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Impact of summer droughts on the water quality of the Meuse river

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Summary Climate change will probably increase the frequency and intensity of low river flows, affecting both water quantity and water quality. Although climate change impacts on water quantity are widely recognised, the impacts on water quality are less known. The aim of this paper is to assess the effects of droughts on the water quality of the river Meuse in western Europe, based on analysis of existing water quality data. Time series of water quality were investigated at two monitoring stations during two severe drought periods, occurring in the years 1976 and 2003. Water quality during these droughts was investigated and compared to water quality during reference periods, representing common hydrological conditions and similar chemical pollution. A total amount of 24 water quality parameters were involved in the analysis, which can be divided into four groups: (1) general water quality variables (water temperature, chlorophyll-*a*, pH, dissolved oxygen and suspended solids), (2) nutrients, (3) major elements (e.g. chloride, fluoride) and (4) heavy metals and metalloids. To assess the effects of changes in discharge and water temperature on the concentration of chemical substances, empirical relations have been established between concentration and discharge, and between concentration and water temperature. The results indicate a general deterioration of the water quality of the Meuse river during droughts, with respect to water temperature, eutrophication, major elements, and some heavy metals and metalloids. This decline in water quality is primarily caused by favourable conditions for the development of algae blooms (high water temperatures, long residence times, high nutrient concentrations) and a reduction of the dilution capacity of point source effluents. © 2008 Published by Elsevier B.V.

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Introduction

Global and regional scale studies indicate that climate change may result in increases in frequency and intensity of droughts in many river basins (e.g. Shabalova et al.,

2003; Arnell, 1999; Kothavala, 1999; Gregory et al., 1997). In Europe, a mean annual air temperature increase of 2.2–5.3 °C is projected for the next century (Christensen et al., 2007) and a decrease in summer precipitation is foreseen for large parts of Europe (EEA, 2004). Additionally, mean and maximum summer temperatures are expected to increase and a continuing increase in precipitation extremes is expected as well (Frei et al., 2006; EEA, 2004; Booi, 2002; Gregory and Mitchell, 1995). It is likely that the frequency of heat waves and droughts in Europe will increase in the future (EEA, 2007; Meehl and Tebaldi, 2004; Stott et al., 2004).

The impact of climate change on hydrology has been studied widely, with a clear focus on water quantity (e.g. Pfister et al., 2004; Middelkoop et al., 2001; Gellens and Roulin, 1998). Recently, also the potential effects of climate change on surface water quality has been increasingly acknowledged (e.g. Murdoch et al., 2000). Several methods have been proposed to assess potential changes in water quality due to climate change. Examples include empirical relations between water quality and climatic trends (Fukushima et al., 2000; Schindler et al., 1996; Williams et al., 1996), and black-box or deterministic modelling approaches to assess potential effects of climate change on surface water quality at continental or regional scale (Krysanova et al., 2004; Mimikou et al., 2000; Clair and Ehrman, 1996; Wolford and Bales, 1996). These studies have indicated that water quality can be directly affected by several climate-related mechanisms on both short and long term. These include effects of air temperature increase, as well as changes in hydrological factors (e.g. limited dilution of point source emissions during low river flows), terrestrial factors (e.g. changes in vegetation and soil structure) and resource-use factors (e.g. increased water use, increased demand for cooling water) (Murdoch et al., 2000). Still, few studies have been reported that assessed the impact of droughts and inherent low-flow conditions on river water quality (Caruso, 2002). Since river water quality may deteriorate to critical values during periods of prolonged low-flow conditions in combination with high water temperatures (e.g. Somville and De Pauw, 1982), insight and understanding of the impact of droughts on water quality is essential, especially for rivers which are highly sensitive to drought conditions.

The river Meuse in Western Europe is a rain-fed river that is characterized by a highly variable discharge regime with commonly low discharges during summer and autumn, and that is highly sensitive to droughts (Berger, 1992). During droughts, important river functions with respect to water quantity are hampered, such as water availability for agriculture and release of cooling water by power plants (Tu et al., 2005). Furthermore, water is extracted for domestic use and drinking water supply of 6 million inhabitants of Belgium and the Netherlands, while at the same time the Meuse has a high recreational and ecological potential (Voltz et al., 2002). These functions depend on the water quality of the Meuse river, which is related to river flow and water temperature. The aim of this paper is to evaluate the impact of droughts on the water quality of the river Meuse. We based our approach on analysis of water quality data of two severe drought periods, occurring in 1976 and 2003. Water quality during these droughts was compared

to water quality under common hydrological regimes (with similar chemical pollution) at two monitoring stations in the lower part of the Meuse. In order to assess the effects of climate change on water quality, concentrations of chemical substances were related to discharge and water temperature.

The Meuse basin

The total length of the river Meuse is 935 km from its source in Pouilly-en-Bassigny on the plateau of Langres in France to the mouth of the Haringvliet in the Netherlands (Middelkoop and Van Haselen, 1999) (Fig. 1). The drainage basin covers an area of 34,548 km², including parts of France (26% of surface area), Luxembourg (<1%), Belgium (41%), Germany (11%) and the Netherlands (22%) (International Meuse Commission, 2005). More than 8.8 million people are living in the entire catchment area. The Meuse basin upstream of the Belgium–Dutch border comprises 34% agricultural land,

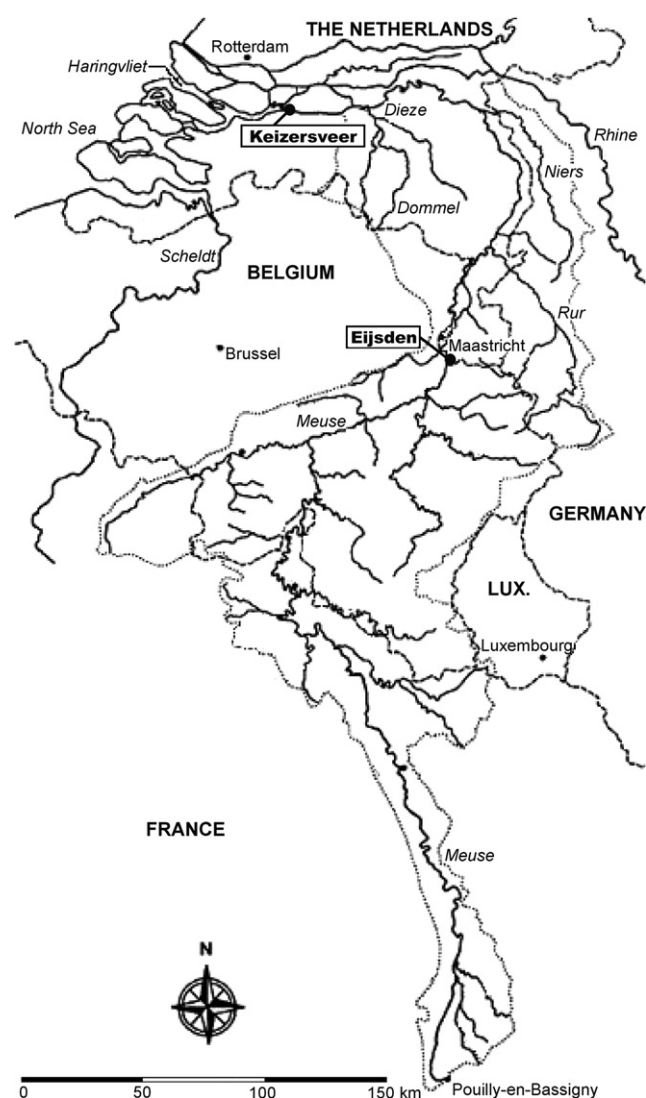


Figure 1 The Meuse basin and locations of measurement stations Eijsden and Keizersveer.

