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Benefits of ozone-activated carbon filtration in integrated treatment processes, including membrane systems

J. P. van der Hoek, J. A. M. H. Hofman and A. Graveland

ABSTRACT

Amsterdam Water Supply introduced ozone-activated carbon filtration (biological activated carbon filtration) in the production plant Leiduin in 1995. This full-scale plant (capacity 70 million m^3 yr^{-1}) can be characterized as an integrated treatment process. Experience since 1995 has revealed that biological activated carbon filtration has several benefits in integrated treatment processes.

It is a very effective barrier for organics in general and for pesticides and organic micropollutants in particular. Due to the preceding ozonation both adsorption and biodegradation take place, resulting in a very long lifetime of the carbon filters (2 years). The ozonation significantly increases the disinfection capacity of the plant. Together with the other process steps, the disinfection capacity is at such a high level that, in combination with the biological stability of the finished water and good engineering practice in the distribution system, no chlorination has to be applied to guarantee hygienic quality. The process stability of the final treatment step, slow sand filtration, is, importantly, increased by the introduction of ozone-activated carbon filtration. Running times of the slow sand filters are longer than 2–3 years, while before the introduction of ozone-activated carbon filtration they never exceeded 1 year.

In integrated membrane processes—which may be used in the treatment plant Leiduin in the future and therefore are being tested on pilot plant scale—ozone-activated carbon filtration also offers many advantages. In a reverse osmosis pretreatment scheme, comprising ozone-activated carbon filtration and slow sand filtration, the reverse osmosis system was shown to have a running time of 1 year without any membrane cleaning, due to the excellent reverse osmosis feed water quality. The application of ozone-activated carbon filtration also enables the use of electrodialysis as an alternative for reverse osmosis in an integrated membrane system.

It is concluded that ozone-activated carbon filtration is a versatile process, offering many benefits in integrated treatment systems.

Key words | biological activated carbon filtration, disinfection, integrated treatment, membranes, organics, ozonation

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INTRODUCTION

The use of biological filtration processes in drinking water treatment is quite common in Europe and is gaining popularity in the USA (Brink & Parks 1996). Slow sand filtration is one well-known biological filtration process; biological activated carbon filtration (BACF) is a more recent example.

BACF is activated carbon filtration preceded by ozonation and is applied for the removal of organic compounds in general, and the removal of organic micropollutants and pesticides in particular, by a combination of adsorption and biodegradation (Graveland 1994).

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In 1992 BACF was introduced to the production plant 'Weesperkarspel' (capacity 31 million m^3 yr^{-1}), and in 1995 to the production plant Leiduin (capacity 70 million m^3 yr^{-1}) (Graveland 1993; Graveland & van der Hoek 1995). One of the reasons for introducing this process to both production plants of Amsterdam Water Supply was the presence of atrazine, bentazone and metolachlor in the product water of the Leiduin plant in 1987. BACF has been in operation for more than four years at the Leiduin plant. Besides being an excellent barrier for organics such as pesticides, the process has been shown to have many more benefits in an integrated treatment process as applied at the Leiduin plant. It results in a significant increase in disinfection capacity, very stable and reliable operation of the slow sand filters following BACF, and the possibility of producing hygienically safe and biologically stable water without the use of a disinfectant residual. In addition, when it is used in an integrated membrane system, which will be applied in an extension scheme of the Leiduin plant, it contributes to the avoidance of colloidal, organic and biological fouling of the membrane system. Stable operation of the reverse osmosis units can be achieved while, as a result of the versatility of BACF, electrodialyis may become an alternative for reverse osmosis in the integrated membrane system.

In this paper, the use of BACF in the Leiduin plant will be evaluated, both for the existing treatment scheme and for a future scheme applying membrane filtration. Results will be presented, originating from the full-scale production plant and from pilot plants, operated in parallel with the full-scale plant.

THE PRINCIPLES OF BIOLOGICAL ACTIVATED CARBON FILTRATION

BACF is based on the combination of ozonation and activated carbon filtration. The water is first ozonated with a relatively low ozone dose (0.3–0.45 mg O_3 mg^{-1} DOC) to produce easily assimilable organic carbon (AOC) up to a concentration of 50–150 μg l^{-1}. As a result, the biodegradation capacity in the subsequent carbon filters is increased, because the increase in AOC yields a higher bacterial density in these filters (Malley *et al.* 1993; Graveland 1994; Price *et al.* 1994). Hence, the organic matter can be adsorbed and/or biodegraded, depending on the nature of the material. Compared with activated carbon filtration without preceding ozonation, the biodegradation of the organic matter is strongly stimulated, and organic matter already adsorbed on the carbon might also be biodegraded. Biological activity in the carbon filters is accompanied by oxygen consumption and alkalinity production. The biodegradation of natural organic matter (NOM) favours adsorption of pesticides and other organic micropollutants, because it lowers NOM-preloading on to the carbon. Also the more persistent organic compounds may be biodegraded as a result of the higher biological activity in the carbon filters. By optimization of the ozone dose in relation to the bromide and DOC content of the water, it is possible to minimize the bromate formation during ozonation and to control the AOC content in the final water, without losing the disinfection capacity of ozonation (van der Hoek *et al.* 1995).

MATERIALS AND METHODS

Leiduin production plant

The process scheme of the Leiduin plant, before and after the introduction of BACF, is shown in Figure 1. The production capacity is 70 million m^3 yr^{-1} (8,000 m^3 h^{-1}). Rhine river water is pretreated in the centre of the Netherlands by coagulation, sedimentation and rapid sand filtration and subsequently transported to the dune area west of Amsterdam where it is infiltrated. After a residence time of approximately 100 days the water is abstracted and collected in an open basin. This recharged water is treated by rapid sand filtration, ozonation, pellet softening, two-stage biological activated carbon filtration and slow sand filtration to achieve drinking water quality. The process conditions of the relevant process units, namely ozonation, BACF and slow sand filtration, are summarized in Table 1.

Ozonation is carried out with a relatively low ozone dose of 0.75–0.9 mg l^{-1} (0.375–0.45 mg O_3 mg^{-1} DOC). The carbon filtration is carried out in two stages. Both the

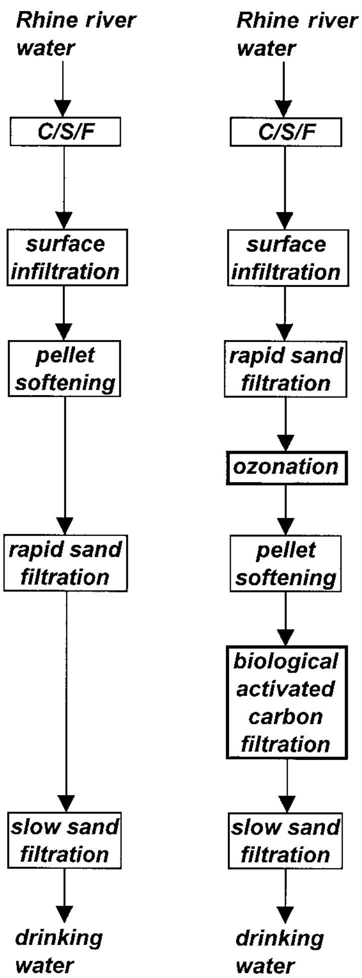

Figure 1 | Leiduin production plant before (left) and after (right) introduction of BACF.

first stage and second stage contain 20 filters and are operated with an EBCT (empty bed contact time) of 20 min. The total contact time of 40 min results in the lowest total costs (Graveland 1993, 1994). After a total operational period of 2 years, a first stage filter is reactivated, and put back into service as a second stage filter. After 1 year of operation this filter is switched to the first stage mode and operated for another year. In this way a pseudo moving bed system is created. A total operational period of 2 years with a total of 40 filters implies that every 2–3 weeks a filter bed is sent to the carbon supplier for reactivation.

