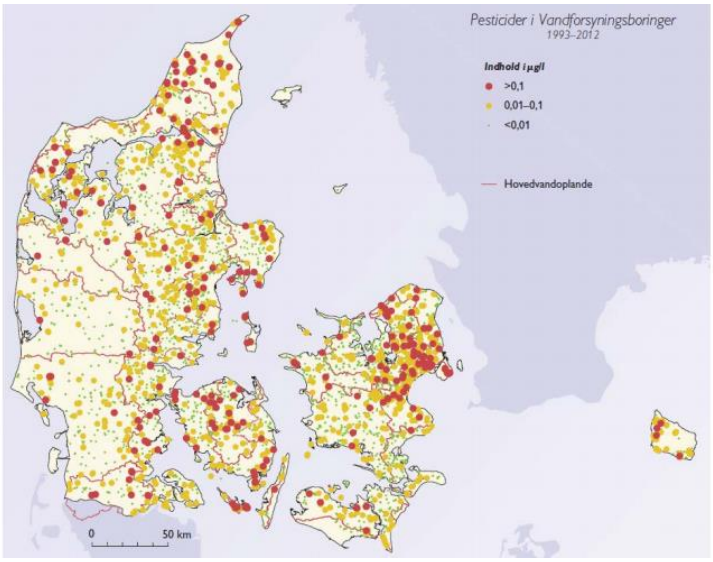

Figure 1 Detected pesticides in waterworks abstraction wells in Denmark during the period 1993-2012. Red dots marks wells with concentrations above the guideline value >0.1 µg/L, yellow dots with 0.01-0.1 µg/L and green dots <0.01 µg/L (GEUS, 2013).

Method

Filter sand was collected at the waterworks and microcosms were set-up within 24 hours with filter sand, water and ^{14}C-labelled pesticide at an initial concentration of 0.03-0.38 µg/L. Water samples were collected over time. Suspended matter was removed from water samples by filtration. The analysis for ^{14}C was based on a double vial system (Figure 2). Each water sample was transferred to a 20mL plastic vial, and a 7 mL vial with 1mL 2M NaOH was positioned inside the 20mL vial. HCl was added to the water sample to strip off produced $^{14}CO_2$ from mineralisation of ^{14}C-pesticide and the double vial system was closed. The $^{14}CO_2$ was caught by the base trap, making it possible to quantify the ^{14}C-pesticide in the water sample and $^{14}CO_2$ in the base.

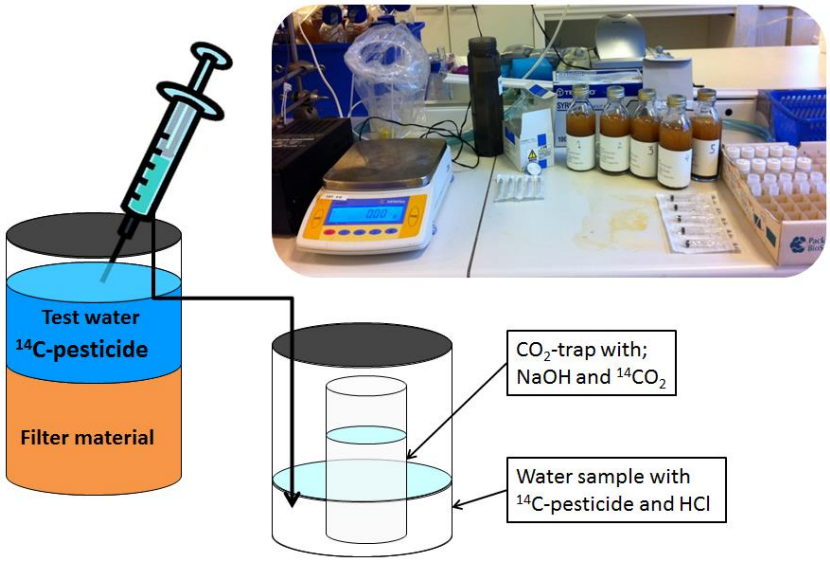

Figure 2 Microcosms consisting of filter sand, water and ^{14}C-pesticide. Water samples were collected, filtered and transferred to 20mL plastic vials. HCl was added to water samples to strip off $^{14}CO_2$ from mineralisation of ^{14}C-pesticide and $^{14}CO_2$ was caught by a base trap.