20% as pasture, 35% as forested and 9% as built-up area (De Wit et al., 2001).

The regime of the Meuse is controlled by rainfall—evapotranspiration, and mainly corresponds with the seasonal variation in net precipitation surplus. The average annual discharge at the Belgium–Dutch border amounts to $265 \text{ m}^3 \text{ s}^{-1}$, with extremes of less than $10 \text{ m}^3 \text{ s}^{-1}$ during droughts and more than $2500 \text{ m}^3 \text{ s}^{-1}$ during floods. Droughts can last from a few weeks up to half a year. During periods of low flow, weirs are operated to maintain a minimum water level for shipping (Berger, 1992). This results in very long residence times with almost stagnant flow conditions of the Meuse in the southern part of the Netherlands. The main tributaries in the lower part are the Rur and Niers, which are located in the region Düsseldorf and Köln, and the Dieze in the province of Brabant (in the Netherlands). During droughts, the contribution of these tributaries and groundwater inflow to the total discharge of the Meuse river is relatively high. De Wit et al. (2001) conclude that the discharge of the Meuse is more variable compared to that of other rain-fed rivers. This implies that the Meuse has a relatively fast response to precipitation and is therefore relatively sensitive to both flooding and drought.

The water quality of the Meuse has been changing in the last fifty years. In the 1960s, water quality of the Meuse deteriorated, and the river was most heavily polluted around 1970. Due to construction of waste water treatment plants, technological innovations in industrial processes, and policy measures (e.g. a ban on phosphate-based detergents) the water quality has slowly improved since the 1970s (Voltz et al., 2002). Despite these measures, present-day concentrations of nutrients, salts and metals are still much higher than the natural (background) concentrations. Water quality of the Meuse also shows a considerable spatial variability. In the upstream reaches (near the source), the river water is least polluted. Water quality generally deteriorates downstream from the inflow of the Sambre river (Belgium), and the water quality remains poor down to the Belgium–Dutch border. A partial recovery occurs in the lower part in the Netherlands (Voltz et al., 2002), due to the inflow of less polluted water from tributaries and deposition of contaminated suspended solids within the channel bed.

Methods

Data collection

Water quality was investigated at the measurement stations Eijsden, situated along the Meuse at the Belgium–Dutch border and Keizersveer, situated approximately 250 km downstream of Eijsden (Fig. 1). The daily discharge record of the Meuse at Eijsden for the period 1975–2005 was used for the selection of droughts. Our selection criterion was defined as the number of days per year for which the discharge was below the threshold of $24 \text{ m}^3 \text{ s}^{-1}$ (5% of daily discharge in 1975–2005). This criterion was used instead of minimum daily discharges, since discharges in the Meuse are commonly low during summer. Furthermore, the duration of critical low-flow conditions is more relevant with respect to water quality and ecology than the absolute minimum discharge measured on a single day. The

number of days for which discharge was below $24 \text{ m}^3 \text{ s}^{-1}$ was highest in 1976 (151 days) followed by 2003 (73 days). In both years the drought started in the beginning of June and remained till the end of November. Therefore, the June–November periods in the year previous to and the year following the drought were defined as reference periods. Due to the short time span between the reference periods and selected droughts, it can be assumed that other changes in the catchment which influence water quality (e.g. land use changes, emission reduction) are negligible. The entire drought period was selected for all water quality parameters which are mainly influenced by discharge. However, for water quality parameters which are highly affected by water temperature, the summer period (June–August) of the droughts and reference periods was selected.

Water quality during both droughts (1976 and 2003) was investigated for station Eijsden, but for station Keizersveer only the 2003 drought was studied due to data availability. The combined effects of geochemical processes and inflow of tributaries on water quality were assessed by comparing water quality at both measurement stations.

Water quality parameters and sampling

The water quality parameters considered in the data analysis are relevant both with respect to the drinking water function and the ecological status of the Meuse. A total number of 24 water quality parameters were selected, which can be divided into four groups:

1. *General water quality variables*, including water temperature (watT), chlorophyll-*a* (Chl-*a*), dissolved oxygen (DO), pH, and suspended solids (SS).
2. *Nutrients*, including ammonium (NH_4^+), nitrite (NO_2^-), nitrate (NO_3^-), and orthophosphate (o-PO_4^{3-}).
3. *Major elements*, including chloride (Cl^-), bromide (Br^-), fluoride (F^-), sulphate (SO_4^{2-}) and potassium (K^+).
4. *Heavy metals and metalloids*, including lead (Pb), copper (Cu), zinc (Zn), nickel (Ni), mercury (Hg), chrome (Cr), cadmium (Cd), arsenic (As), selenium (Se) and barium (Ba).

The majority of the data were obtained from the routine water quality monitoring programme of the Netherlands, carried out by the National Institute for Inland Water Management and Waste Water Treatment (RIZA). In this programme, sampling and storage of water samples is carried out according to standard protocols. Chemical analyses are performed according to standard methods. Water temperature and dissolved oxygen are measured in the river water (around 12:00 a.m.) using a multi-purpose sensor system, chlorophyll-*a* is determined by reversed-phase HPLC, major ions and nutrients are determined using standard auto-analyser methods, heavy metals are analysed by AAS until 1998, and by ICP-MS in recent years. Full analytical details are published by the RIZA laboratory in annual reports (Geerdink and Kotte, 2004). Data series for station Eijsden were available on a daily or weekly basis; data series for station Keizersveer were available on a two-weeks or monthly basis.

Data analysis

To evaluate variations in water quality during droughts, concentrations of each quality parameter were plotted as time series for the years with the drought (1976 and 2003) and reference years (1975 and 1977; 2002 and 2004, respectively). In addition, it was tested whether water quality during droughts deviated significantly from water quality of the reference periods. Mean, median, minimum and maximum values were computed, and Wilcoxon Rank Sum tests were performed to test whether differences in water quality were significant (at 95% and 99% confidence level). In order to assess the effects of discharge and water temperature on water quality, empirical relations were established between concentration and discharge, and between concentration and water temperature at the station Eijsden for 2002–2004.

Results

Meteorological conditions

The droughts of 1976 and 2003 were characterized by large precipitation deficits. According to Beersma and Buishand (2004) precipitation deficits for the Dutch part of the Meuse basin were 381 mm for the summer half year (April–September) of 1976, and 189 mm for the summer half year of 2003. The mean recurrence times of these droughts were estimated to be 89 years (1976) and 10 years (2003) (Beersma and Buishand, 2004). The drought of 1976 was thus more severe and exceptional than the drought of 2003. In addition to large precipitation deficits, heat waves occurred during both droughts. In 1976, a heat wave (air $T_{\max} > 25^{\circ}\text{C}$) extended

for 18 days (22 June–9 July), with 15 days of maximum air temperatures higher than 30°C (recorded at the meteorological station of Maastricht, situated approximately 7 km North of Eijsden; Fig. 1). In 2003, a heat wave occurred for 14 days (31 July–13 August), with 9 days of maximum air temperatures exceeding 30°C (data of Royal Dutch Meteorological Institute (KNMI)).