The final process step at the Leiduin production plant is slow sand filtration (van der Hoek *et al.* 1996; Kors *et al.* 1996). The total surface area is 32,000 m^2 divided over 25 filters. All filters are covered to exclude sunlight and to prevent them from freezing. The filters are operated with a filtration rate of 0.25 m h^{-1}. After this final process step the water is hygienically safe and biologically stable. In combination with a reliable, stringently controlled distribution system, good engineering practice and the use of biostable materials, no disinfectant residual is necessary to ensure the quality of the drinking water during distribution. Therefore, chlorination is not applied at Amsterdam Water Supply.

Pilot plants

Two pilot plants have been operated:

1. Pilot plant A has the same process scheme as the full-scale Leiduin plant and uses the same feed water. The feed flow is 14 m^3 h^{-1}. In contrast to the full-scale plant, the carbon filters have not been reactivated since the start-up in April 1995.
2. Pilot plant B is an integrated membrane system in which BACF in combination with slow sand filtration is used as pretreatment for reverse osmosis. Figure 2 shows the process scheme. This process scheme is studied as an extension scheme for the full-scale Leiduin plant. In this treatment scheme, pretreated Rhine river water (coagulation/sedimentation/filtration) is directly treated without artificial recharge in the dune area. The process conditions of the pretreatment units are summarized in Table 2. In contrast to the full-scale plant and pilot plant A, the carbon filters are operated as a single-stage filtration process. The reverse osmosis system consists of seven pressure vessels in a 4-2-1 arrangement and is described in detail in van der Hoek *et al.* (1998, 1999).

Analyses

Atrazine, desisopropylatrazine and desethylatrazine were measured via an analytical method standardized by Amsterdam Water Supply (1998). This analysis involves

Table 1 | Process conditions for Leiduin production plant

Ozonation	Ozone dose	0.75–0.9 mg/l
	Contact time	20 min
Biological activated carbon filtration	Number of 1st stage filters	20
	Number of 2nd stage filters	20
	EBCT per stage	20 min
	Filtration rate	7 m h^{-1}
	Carbon type	Norit ROW 0.8S
	Carbon volume per filter	145 m^3
	Reactivation frequency	Once every 2 years
Slow sand filtration	Surface area	32,000 m^2
	Filtration rate	0.25 m h^{-1}

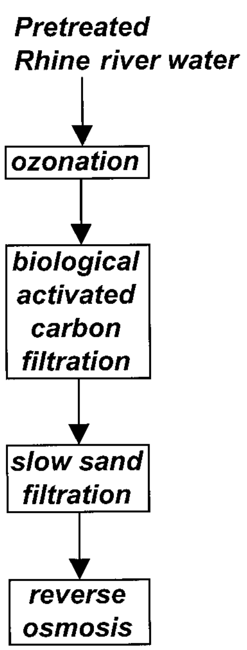

Figure 2 | Process scheme pilot plant B, the integrated membrane system.

Table 2 | Process conditions pilot plant B

Ozonation	Ozone dose	0.8 mg l^{-1}
	Contact time	18–30 min
Biological activated carbon filtration	EBCT	40–70 min
	Bed height	2.5 m
	Carbon type	Norit ROW 0.8S
Slow sand filtration	Filtration rate	0.2–0.5 m h^{-1}
	Bed height	0.8–1.2 m
	Sand size	0.2–1.0 mm

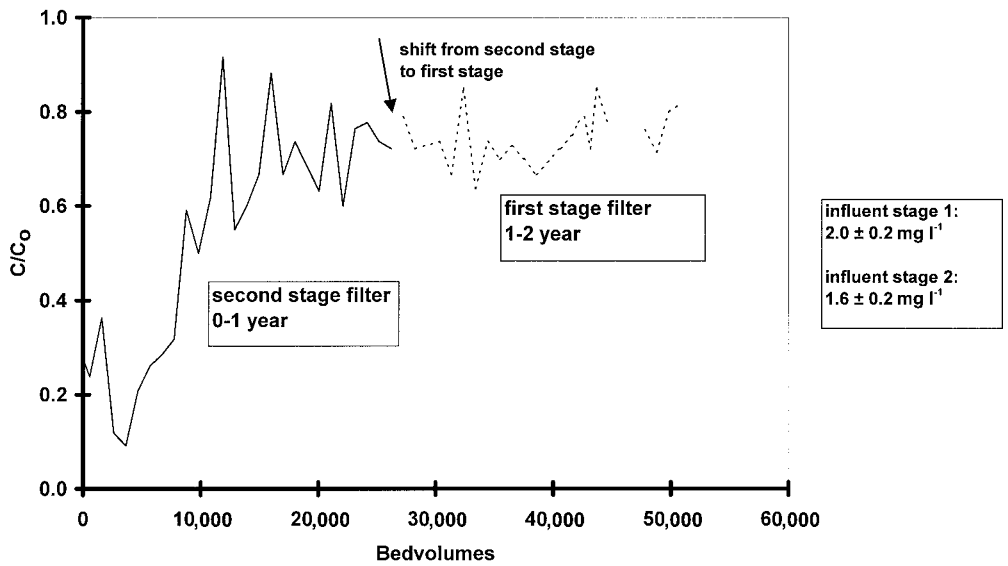

Figure 3 | DOC break-through profile of a single carbon filter in the Leiduin full-scale production plant.

liquid–liquid extraction with ethylacetate, and gas chromatography with nitrogen-phosphorus detection. The concentration of atrazine is determined by comparing the ratio between the area of the atrazine peak and the peak of an internal standard (phosphamidone) in the sample analysed and in the standard with known atrazine concentration. For the analysis of diuron and isoproturon the filtered water is led through an adsorbence column. After a clean-up to remove the humic fraction, this column is connected with the eluent stream of a HPLC, by which the adsorbed components are eluated on the analytical column where they are separated. Identification and quantification is carried out with a UV-spectrometer/PDA combination (photodiode array detection). Identification is based on retention times related to the internal standards fenuron and chloroxuron. Quantification is based on an external standard method in relation to a calibration line of the standard that went through the same procedure (Amsterdam Water Supply 1998).

Assimilable organic carbon (AOC) was determined according to the method described by van der Kooij (1992). The biofilm formation rate (BFR) was determined with a biofilm monitor as described by van der Kooij *et al.* (1997). The filtration characteristics and fouling characteristics of the water were measured using the modified fouling index (Schippers & Verdouw 1980). All other analyses were performed according to the standard methods as applied by Amsterdam Water Supply (1998).