Results

There was an evident removal potential of pesticides in the investigated filter sand. All investigated pesticides and degradation compounds were partially removed in samples from all the investigated rapid sand filters after 6-13 days (Table 1). The pesticide concentration in the water decreased substantially – MCPP decreased to 42-85%, bentazone to 15-35%, glyphosate to 7-14% and *p*-nitrophenol 1-3%. Comparing results from abiotic controls with removal in microcosms, it was clear that the removal was partially biological, since less pesticide was left in the microcosms than in the abiotic controls, except from the MCPP and glyphosate removal at Sjælsø waterworks Plant I (Table 1). Biological removal was largest in Sjælsø waterworks Plant II, where removal of bentazone, glyphosate and *p*-nitrophenol lead to mineralisation – 8-14% of the initial betazone, 42-43% of the initial glyphosate and 7-10% of the initial *p*-nitrophenol was recovered as CO_2 after 6 days (Table 1).
Bentazone is still legally used in the European Union (EU pesticide database, 2013), and it is hard to degrade in aquifer material (Broholm et al., 2001). Due to the promising biological removal of bentazone at Sjælsø waterworks Plant II (Table 1), the removal rate of bentazone was investigated with sand from these filters. The primary rapid sand filters at Sjælsø waterworks Plant II had a contact time of 43 minutes.
Bentazone was removed rapidly in the microcosms – After 20 minutes less than 50% of the initial bentazone was left in the microcosms (Figure 3). There was no detected removal in the corresponding autoclaved control or in a microcosm with water only. Hence, the removal of bentazone was a microbial process and removal must have been caused by processes in the filter sand since there was no removal in the microcosm with water only.

Table 1 The fractionation of ^{14}C-bentazone of the initial $^{14}C_0$ in the water phase. Incubation with filter material from different rapid sand filters. Data are from microcosms (two replicates) and abiotic controls (Modified from Hedegaard and Albrechtsen (2014)).

	Remaining bentazone in water phase ($^{14}C/^{14}C_0$)		**$^{14}CO_2$-production from degradation ($^{14}CO_2/^{14}C_0$)**	
	Microcosms	**Abiotic control**	**Microcosms**	**Abiotic control**
Islevbro	**13 days**		**13 days**	
MCPP	42-48%	57-61%	-	-
Bentazone	26-33%	74-83%	-	-
Sjælsø Plant I	**6 days**		**6 days**	
MCPP	67-74%	67%	-	-
Bentazone	31-35%	62%	-	-
Glyphosate	7-8%	4%	-	-
p-nitrophenol	1-3%	22%	-	-
Sjælsø Plant II	**6 days**		**6 days**	
MCPP*	70-85%	92%	-	-
Bentazone	15-18%	103%	8-14%	-
Glyphosate	9-14%	8%	42-43%	-
p-nitrophenol	3%	96%	7-10%	-

* low initial concentrations (0.033-0.036 µg/L) – uncertain results.

- no evident tendency in results

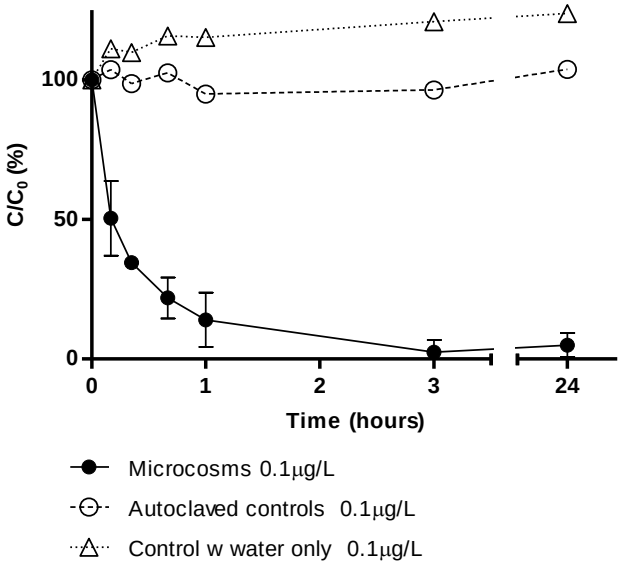

Figure 3 Removal of bentazone in microcosms consisting of 100 g filter sand and 100 ml water from Sjælsø waterworks Plant II. The mean concentrations and standard deviation are given as percentage of the initial concentration. Microcosms with an initial concentration of 0.1 µg/L (triplicate) are depicted along with two corresponding controls (initial concentration 0.1 µg/L), one with autoclaved filter sand and one with water only.

Conclusions

The investigations showed:

- An evident removal potential of MCPP, bentazone, glyphosate, and *p*-nitrophenol in samples from rapid sand filters at Danish waterworks. The microbial removal was largest in filter material taken from Sjælsø Plant II.
- In filter sand from Sjælsø waterworks Plant II bentazone concentration in the water phase decreased to less than 50% of the initial concentration within 20 minutes as a result of microbial removal.

This study showed that substantial microbial pesticide removal is possible within the contact time of rapid sand filters and thereby a potential for treatment of pesticide contaminated groundwater in Danish waterworks. This is of commercial interest due to the economical and environmental sustainability of this water treatment method.

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