Comparison of annual precipitation sums, and mean and maximum air temperatures at the meteorological station of Maastricht for 1976 and 2003, with the same parameters for the reference periods also demonstrate the drier and warmer conditions during the droughts. The annual precipitation sum was substantially lower for 1976 (554 mm) compared to the values of the reference years 1975 (639 mm) and 1977 (796 mm). The same observation applies to the year 2003 (614 mm), compared to 2002 (954 mm) and 2004 (779 mm). The mean and maximum air temperatures for the summer (June–August) of 1976 (18.9°C and 35.9°C) and 2003 (19.9°C and 36.2°C) were significantly higher ($p < 0.01$) than the values of the summers of the reference periods (Table 1). Considering the overall average of the entire period of 1975–2005 for annual precipitation sum (744 mm) and mean and maximum air temperature (17.2°C and 32.6°C), it can be concluded that the reference years represent the average meteorological conditions over the last three decades fairly well (Table 1).

Hydrology

Discharge at Eijsden was extremely low during the drought of June–November 1976 (median $Q = 8 \text{ m}^3 \text{ s}^{-1}$) compared to the reference periods of 1975 ($50 \text{ m}^3 \text{ s}^{-1}$) and 1977

Table 1 Annual precipitation sum and mean and maximum summer air temperature at the meteorological station of Maastricht for the drought years (1976 and 2003), reference years, and averaged over the entire period 1975–2005 (based on data of Royal Dutch Meteorological Institute (KNMI))

	Drought	Reference		Drought	Reference		Average
	1976	1975	1977	2003	2002	2004	1975–2005
Annual P sum (mm)	554	639	796	614	954	779	744
Mean summer (JJA) AirT ($^{\circ}\text{C}$)	18.9	17.5	16.0	19.9	18.0	17.0	17.2
Max summer (JJA) AirT ($^{\circ}\text{C}$)	35.9	32.6	30.1	36.2	33.6	32.4	32.6

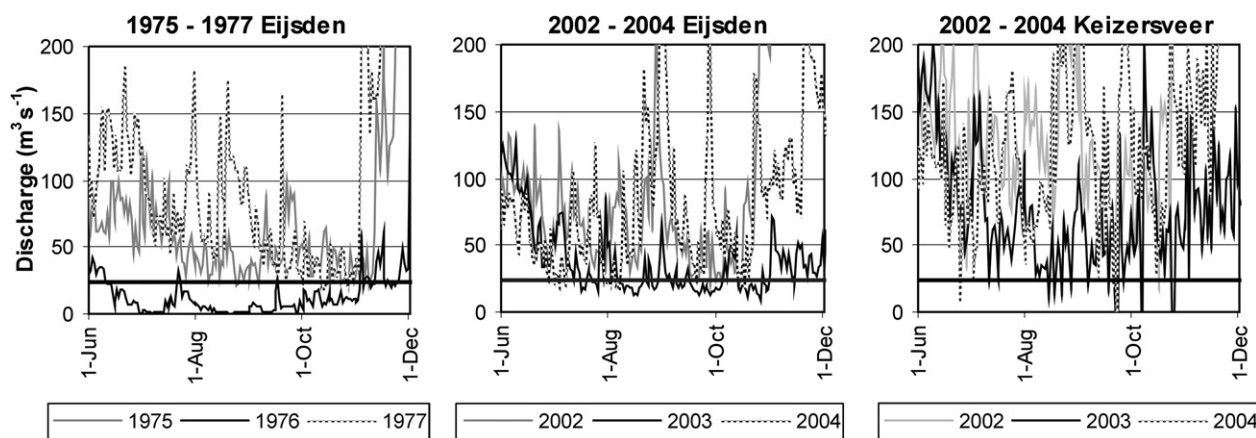


Figure 2 River flow of the Meuse at Eijsden during the drought of 1976 and 2003 and reference periods, and at station Keizersveer during the 2003-drought and reference periods. The horizontal line presents the threshold of $24 \text{ m}^3 \text{ s}^{-1}$.

($79 \text{ m}^3 \text{ s}^{-1}$) (Fig. 2). Significantly lower ($p < 0.01$) discharges were also found for the drought of June–November 2003 (median $Q = 30 \text{ m}^3 \text{ s}^{-1}$) compared to the reference periods of 2002 ($75 \text{ m}^3 \text{ s}^{-1}$) and 2004 ($70 \text{ m}^3 \text{ s}^{-1}$). Median discharges of the reference periods of 1977, 2002 and 2004 were very close to the median value estimated for the June–November period of the last 30 years ($77 \text{ m}^3 \text{ s}^{-1}$). The reference period of 1975 was characterized by somewhat drier conditions resulting in slightly lower discharges. However, in comparison to the extreme low discharge during the drought of 1976, this reference period is still considered to be appropriate for investigating the impact of droughts on water quality.

Discharges at Keizersveer are generally higher than those at Eijsden, as a result of inflow of surface water from tributaries (e.g. Rur, Niers and Dieze; Fig. 1), and groundwater flow to the Meuse downstream of Eijsden. Low-flow conditions during the 2003-drought were therefore less severe at Keizersveer (median $Q = 65 \text{ m}^3 \text{ s}^{-1}$) compared to Eijsden ($30 \text{ m}^3 \text{ s}^{-1}$). Still, Wilcoxon Rank Sum test results indicated that the discharge at Keizersveer was also significantly lower ($p < 0.01$) during the drought of 2003 than during the reference periods of 2002 ($138 \text{ m}^3 \text{ s}^{-1}$) and 2004 ($118 \text{ m}^3 \text{ s}^{-1}$).

Water quality

General water quality parameters

Water temperatures were only investigated for the recent drought of 2003 and reference periods, as no data were available at Eijsden for the period of 1975–1977. During the summer period (June–August) of 2003, high water temperatures were observed at both measurement stations with maximum values of 26.9°C at Eijsden and 24.2°C at Keizersveer. This resulted in exceedance of the surface water standard for ecological status and drinking water production (25°C) at Eijsden for several weeks. The difference in median water temperature between the drought and reference periods was more than 2.0°C , and was significant for both reference periods at Eijsden ($p < 0.01$) and Keizersveer ($p = 0.04$) (Table 2). Water temperatures were generally lower at Keizersveer compared to Eijsden. This can be explained by cooling of the river water downstream from Eijsden (despite the presence of some cooling water discharges), and by inflow of tributaries with lower water temperatures along the stretch Eijsden–Keizersveer. Water temperature at Eijsden is strongly related to daily mean air temperature ($R^2 = 0.79$; $p < 0.01$; Fig. 3a). In addition, an inverse relation was found between water temperature and river discharge ($R^2 = 0.47$; $p < 0.01$; Fig. 3b), reflecting higher warming rates of river water under low-flow conditions.