RESULTS AND DISCUSSION

Benefits of BACF in the existing treatment scheme

Removal of organics

Figure 3 shows a typical break-through profile of a carbon filter over a total period of 2 years (52,560 bedvolumes). A filter starts as a second stage filter and gradually shows a DOC break-through of 70% after a period of 1 year. Then the filter is switched to the first stage and still shows a constant DOC removal efficiency of about 20–30% ($C/C_o = 0.7–0.8$). The constant DOC removal of the filters in the first stage implies biodegradation of organic material in these first stage filters (Cauchi *et al.* 1993).

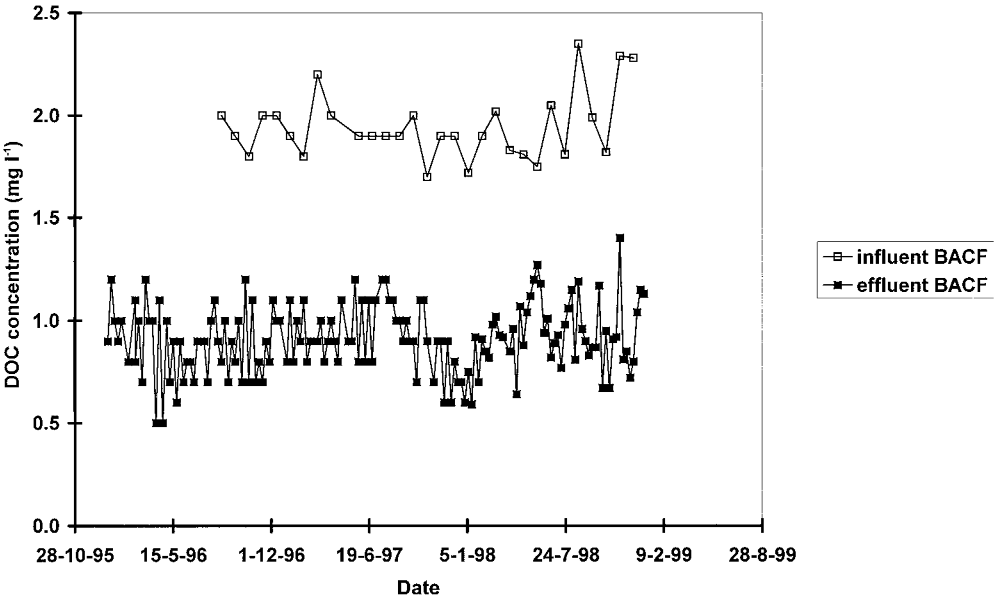

Figure 4 | DOC concentration after biological activated carbon filtration in the full-scale Leiduin plant 1996–1998.

The 20 filters in the second stage of the full-scale plant are reactivated in sequence and thus have a running time varying from 0 to 1 year. As a consequence, the mixed filtrate of these second stage filters has a DOC content of about 1 mg l^{-1}. Figure 4 shows the concentration over the period 1996–1998.

As mentioned in the Introduction, one of the main reasons for introducing BACF to the treatment scheme of the Leiduin production plant was to create a robust barrier for pesticides, besides DOC reduction and AOX reduction. Therefore, at pilot-plant scale, several spiking experiments were carried out with mixtures of pesticides, ranging in concentration from 5 to 50 μg l^{-1}, and a dosing period ranging from 1 to 28 days. After a 3-year operational period for the carbon filters of pilot plant A, without any carbon reactivation, a mixture of atrazine, diuron and isoproturon was dosed in the feed water of the carbon filters. To avoid chemical oxidation of the pesticides during ozonation, the pesticides were dosed after the ozonation section, each in a concentration of 5 μg l^{-1}. This spiking experiment lasted for 696 h (29 days). Figure 5 shows the removal efficiencies over the first stage and second stage carbon filters of pilot plant A.

All three pesticides were completely removed in the carbon filters. Only atrazine showed a little break-through in the first stage filters, but the residual atrazine was completely removed in the second stage filters. During the whole spiking period (696 h) the filters performed very well. The pilot plant carbon filters, showing a 80% DOC break-through after a lifetime of 3 years without reactivation, still showed an excellent pesticide removal efficiency, exceeding 99%.

From the literature it is known that pesticides are removed by biodegradation in carbon filters (Huang & Banks 1996). Two additional experiments showed that in this specific case biodegradation might also have contributed to pesticide removal. Figure 6 shows the results of a 2-day atrazine spiking experiment of a carbon filter which had already been in operation for 36,000 BV (EBCT 40 min). In this experiment atrazine was dosed preceding the ozonation at a concentration of 22 μg l^{-1}. After ozonation the atrazine concentration was 16 μg l^{-1}, while desisopropylatrazine and desethylatrazine were formed at a concentration of 0.7 μg l^{-1} and 2.4 μg l^{-1} respectively. Besides atrazine, the AOC concentration and DOC concentration over the filter bed height were also measured.