Considerably higher chlorophyll-*a* concentrations were observed at Eijsden during the summer periods of 1976 (median of $25 \mu\text{g l}^{-1}$) and 2003 ($38 \mu\text{g l}^{-1}$) compared to the reference periods (Table 2). During the summer of 2003 a maximum value of $86 \mu\text{g l}^{-1}$ was observed, which can be considered as an exceptionally high concentration for the Meuse river. In addition, distinctly higher maximum values of pH were observed during the summer periods of the droughts (8.6 in 1976 and 8.2 in 2003) (Table 2), reflecting a decrease in dissolved CO_2 concentration. Peaks in chlorophyll-*a* and pH coincided, indicating the presence of algae

blooms at Eijsden during the summer periods of the droughts. In contrast, no distinct increase in chlorophyll-*a* concentration and pH was observed at Keizersveer during the summer period of 2003, and chlorophyll-*a* concentrations here were an order of magnitude lower than at Eijsden during the entire period 2002–2004. This discrepancy in responses of chlorophyll-*a* and pH between both measurement stations can probably be attributed to the higher river flows at Keizersveer compared to Eijsden, resulting in less stagnant conditions and consequently less favourable conditions for algae growth at Keizersveer during the drought. Moreover, the water temperature at Keizersveer was some 2°C less than that at Eijsden.

Dissolved oxygen concentrations at Keizersveer during the summer of 2003 (median of 6.8 mg l^{-1}) were significantly lower ($p \leq 0.05$) compared to the summers of 2002 (7.8 mg l^{-1}) and 2004 (8.1 mg l^{-1}) (Fig. 4; Table 2). This is to be expected since dissolved oxygen solubility is lower under higher water temperatures, as confirmed by a strong negative linear relation between dissolved oxygen and water temperature ($R^2 = 0.76$, $p < 0.01$). In contrast, increases in dissolved oxygen concentrations (measured during day-time) were observed at Eijsden during large parts of the summer periods of 1976 and 2003, reaching concentrations of more than 15 mg l^{-1} in 1976 (i.e., supersaturation). As peaks in dissolved oxygen concentration corresponded with peaks in chlorophyll-*a* concentrations and pH, the increase in dissolved oxygen during day-time is very likely related to algae blooms.

Suspended solid concentrations at Eijsden were slightly lower during the drought period of 1976 (median of 8 mg l^{-1}) compared to the reference periods 1975 (9 mg l^{-1}) and 1977 (13 mg l^{-1}). The same observation applies to the year 2003 (4 mg l^{-1}) compared to 2002 (7 mg l^{-1}) and 2004 (7 mg l^{-1}) (Table 2). This is to be expected, due to the lower transport capacity of suspended solids under low-flow conditions. Differences in median concentration were significant ($p < 0.01$) for the drought of 2003. In contrast, no concentration changes were observed at Keizersveer, where the decrease in discharge was less severe compared to Eijsden during the drought of 2003.

Nutrients

Ammonium, nitrite and orthophosphate concentrations at Eijsden were considerably higher during both droughts compared to the reference periods (Fig. 5, Table 3). For ammonium, concentration increases were significant for the droughts of 1976 ($p \leq 0.01$) and 2003 ($p \leq 0.05$). Significantly ($p \leq 0.01$) higher concentrations were also observed for orthophosphate (except for the 2003-drought compared to reference period of 2004). In contrast, slightly lower nitrate concentrations were observed during both droughts compared to the reference periods ($p \leq 0.01$). This discrepancy in concentration response can be explained by different pollutant sources and geochemical processes during droughts.

The increased concentrations of ammonium under low-flow conditions (Fig. 6a) were probably due to a reduced dilution of effluents of waste water treatment plants, which operate as constant point sources of ammonium. Additionally, increased ammonium release from sediment under stagnant conditions may have contributed to the

Table 2 Summary statistics of general water quality parameters and *p*-values for significant differences (based on Wilcoxon Rank Sum tests) for the droughts of 1976 (at Eijsden) and 2003 (at Eijsden and Keizerseer), and reference periods

	Eijsden					Eijsden					Keizersveer				
	Drought		Reference		<i>p</i> -Value	Drought		Reference		<i>p</i> -Value	Drought		Reference		<i>p</i> -Value
	1976		1975	1977	1976–1975 1976–1977	2003		2002	2004	2003–2002 2003–2004	2003		2002	2004	2003–2002 2003–2004
WatT^a															
Mean						23.7		21.7	21.1		22.5		20.5	19.4	
Median						23.7		21.7	21.5	0.00	22.4		20.8	20.3	0.04
Min						20.7		19.2	18.6		20.4		18.9	18.9	
Max						26.9		24.1	25.6		24.2		23.8	23.2	
<i>n</i>						92		92	92		6		6	7	
Chl-<i>a</i>^a															
Mean	25.4			14.7		39.8		34.3	14.9		3.8		3.8	5.1	
Median	25.0			18.0		38.0		26.0	12.0	0.00	3.5		3.0	3.0	
Min	11.0			6.0		14.0		17.0	7.0		3.0		2.0	2.0	
Max	45.0			20.0		86.0		68.0	36.0		5.0		6.0	19.0	
<i>n</i>	5			3		13		13	14		6		6	7	
O₂^a															
Mean	8.5		8.1	7.3		6.2		5.5	5.7		6.8		7.7	8.0	
Median	8.6		7.8	7.3		6.0		5.3	5.6		6.8		7.8	8.1	0.02
Min	4.3		6.4	6.1		4.9		3.3	3.6		6.4		6.9	6.5	0.04
Max	15.7		10.8	8.4		8.5		6.9	7.7		7.0		8.1	10.1	
<i>n</i>	13		9	13		13		13	14		6		6	7	
pH^a															
Mean	7.9		7.9	7.8		7.7		7.5	7.6		7.6		7.9	7.7	
Median	7.9		7.9	7.8		7.6		7.5	7.5	0.01	7.6		7.9	7.8	0.00
Min	7.4		7.6	7.4		7.5		7.4	7.4	0.05	7.6		7.8	7.5	
Max	8.6		8.0	8.3		8.2		7.7	7.8		7.7		8.0	8.0	
<i>n</i>	13		11	14		13		13	14		6		6	7	
SS															
Mean	10		11	19		5		18	9		7		8	6	
Median	8		9	13	0.05	4		7	7	0.00	6		6	4	
Min	1		1	2		2		1	2		2		2	2	
Max	25		36	82		35		446	83		10		26	13	
<i>n</i>	25		20	26		183		171	183		7		11	7	

^a Statistics based on period June–August (instead of June–November).

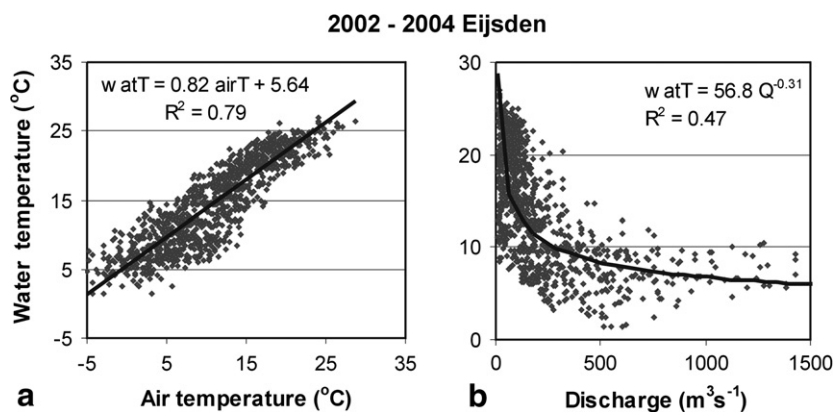


Figure 3 Relation between water temperature at Eijsden and daily mean air temperature at Maastricht (a), and between water temperature and discharge at Eijsden (b) for 2002–2004.

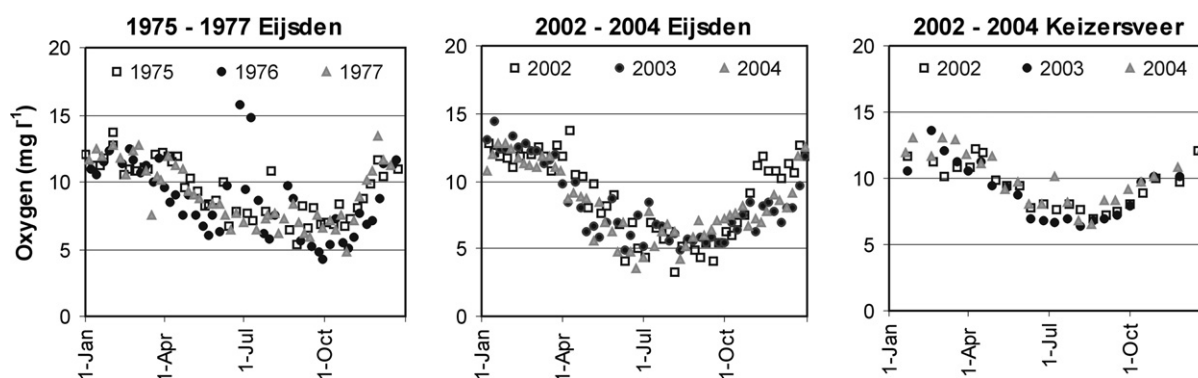


Figure 4 Dissolved oxygen concentration at Eijsden during the drought of 1976 and 2003 and reference periods, and at station Keizersveer during the 2003-drought and reference periods.

higher ammonium concentration during droughts. The sediments of the Meuse are rich in organic matter and, consequently, ammonium concentrations in the pore water are high (up to 2 mM = 28 mg N/l, Van den Berg et al., 2000). Pore water profiles of ammonium indicate that the sediments act as a source of ammonium to the water column (Van den Berg et al., 2000), which will affect surface water quality more if river flow is low. Increases in ammonium concentration due to sediment release during droughts were previously demonstrated by e.g. Mulholland et al. (1997) and Porter et al. (1996). An increase in nitrite concentration during the drought of 2003 can also be attributed to decreased dilution, as a very strong negative relation was found between nitrite concentration and discharge (Fig. 6b). Similar to ammonium, less dilution of effluents from waste water treatment plants should increase the nitrate concentration of the Meuse river. However, the nitrate–discharge relation (Fig. 6c) showed a distinct decrease in concentration under lower discharges, indicating predominant effects of other processes during droughts. As nitrate is a very mobile substance transported by soil water and groundwater, lower concentrations of nitrate might be explained by a reduced supply from soil leaching and overland flow during drought conditions. Furthermore, the negative linear relations found between ni-

trate and chlorophyll-*a*, and between nitrate and water temperature (Fig. 7) may also reflect an intensified uptake of nitrate by algae and an increased denitrification. These processes probably contributed to the slight decrease in nitrate concentration during droughts. The increase in orthophosphate is probably due to the dilution effect (similar to ammonium), as orthophosphate showed a clear negative relation with discharge (Fig. 6d). Increases in phosphate release from sediment under stagnant conditions, probably also contributed to an increased concentration during droughts. Comparison of concentrations levels of orthophosphate and ammonium for 1975–1977 with 2002–2004, clearly reflects the effects of emission reduction measures (e.g. construction of waste water treatment plants).

At Keizersveer, increases in concentration of ammonium, nitrite, and orthophosphate were less pronounced during the investigated drought (2003) compared to Eijsden (Fig. 5), due to higher river flows at Keizersveer (more dilution). Moreover, the residence time between Eijsden and Keizersveer during low-flow periods is estimated to be at least one month, which is long enough for significant losses due to biological and geochemical processes (e.g. nitrification, uptake by algae, and adsorption of orthophosphate by suspended solids and sediments).

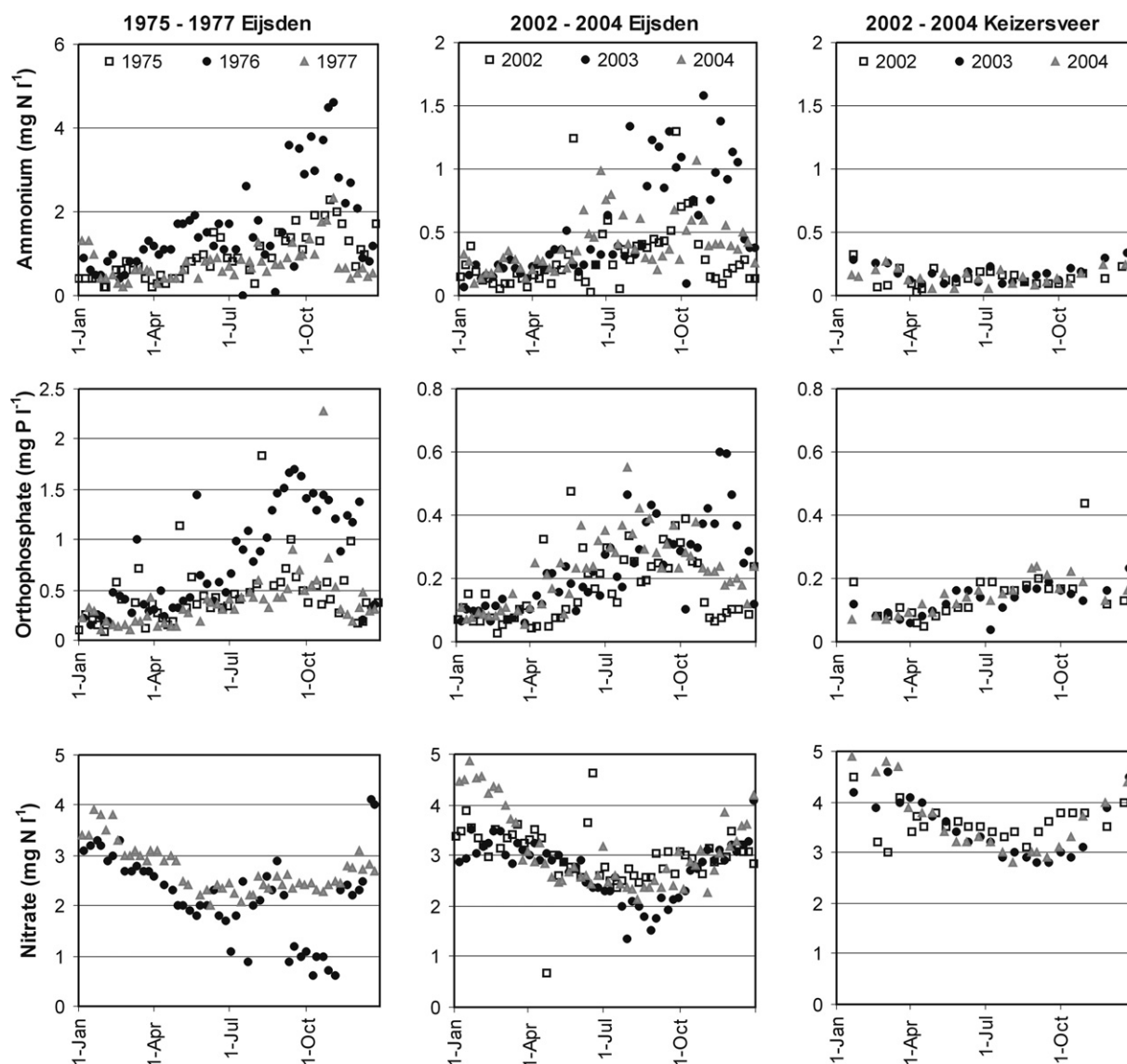


Figure 5 Ammonium, orthophosphate and nitrate concentrations at Eijsden during the drought of 1976 and 2003 and reference periods, and at station Keizersveer during the 2003-drought and reference periods.