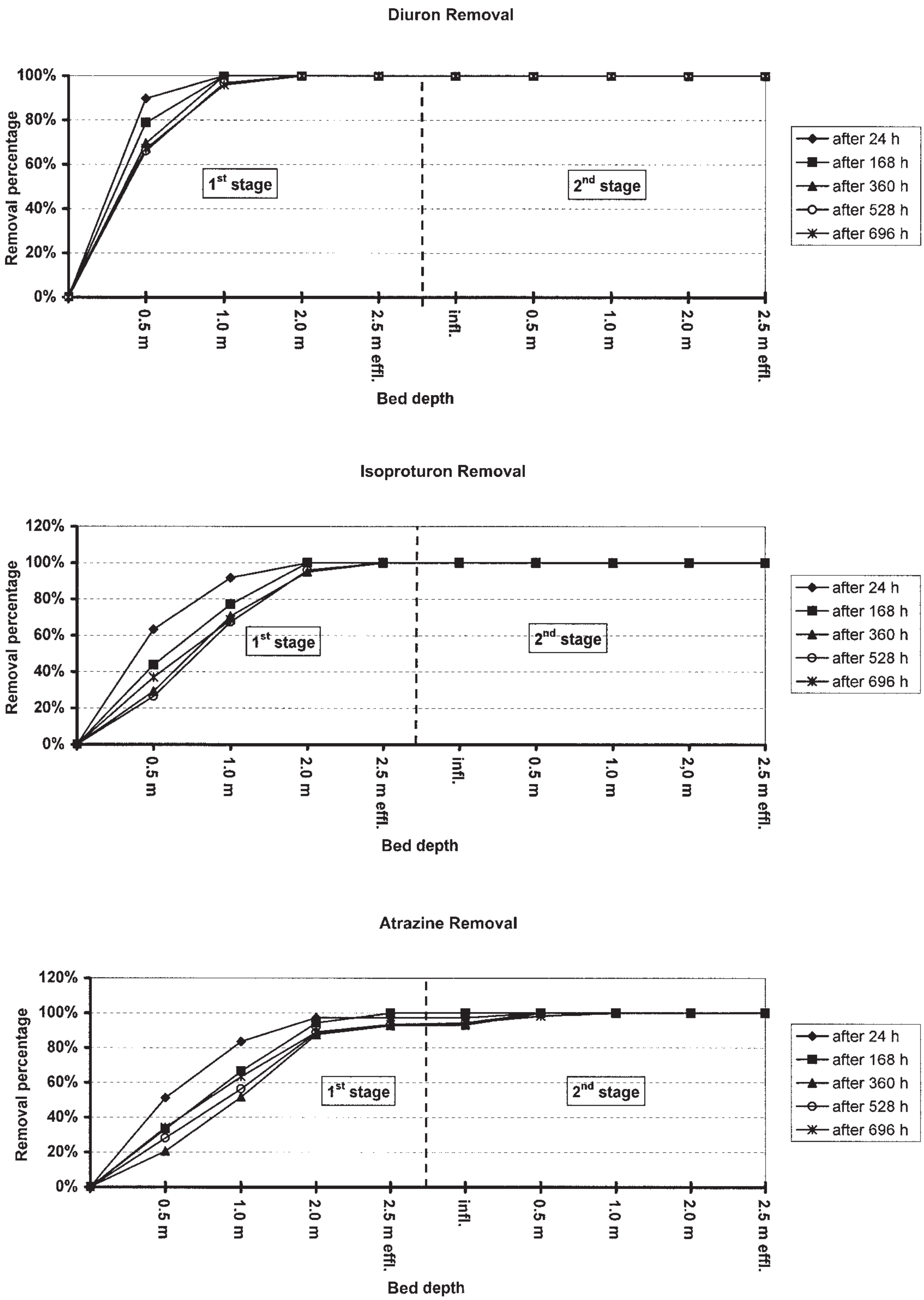

Figure 5 | Removal of diuron, isoproturon and atrazine in a two-stage activated carbon filtration process during a 29-day spiking experiment after a carbon lifetime of 3 years.

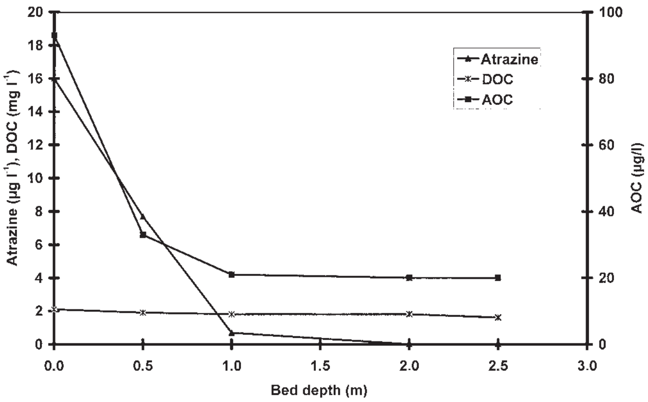

Figure 6 | Removal of atrazine, DOC and AOC over the filter bed height during a 2-day spiking experiment, using carbon with a lifetime of 36,000 BV (influent DOC 2.1 mg l^{-1}; preceding ozone dose 0.8 mg l^{-1}).

The AOC removal took place in the upper part of the filter and coincided with the atrazine removal, while the DOC removal was only 20% (from 2.1 mg l^{-1} to 1.7 mg l^{-1}). Atrazine and atrazine degradation products were below the detection limits in the effluent of the carbon filter. As AOC is removed mainly by biological processes, the biologically active part is in the upper part of the filter. This indicates that atrazine removal, located in the upper part of the filter, might also have taken place through biodegradation.

In the second additional experiment, the effect of doubling the EBCT on the increase of maximum running time for pesticide removal was determined. The maximum running time is defined as the running time until the effluent concentration exceeds 1 µg l^{-1} with a feed concentration of 5 µg l^{-1} (break-through 20%). This experiment was carried out with carbon that had already been used for more than 3 years (47,000 BV, influent DOC concentration 2 mg l^{-1}). In one carbon filter, biological activity was excluded by dosing sodium azide in the influent. Therefore, in this filter, only adsorption of the pesticides could take place. In the other carbon filter, both adsorption and biodegradation were allowed to take place. Table 3 shows the results.

For all pesticides, in the filter in which no biocide (sodium azide) was dosed (filter 2) the doubling of the contact time showed a much greater effect on the maximum running time as compared with the filter in which only adsorption took place (filter 1). This strongly suggests that biodegradation of pesticides plays an important role in biologically active carbon filters.

Table 3 | Effect of doubling the EBCT on the increase in running time

	Increase in maximum running time (multiplier)	
Pesticide	**Filter 1: adsorption**	**Filter 2: adsorption and biodegradation**
Atrazine	2.1–3.6	3.0–4.0
Bentazone	1.5–2.0	6.0
Metribuzine	2.0–3.0	> 17.0
Pirimicarb	2.0–4.0	3.0–4.0

Disinfection capacity

Before the introduction of ozone and BACF, chlorine was occasionally dosed in the finished water of the Leiduin plant to maintain the microbiological quality of the drinking water during distribution. This was necessary only in winter times during periods of frost. During such periods, the hygienic load of the Leiduin production plant is high due to the fact that in the open collecting basin after the dune passage, many birds gather on the only open, unfrozen water available in the winter. In addition, at very low water temperatures (<5°C) the slow sand filters have a lower disinfection capacity. To ensure the hygienic quality of the water leaving the plant, a low chlorine dose of 0.6 mg l^{-1} was applied, resulting in a residual concentration of 0.2 mg l^{-1} after 20 min contact time.

The first winter during which the ozonation was in operation (1995–1996) was characterized by a very long frost period. Therefore, it could be tested whether the presence of an ozonation step in the process scheme resulted in an increased disinfection capacity, making the use of chlorine unnecessary, even under extreme conditions. During this winter the ozonation was operated with an ozone dose of 0.9 mg l^{-1}, resulting in a contact time (CT) value of 2 mg · min l^{-1}. Tables 4 and 5 show the results of some specific analyses during this period.

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Table 4 | Coliforms 44°C, fecal streptococci and spores of sulphite reducing clostridia in the rapid sand filtrate and in the finished water (1 December 1995–20 March 1996)

	Rapid sand filtrate				Finished water				
	p10	p50	p90	N	p10	p50	p90	N	MAC*
Coliforms 44°C (no. 100 ml^{-1})	0.8	13	60	147	< 0.1	< 0.1	0.2	93	< 0.3
Fecal streptococci (no. 100 ml^{-1})	1.0	15	120	118	< 0.2	0.3	0.6	12	< 1
Clostridia (no. 100 ml^{-1})	0.5	1.0	2.4	22	< 0.1	0.2	0.4	20	< 1

*Maximum admissible concentration according to Dutch legislation.

Table 5 | Campylobacter (no. 100 ml^{-1}) in the raw water after dune passage, in the water after rapid sand filtration and in the water after ozonation

Date	After dune passage	After rapid sand filtration	After ozonation
11 December 1995	24	7.5	< 0.03
8 January 1996	240	15	< 0.03
12 February 1996	150	4.6	< 0.03

Coliforms, fecal streptococci and clostridia were measured in the rapid sand filtrate, and in the finished water. The concentrations in the finished water were all below the maximum admissible concentration (MAC) and no chlorine was necessary to comply with the regulations. On three occasions the number of campylobacter was also measured in the rapid sand filtrate and in the water after ozonation. From these values a logarithmic reduction capacity of the ozonation can be estimated of at least 2.2 units.

Hence, due to the introduction of ozonation, the Leiduin production plant produces hygienically safe water without the use of a chlorine residual in the drinking water during distribution, also during periods with a low water temperature and a high microbial load.