Major elements

Distinct increases in concentration of major elements were observed at Eijsden during the droughts of 1976 and 2003 compared to the reference periods (Table 4). This is presented for chloride in Fig. 8. As chloride and other major elements are non-reactive in surface water (e.g. Van der Weijden and Middelburg, 1989), concentration increases of these conservative substances during low river flows can be explained by limited dilution of the anthropogenic input by point sources. Wilcoxon Rank Sum test results indicated that differences in concentration during the drought of 1976 and 2003 compared to the reference periods were significant ($p \leq 0.01$) for chloride, fluoride and sulphate at Eijsden (Table 4). Potassium also showed higher concentrations during the droughts. However, concentration increases were less clear and also less significant ($p \geq 0.05$) than those found for chloride, fluoride, and sulphate (Table 4). This can be explained by a relatively lower supply of potassium by point sources and higher supply by diffuse

sources (e.g. fertilizer leaching) compared to the other major elements. In contrast to point sources from which releases are, more or less, constant in time, input of substances by diffuse sources is limited during drought conditions, due to less supply by soil leaching and overland flow (cf. nitrate).

Concentration increases were also observed for the majority of major elements at Keizersveer during the drought of 2003. However, increases were generally less pronounced at Keizersveer when compared to Eijsden. This can be explained by the less severe decrease in discharge at Keizersveer during the drought of 2003, which resulted in smaller concentration effects due to limited dilution.

Due to their conservative nature, the concentration of major elements in surface water is a simple function of river discharge and emission load. The relation between discharge and concentration of these substances can be described as (Van der Weijden and Middelburg, 1989):

Table 3 Summary statistics of nutrients and *p*-values for significant differences (based on Wilcoxon Rank Sum tests) for the droughts of 1976 (at Eijsden) and 2003 (at Eijsden and Keizerseer), and reference periods

	Eijsden					Eijsden					Keizersveer				
	Drought	Reference		p-Value		Drought	Reference		p-Value		Drought	Reference		p-Value	
	1976	1975	1977	1976–1975 1976–1977		2003	2002	2004	2003–2002 2003–2004		2003	2002	2004	2003–2002 2003–2004	
NH ₄ ⁺															
Mean	2.15	1.30	0.95	0.01	0.00	0.75	0.39	0.52	0.00	0.05	0.17	0.14	0.14		
Median	1.75	1.30	0.83			0.76	0.39	0.46			0.17	0.14	0.14		
Min	0.00	0.30	0.41			0.09	0.03	0.20			0.10	0.09	0.06		
Max	4.60	2.30	2.32			1.58	1.30	1.07			0.30	0.19	0.24		
n	26	24	26			26	26	27			12	12	12		
NO ₂ [−]															
Mean						0.22	0.17	0.15			0.05	0.06	0.04		
Median						0.20	0.18	0.13		0.00	0.05	0.05	0.04		
Min						0.05	0.03	0.06			0.02	0.03	0.03		
Max						0.37	0.37	0.31			0.07	0.09	0.06		
n						26	26	27			12	12	12		
NO ₃ [−]															
Mean	1.7		2.4			2.3	2.9	2.7			3.1	3.5	3.2		
Median	1.8		2.4		0.00	2.3	2.8	2.6	0.00	0.01	3.0	3.5	3.2	0.01	
Min	0.6		2.0			1.4	2.4	2.1			2.8	3.1	2.8		
Max	2.9		2.8			3.1	4.6	3.9			3.9	3.8	4.0		
n	26		26			26	26	27			12	12	12		
o-PO ₄ ^{3−}															
Mean	1.12	0.57	0.53			0.31	0.22	0.30			0.14	0.19	0.18		
Median	1.19	0.48	0.43	0.00	0.00	0.30	0.24	0.28	0.01		0.16	0.17	0.17		
Min	0.39	0.28	0.19			0.10	0.06	0.18			0.04	0.11	0.13		
Max	1.70	1.84	2.28			0.60	0.39	0.55			0.19	0.44	0.24		
n	26	24	26			26	26	27			12	12	12		

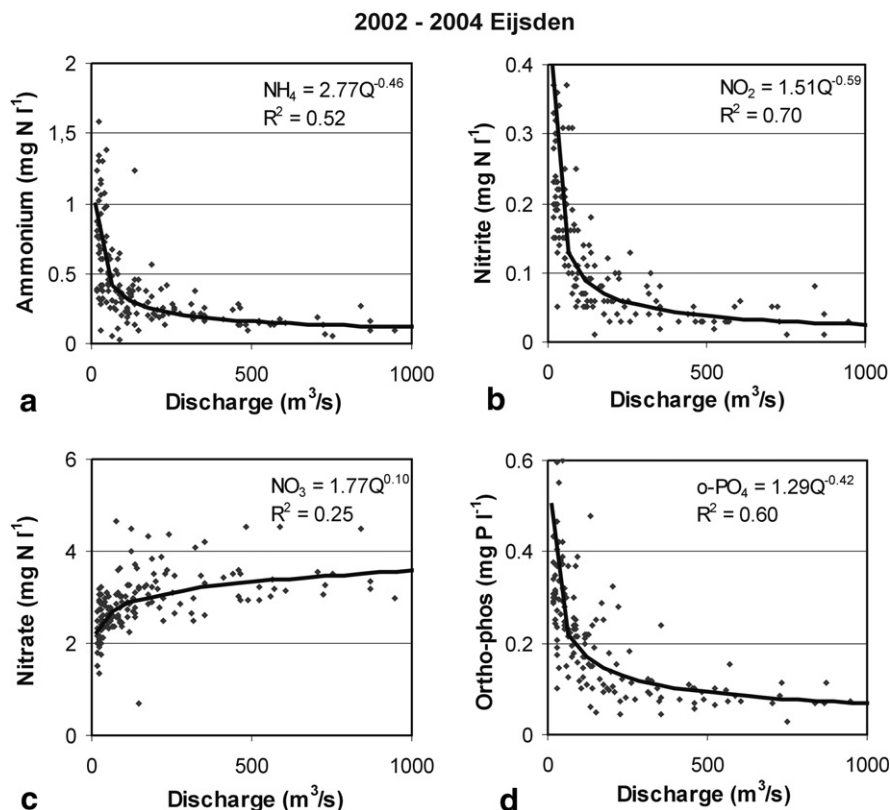


Figure 6 Concentration–discharge relations for ammonium (a), nitrite (b) nitrate (c) and orthophosphate (d) at Eijsden for 2002–2004.

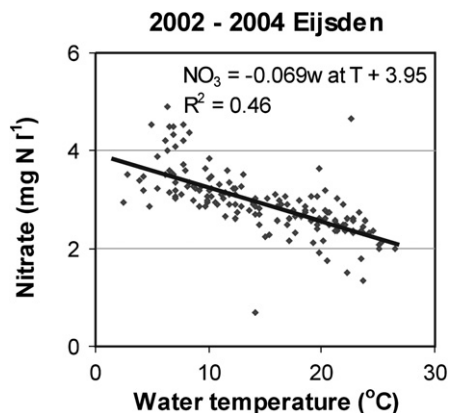


Figure 7 Concentration–water temperature relation for nitrate at Eijsden for 2002–2004.

$$C = a/Q + b$$

where C = concentration of substance in river water (mg l^{-1}), Q = discharge ($\text{m}^3 \text{s}^{-1}$), a = chemical load (g s^{-1}), b = background concentration (mg l^{-1}).

The chemical load (a) is the anthropogenic input of chemicals, mainly by point sources. The background concentration (b) both comprises the natural concentration of the river water and the input of chemicals due to overland flow in the catchment (diffuse sources). As both a and b can be regarded as constants within the short time frame considered in this study (individual drought period plus its

reference periods), concentration responses of major elements are mainly determined by discharge fluctuations, which affect the dilution of the chemical input. As an example, the relation between concentration and discharge for chloride and fluoride at Eijsden for the period 2002–2004 is shown in Fig. 9.

The obtained values of the chemical load and background concentration for all investigated major elements at Eijsden for the current situation (2002–2004) and the past situation (1975–1977) are listed in Table 5. The values of a and b for Keizersveer are presented for 2002–2004. All established relations were significant ($p \leq 0.05$). Comparison of calculated chemical loads (a) at Eijsden for both periods clearly shows the effect of emission reduction by point sources during the last three decades. Chloride and fluoride loads were reduced by 47% and 45%, respectively, and sulphate loads decreased by 20%. Emission reduction of diffuse sources also resulted in decreases in the background concentration. A reduction in b of 19% for chloride, 15% for fluoride and 9% for sulphate was found over the last three decades.

By comparing the calculated values of a and b at Eijsden to those at Keizersveer for the period of 2002–2004, downstream increases in both chemical load and background concentration could be demonstrated for chloride, bromide, sulphate and potassium. The increase in chemical load (a) amounted to 133% for potassium, 80% for bromide, 45% for chloride and 36% for sulphate (Table 5). In contrast, no increases were found for fluoride, indicating the absence of significant emission sources downstream from Eijsden.

Table 4 Summary statistics of major elements and *p*-values for significant differences (based on Wilcoxon Rank Sum tests) for the droughts of 1976 (at Eijsden) and 2003 (at Eijsden and Keizerseer), and reference periods

	Eijsden					Eijsden					Keizersveer				
	Drought	Reference		p-Value		Drought	Reference		p-Value		Drought	Reference		p-Value	
	1976	1975	1977	1976–1975	1976–1977	2003	2002	2004	2003–2002	2003–2004	2003	2002	2004	2003–2002	2003–2004
Cl [−]															
Mean	101	73	57	0.00	0.00	72	45	53	0.00	0.00	57	50	52	0.04	0.03
Median	102	72	51			72	45	50			59	52	52		
Min	60	46	32			30	16	27			42	27	45		
Max	137	104	133			160	77	96			67	61	57		
n	26	24	26			26	26	27			12	12	12		
Br [−]															
Mean						0.14		0.10			0.12	0.09	0.10		
Median						0.14		0.08			0.12	0.09	0.10		
Min						0.04		0.06			0.06	0.06	0.07		
Max						0.30		0.20			0.17	0.10	0.12		
n						13		13			6	6	6		
F [−]															
Mean	1.45	0.99	0.90	0.01	0.01	1.15	0.75	0.41	0.00	0.00	0.42	0.41	0.40		
Median	1.40	0.80	0.85			1.10	0.69	0.33			0.45	0.41	0.37		
Min	0.10	0.30	0.30			0.81	0.15	0.10			0.28	0.25	0.32		
Max	3.40	2.80	2.80			1.80	1.40	0.84			0.50	0.52	0.58		
n	25	16	26			13	13	13			7	7	7		
SO ₄ ^{2−}															
Mean	72	64	54	0.01	0.00	63	47	50	0.00	0.00	67	60	59	0.02	0.02
Median	73	64	51			67	49	48			70	61	60		
Min	49	43	38			35	25	37			53	39	53		
Max	97	90	79			79	63	65			77	67	64		
n	26	21	23			26	26	27			7	12	7		
K ⁺															
Mean						5.5	4.7	4.7			8.3	6.8	7.3		0.05
Median						5.9	4.5	4.7			8.5	7.1	7.3		
Min						3.8	2.8	3.8			6.2	4.8	6.5		
Max						6.6	6.4	5.5			9.6	8.0	8.0		
n						7	7	7			6	6	6		

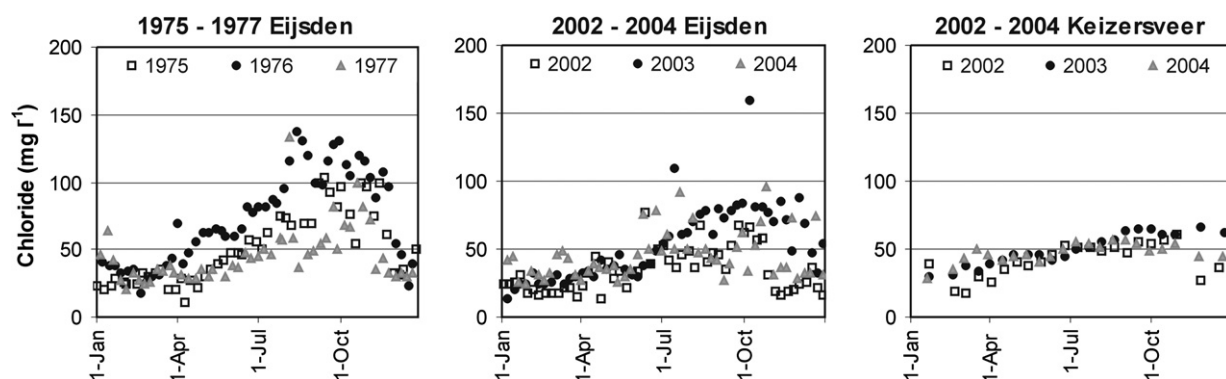


Figure 8 Chloride concentrations at Eijsden during the drought of 1976 and 2003 and reference periods, and at station Keizersveer during the 2003-drought and reference periods.