Process stability of the slow sand filters

Slow sand filtration, in combination with ozone-activated carbon filtration (BACF), is one of the important process steps in the Leiduin plant, which enables the production of hygienically safe and biologically stable water without the use of a disinfectant residual. The use of covered slow sand filters already contributes to stable performance, as sunlight is excluded and freezing of the water surface in winter is prevented. However, the introduction of BACF showed an important increase in the running time of the slow sand filters between successive scraping procedures. Scraping of the slow sand filters is necessary when the maximum headloss is reached and involves the removal of the upper 2–3 cm of the sandbed (the so-called 'Schmutzdecke' or biological mat) to reduce the headloss. Figure 7 shows the running times of the slow sand filters before the introduction of BACF in the period 1985–1994. About 50% of the running times did not exceed 100 days. In the old process configuration, only one filtration step, the rapid sand filtration, preceded the slow sand filters, and the turbidity of the feed water of the slow sand filters was 0.39 ± 0.21 NTU.

Introduction of the ozone-activated carbon filtration resulted in three filtration steps preceding the slow sand filters: the rapid sand filtration and the two-stage carbon filtration. The turbidity of the feed water of the slow sand filters decreased to 0.11 ± 0.04 NTU and resulted in a dramatic increase in the running time. Figure 8 shows the running times in the new process configuration as compared with the old process configuration. In the initial 1.5 years after the introduction of BACF, running times were kept shorter than approximately 500 days, even though the maximum headloss had not yet been reached, because it was not yet certain whether longer running

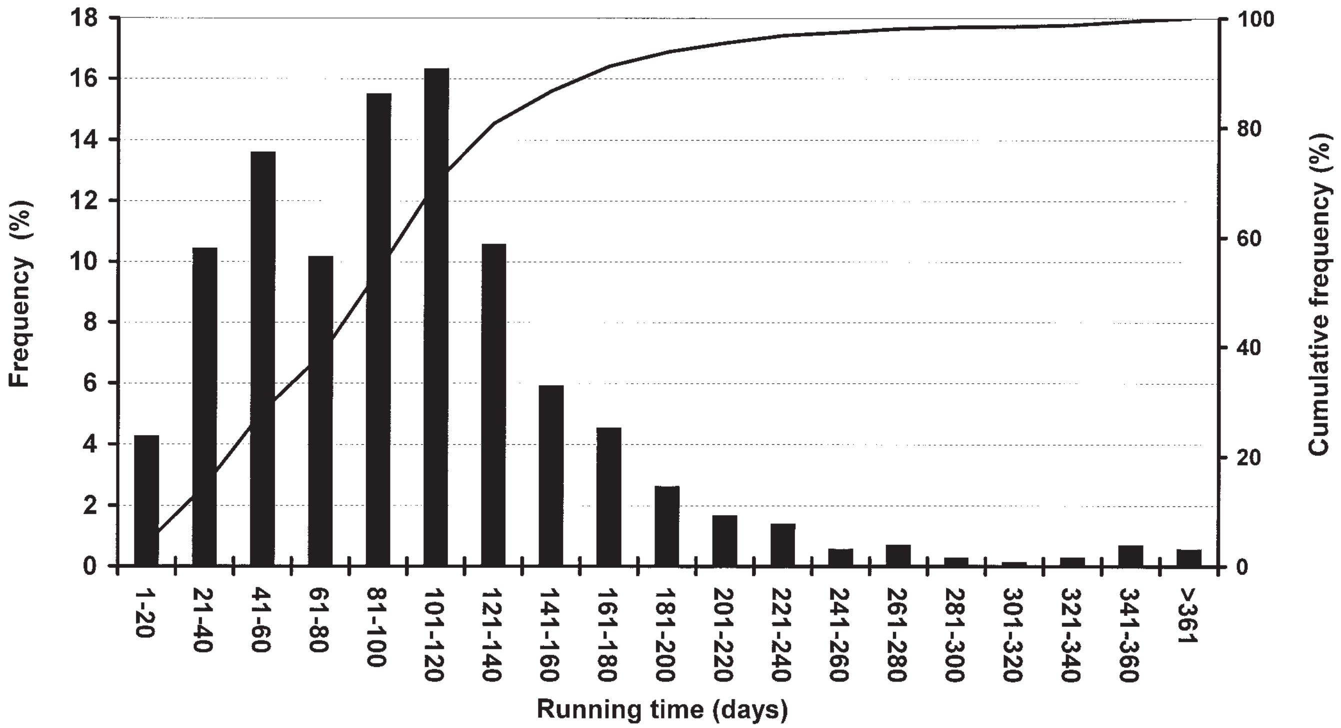

Figure 7 | Running time of the slow sand filters of Leiduin production plant before the introduction of BACF (1985–1994).

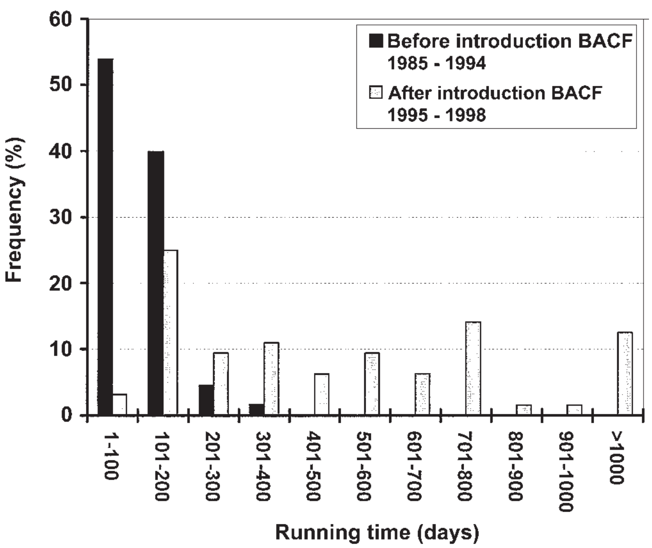

Figure 8 | Running time of the slow sand filters of Leiduin production plant before and after the introduction of BACF.

times would have a negative effect on the effluent quality. After it became clear that extremely long running times did not result in a deterioration of the effluent quality nor in a deterioration of the filter performance, long running times were accepted and nowadays all slow sand filters operate with a running time of at least 2 years. Hence, introduction of BACF has resulted in the very stable operation and reliable performance of the slow sand filters.

Biological stability of the finished water

As discussed in the previous section, ozonation contributes significantly to the final disinfection capacity of the Leiduin production plant. However, during ozonation easily biodegradable organic material is produced and this may reduce the biological stability of the finished

Table 6 | Biological stability of the finished water from Leiduin production plant before and after introduction of BACF (average±standard deviation, *n*=number of determinations)

	Finished water quality	
	Before introduction of BACF	After introduction of BACF
AOC ($\mu g\ l^{-1}$)	5.1 ± 1.1 ($n = 8$)	5.4 ± 1.7 ($n = 20$)
BFR (pg ATP $cm^{-2} \cdot day^{-1}$)	0.25 ± 0.03 ($n = 16$)	0.84 ± 0.14[1] ($n = 8$)
		0.25 ± 0.03[2] ($n = 9$)

[1]March 1997–August 1997.
[2]September 1997–January 1998.

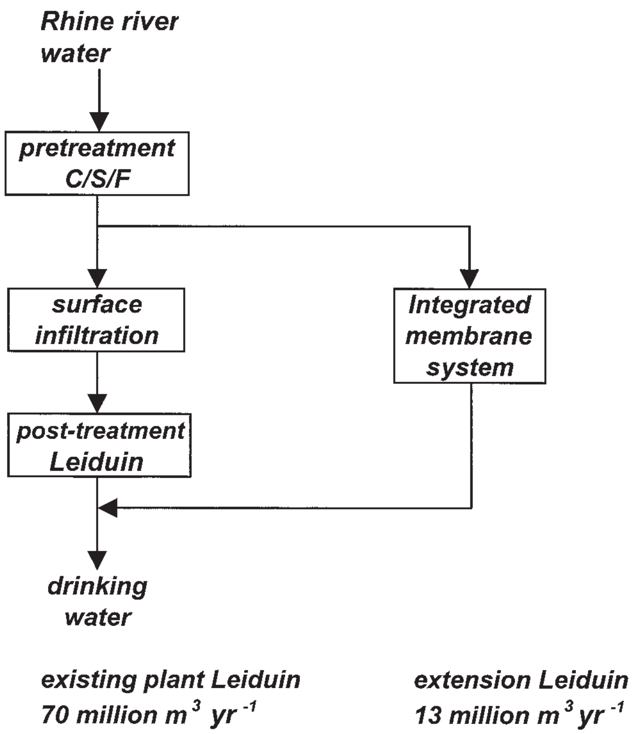

Figure 9 | Extension of the Leiduin plant with an integrated membrane system.

water. With an ozone dose as applied in the Leiduin plant, the AOC concentration increases up to 60–100 $\mu g\ l^{-1}$ (van der Hoek *et al.* 1995). This has to be removed in the subsequent filtration steps, namely the two-stage carbon filtration and the slow sand filtration. In the treatment philosophy of AWS, safety disinfection has to be achieved by nutrient removal, preventing regrowth of bacteria during transport and distribution. Production of biologically stable water is a prerequisite in this approach.

Therefore, the biological stability of the finished water was judged by measuring the AOC concentration and the biofilm formation rate (BFR). Water is assumed to be biologically stable when the AOC concentration is below 10 $\mu g\ l^{-1}$ (van der Kooij 1992) and the BFR does not exceed 1 pg ATP $cm^{-2} \cdot day^{-1}$ (van der Kooij *et al.* 1995*a*, *b*). The risk of exceeding the guideline value for aeromonads (200 cfu 100 ml^{-1}) is ≤20% at BFR values ≤10 pg ATP $cm^{-2} \cdot day^{-1}$ (van der Kooij *et al.* 1999). From Table 6 it can be concluded that both before and after the introduction of BACF the finished water can be regarded as biologically stable, using the criteria as defined above. The BFR remains very low compared with values that are normally observed in drinking water in the Netherlands (1–60 pg ATP $cm^{-2} \cdot day^{-1}$; van der Kooij *et al.* 1995*b*). Hence, the increased concentration of biodegradable organic material resulting from the ozonation is very effectively removed in the filtration steps, following the ozonation. The carbon filters play an important role in this removal. Thus, biological filtration processes as BACF still offer the possibility to produce biologically stable water.

Table 7 | Reverse osmosis fouling: key parameters, guide lines and pretreatment results (average±standard deviation, n=number of determinations)

Fouling aspect	Key parameter	Guideline	Result pretreatment O_3-BACF-SSF
Particulate/colloidal fouling	MFI (s l^{-2})	<1	0.69 ± 0.25 ($n = 42$)
Organic fouling	DOC (mg l^{-1})	<2	1.4 ± 0.3 ($n = 25$)
Biofouling	AOC (μg l^{-1})	<10	5.7 ± 2.5 ($n = 7$)
	BFR (pg ATP cm^{-2}·day^{-1})	<1	0.84 ± 0.14 ($n = 8$)

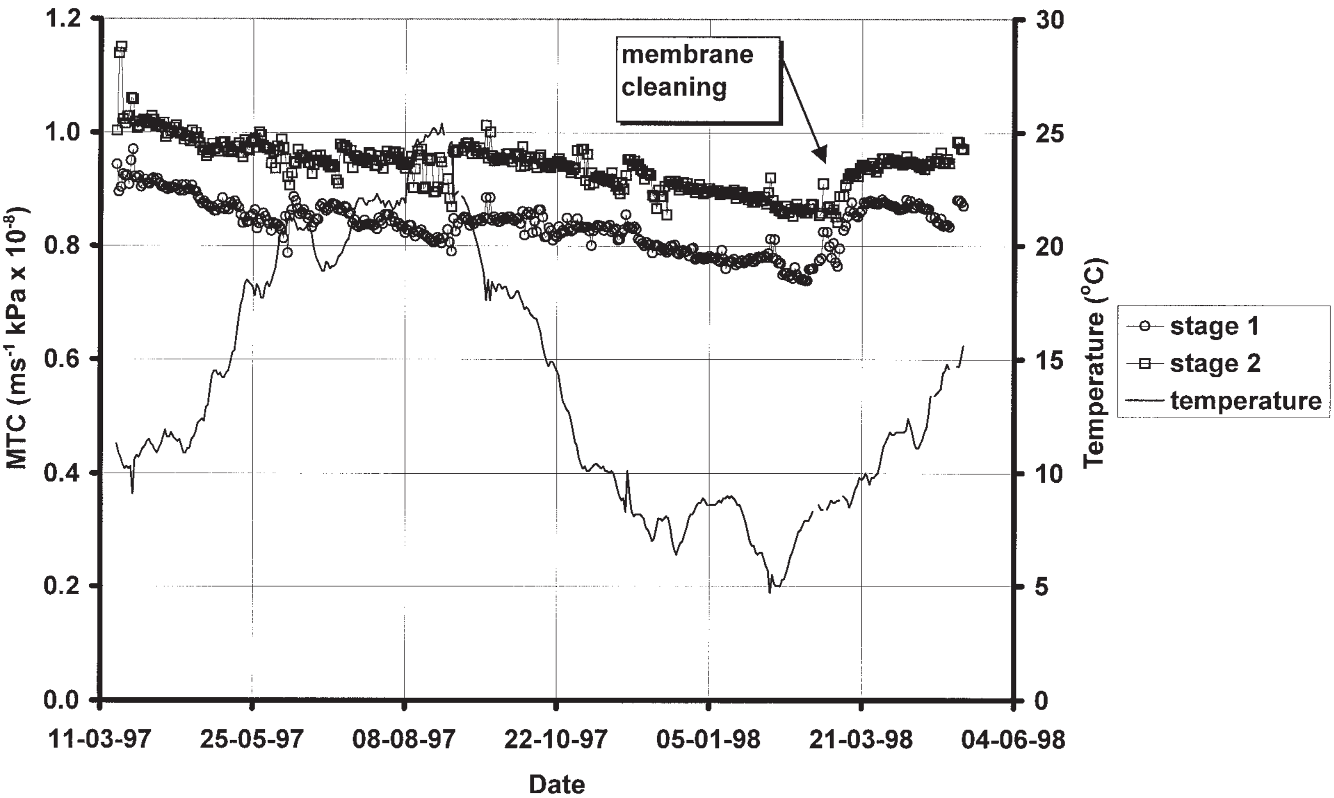

Figure 10 | Mass transfer coefficient of a reverse osmosis membrane unit, using ozone-activated carbon filtration and slow sand filtration as pretreatment.

Benefits of BACF in integrated membrane systems

Integrated membrane systems using reverse osmosis

As mentioned in the Introduction, process schemes have been developed and studied at pilot-plant scale for the extension of the capacity of the Leiduin production plant. One alternative concerns the direct treatment of pretreated Rhine river water without soil passage, using an integrated membrane system. In this alternative, 70 million m^3 yr^{-1} will be produced with the existing plant and 13 million m^3 yr^{-1} will be produced with the integrated membrane system. Figure 9 shows this combination of the existing plant and the integrated membrane system and an effective set-up of the integrated membrane system is shown in Figure 2.

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In the existing plant, the ozonation, carbon filters and slow sand filters already have a hydraulic capacity of 83 million $m^3\ yr^{-1}$, so a part of these units (13 million $m^3\ yr^{-1}$) will be separated from the existing plant and used in the integrated membrane system. They act as pretreatment for the final stage, which is a reverse osmosis unit. This pretreatment has proven to result in a very stable operation of the reverse osmosis system, as fouling of the membrane elements is prevented by the high quality of the feed water produced by the pretreatment. Table 7 shows three types of fouling and the key parameters which govern the fouling characteristics of the feed water, including the values below which no severe fouling will occur (van der Hoek *et al.* 1997, 1999). The actual concentrations of the feed water as produced by the pretreatment scheme comprising ozonation, BACF and slow sand filtration are below the guidelines.

The benefits of the use of ozone-activated carbon filtration in combination with slow sand filtration as pretreatment for reverse osmosis in an integrated membrane system are even more pronounced in Figure 10. The figure shows the mass transfer coefficient (MTC), a standard for the productivity of reverse osmosis membranes. Using ozone-activated carbon filtration and slow sand filtration as pretreatment, the MTC only showed a decrease of 16% over a whole year, without any chemical cleaning of the membrane elements. As compared with literature data (Harfst 1994) this long period without chemical cleaning, while maintaining the productivity at a high value, can be regarded as an excellent performance of the pretreatment and it results in a very stable and reliable operation of the reverse osmosis system.

Hence, in integrated membrane systems, BACF has also proven its effectiveness, as it reduces the modified fouling index (MFI), DOC, AOC and BFR content of the feed water to such low values that operation of the reverse osmosis system is not hampered by colloidal fouling, organic fouling and biofouling.

Additional perspectives of BACF in integrated membrane systems

In the integrated membrane system using reverse osmosis as developed for the extension of the Leiduin plant, the membrane system originally had to fulfil four functions: desalination, softening, removal of pesticides and disinfection. The use of ozonation and BACF in the integrated membrane system reduces the number of functions the membrane system has to fulfil, as ozone-activated carbon filtration already offers a reliable barrier for pesticides and already offers a high disinfection capacity.

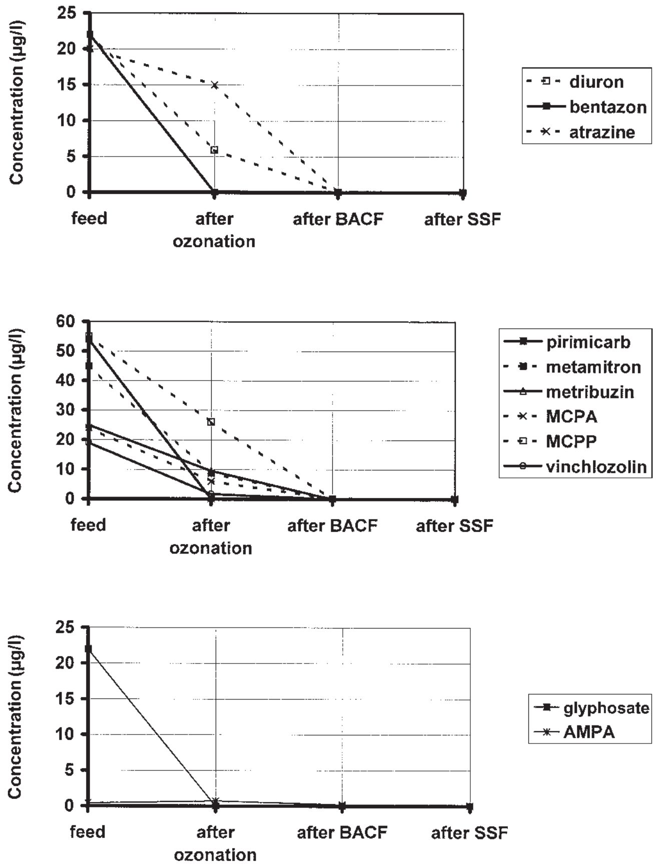

Figure 11 | Removal of pesticides during a 3-day spiking experiment by ozonation, activated carbon filtration and slow sand filtration in the integrated membrane system.

The removal of pesticides by ozonation, BACF and slow sand filtration is shown in Figure 11. Even at very high peak loadings the pesticides are removed below the maximum admissible concentration. The pesticide spiking in this experiment lasted for 3 days, after the carbon filters had already been in operation for 3 years and several previous spiking experiments had been carried out.

From an evaluation of the hygienic quality of the pretreated Rhine river water, it can be calculated that

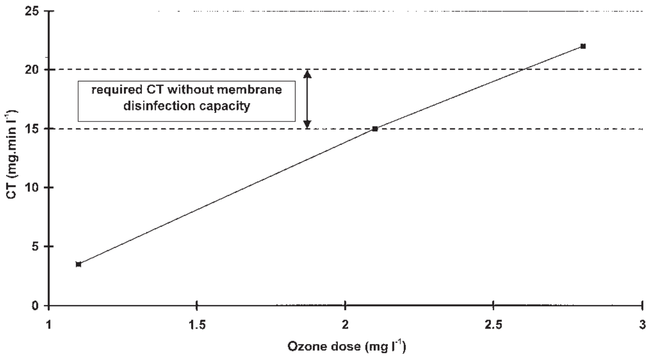

Figure 12 | Relation between ozone dose and CT value for the pretreated Rhine river water (DOC 2 mg l^{-1}; Br^- 100–200 μg l^{-1}).

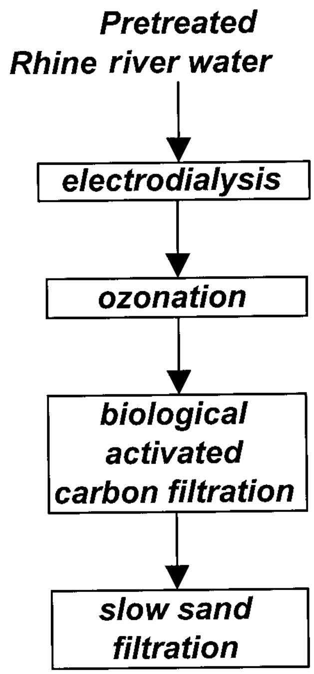

Figure 13 | An integrated membrane system comprising electrodialysis as alternative for reverse osmosis.

the ozonation has to result in a CT value of at least 15–20 mg · min l^{-1} to create sufficient final disinfection capacity in the integrated membrane system, when the membrane unit itself does not have disinfection capacity and does not contribute to the disinfection capacity of the integrated membrane system (Graveland & van der Hoek 1997; van der Hoek *et al.* 1998). Due to the relatively low DOC concentration (average value 2 mg l^{-1}) this required CT value can already be reached with an ozone dose of 2–2.5 mg l^{-1}, as shown in Figure 12. Final disinfection is still achieved through nutrient removal by the carbon filtration and slow sand filtration.

So, in the integrated membrane system, the removal of pesticides and disinfection can be guaranteed by ozonation, BACF and slow sand filtration, leaving only the functions desalination and softening to be fulfilled by the membrane process. In that case electrodialysis becomes an alternative for reverse osmosis, and a second integrated membrane system comprising electrodialysis instead of reverse osmosis becomes a realistic option. Figure 13 shows this integrated membrane system. As electrodialysis is less sensitive to fouling it can be used as first process unit. With this sequence electrodialysis removes bromide to such a low concentration, that bromate production during ozonation is not a problem and the product water is in compliance with the Dutch bromate standard of 5 μg l^{-1} (van der Hoek *et al.* 1998). Figure 14 shows the desalination characteristics of a two-stage and three-stage electrodialysis system as compared with a three-stage reverse osmosis system (van der Hoek *et al.* 2000).

Thus, by applying ozonation and BACF in integrated membrane systems the use of electrodialysis as an alternative for reverse osmosis becomes a viable option and offers the possibility of making a more deliberate choice of the type of membrane process in the integrated membrane system.

Costs of BACF

In 1997, the drinking water production costs of the Leiduin plant were 0.97 Dutch guilders. These costs include operation and maintenance, and interest and depreciation. The costs of ozonation were 0.03 Dutch guilders and the costs of the carbon filtration were 0.11 Dutch guilders. BACF accounts for only 14% of the total production costs and thus can be regarded as a very cost-effective process related to the many benefits the process offers.

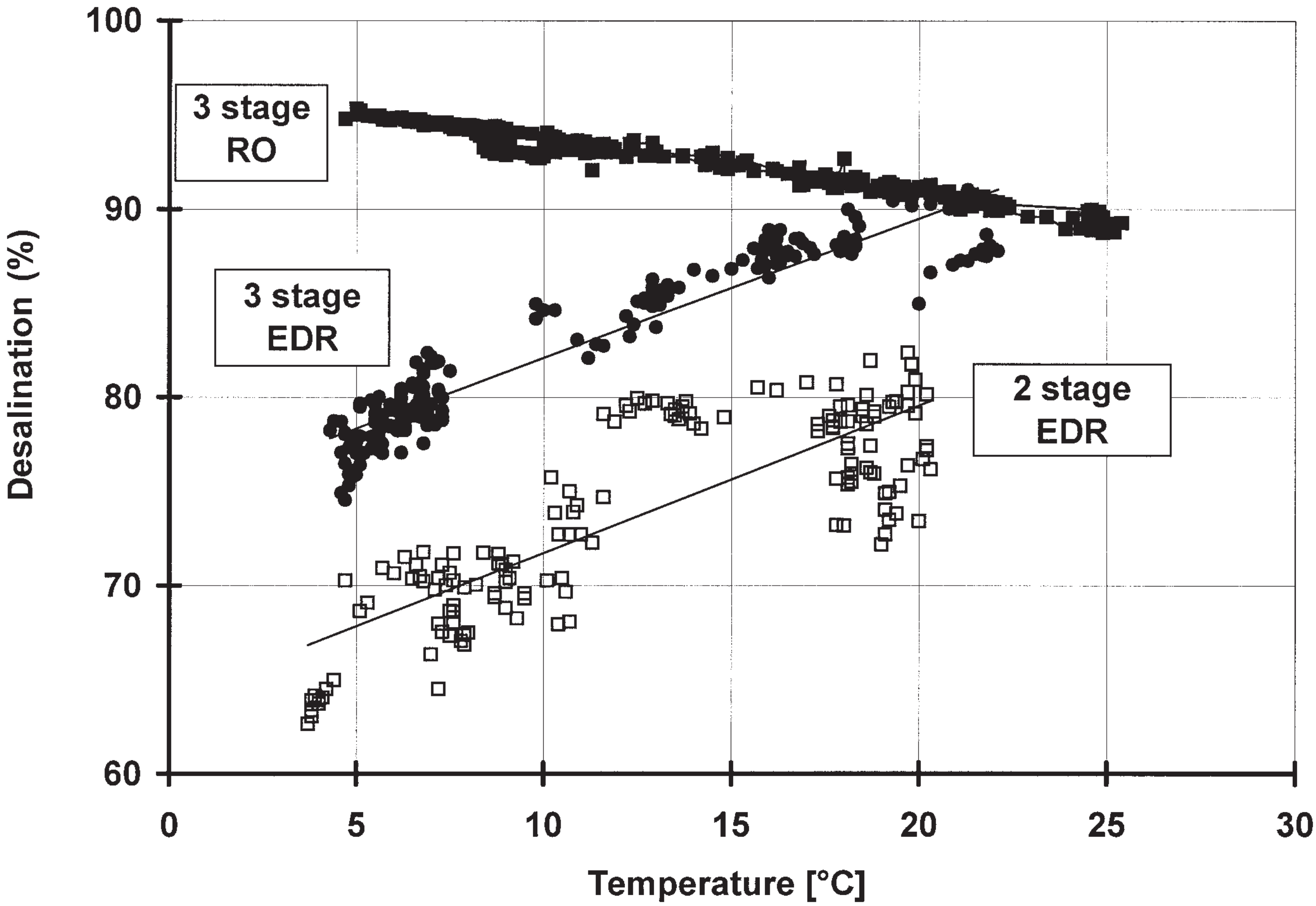

Figure 14 | Desalination characteristics of two stage electrodialysis, three stage electrodialysis, and three stage reverse osmosis.

CONCLUSIONS

From full-scale experience and pilot plant experiments it can be concluded that ozone-activated carbon filtration is a versatile process which has many benefits in integrated treatment schemes.

In the drinking water treatment plant Leiduin in which ozone-activated carbon filtration was introduced in 1995, it has shown to be a very effective process for the removal of organics in general and the removal of pesticides in particular. Due to the preceding ozonation the carbon filtration acts as a biological filtration process, resulting in adsorption and biodegradation of organics with an accompanying long lifetime of the carbon filters.

In the Leiduin production plant, typical additional benefits of ozone-activated carbon filtration are an increased disinfection capacity and a very stable operation of the slow sand filters following BACF. The increased disinfection capacity and the effective removal of biodegradable organic material make it possible to produce hygienically safe and biologically stable drinking water without the use of chlorine.

Also, in integrated membrane systems ozone-activated carbon filtration can play an important role. When it is applied as pretreatment for reverse osmosis, the feed water quality of the reverse osmosis system is of such a high level that a very stable operation of the system can be obtained. Due to its disinfection capacity and its effective removal of pesticides, other membrane processes may become attractive in integrated membrane systems. In the extension scheme for the Leiduin plant, ozone-activated carbon filtration creates possibilities for electrodialysis as an alternative for reverse osmosis.

In summary it is concluded that ozone-activated carbon filtration, applied in integrated treatment processes, does not only result in disinfection and a very effective and efficient removal of organics, but also in several other benefits, which make this process very attractive for incorporation into drinking water treatment systems.

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