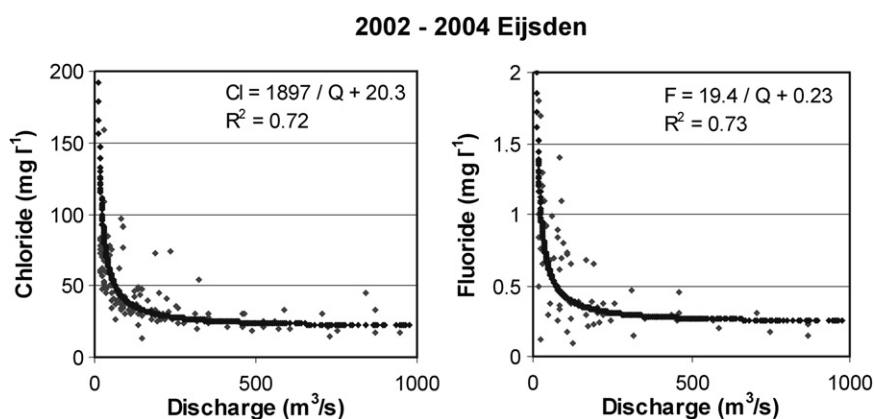


Figure 9 Concentration–discharge relations for chloride and fluoride at Eijsden for 2002–2004.

Table 5 Values of chemical loads (a), background concentration (b) and explained variance (R^2) derived from the relation between concentration of major elements and discharge at Eijsden for 1975–1977 and 2002–2004, and at Keizersveer for 2002–2004

Parameter	Location	Period	a (g s^{-1})	b (mg l^{-1})	R^2
Chloride	Eijsden	1975–1977	3576	25.2	0.55
	Eijsden	2002–2004	1897	20.3	0.72
	Keizersveer	2002–2004	2751	23.8	0.62
Bromide	Eijsden	2002–2004	1.94	0.055	0.63
	Keizersveer	2002–2004	3.49	0.060	0.64
Fluoride	Eijsden	1975–1977	35.4	0.27	0.40
	Eijsden	2002–2004	19.4	0.23	0.73
	Keizersveer	2002–2004	19.2	0.24	0.56
Sulphate	Eijsden	1975–1977	1951	29.4	0.50
	Eijsden	2002–2004	1569	26.7	0.83
	Keizersveer	2002–2004	2133	38.3	0.67
Potassium	Eijsden	2002–2004	121	2.7	0.82
	Keizersveer	2002–2004	281	4.51	0.83

Heavy metals and metalloids

Total concentrations of heavy metals (lead, copper, zinc, nickel, mercury, chrome, cadmium), metalloids (arsenic

and selenium), and barium in the Meuse river during droughts and reference periods are shown in Table 6. Higher concentrations were observed for selenium, barium

Table 6a Summary statistics of heavy metals and *p*-values for significant differences (based on Wilcoxon Rank Sum tests) for the droughts of 1976 (at Eijsden) and 2003 (at Eijsden and Keizersveer), and reference periods

	Eijsden					Eijsden					Keizersveer				
	Drought		Reference		<i>p</i> -Value	Drought		Reference		<i>p</i> -Value	Drought		Reference		<i>p</i> -Value
	1976	1975	1977	1976–1975	1976–1977	2003	2002	2004	2003–2002	2003–2004	2003	2002	2004	2003–2002	2003–2004
Pb															
Mean	6.4		14.5			1.8	5.8	1.8			1.9	3.0	2.7		
Median	4.0		8.0	0.01		1.8	2.4	1.8	0.00		2.0	1.0	2.0		
Min	1.0		2.0			0.2	1.1	0.6			1.0	1.0	1.0		
Max	23.0		70.0			3.0	47.0	3.7			4.0	12.0	9.0		
<i>n</i>	12		13			26	26	27			7	7	7		
Cr															
Mean	4.9	8.1	9.2			0.9	4.2	1.1			1.3	1.6	1.4		
Median	3.0	6.0	7.0	0.01	0.01	0.8	1.5	1.0	0.00		1.0	1.0	1.0		
Min	1.0	3.0	2.0			0.1	0.1	0.5			1.0	1.0	1.0		
Max	24.0	28.0	27.0			2.9	43.0	2.3			2.0	5.0	4.0		
<i>n</i>	12	14	13			26	25	27			7	7	7		
Hg															
Mean	0.08	0.21	0.26			0.00	0.02	0.01			0.02	0.02	0.03		
Median	0.10	0.20	0.20	0.02	0.01	0.00	0.01	0.01	0.00	0.00	0.03	0.02	0.03		
Min	0.00	0.00	0.00			0.00	0.00	0.00			0.01	0.01	0.03		
Max	0.30	0.50	0.90			0.01	0.15	0.07			0.03	0.03	0.03		
<i>n</i>	12	14	12			25	25	27			7	7	7		
Cd															
Mean	1.54	2.37	1.64			0.11	0.25	0.18			0.11	0.15			
Median	0.75	1.50	1.30	0.04		0.10	0.10	0.20	0.04	0.00	0.10	0.11			
Min	0.10	0.60	0.30			0.05	0.05	0.08			0.08	0.08			
Max	7.00	5.80	4.90			0.20	1.70	0.30			0.16	0.40			
<i>n</i>	12	14	13			26	26	27			7	7			
Zn															
Mean	64	423	146			27	42	17			21	25	26		
Median	50	249	115	0.00	0.00	24	24	15			20	19	21		
Min	20	55	45			13	10	5			17	16	16		
Max	186	1972	440			47	261	29			26	57	54		
<i>n</i>	12	14	13			26	23	27			7	7	7		

Table 6b Summary statistics of heavy metals and metalloids and *p*-values for significant differences (based on Wilcoxon Rank Sum tests) for the droughts of 1976 (at Eijsden) and 2003 (at Eijsden and Keizerseer), and reference periods

	Eijsden					Eijsden					Keizersveer				
	Drought		Reference		<i>p</i> -Value	Drought		Reference		<i>p</i> -Value	Drought		Reference		<i>p</i> -Value
	1976	1975	1977	1976–1975	1976–1977	2003	2002	2004	2003–2002	2003–2004	2003	2002	2004	2003–2002	2003–2004
Cu															
Mean	6.8	9.4	8.1			3.8	5.6	3.4			3.4	3.9	4.1		
Median	6.0	8.5	6.0			3.4	3.4	3.1			4.0	4.0	4.0		
Min	3.0	5.0	4.0			1.9	1.4	1.8			2.0	3.0	3.0		
Max	14.0	19.0	21.0			8.8	31.0	5.9			4.0	5.0	7.0		
<i>n</i>	12	14	11			26	26	27			7	7	7		
As															
Mean	25.3	13.4	6.9			1.5	1.3	1.2			1.29	1.43			
Median	18.3	13.8	6.0	0.05	0.00	1.5	1.3	1.2			1.0	1.0			
Min	12.8	4.0	3.0			1.1	0.9	1			1.0	1.0			
Max	62.4	21.0	14.0			1.8	1.8	1.6			2.0	3.0			
<i>n</i>	12	8	11			7	7	7			7	7			
Ni															
Mean	7.3	5.9	8.4			4.9	6.1	3.6			4.7	4.4	4.4		
Median	7.0	5.0	6.0			4.3	3.5	3.2		0.00	5.0	4.0	4.0		
Min	4.0	4.0	4.0			2.5	1.3	0.5			4.0	3.0	3.0		
Max	12.0	11.0	17.0			12.0	33.0	11.0			5.0	7.0	6.0		
<i>n</i>	12	14	11			26	26	27			7	7	7		
Ba															
Mean						27.0	24.4	22.9			30.6	31.6	28.6		
Median						26.3	24.9	22.1	0.05	0.00	31.0	30.0	29.0		0.02
Min						24.4	18.5	21.8			28.0	28.0	26.0		
Max						31.7	26.7	25.4			32.0	44.0	30.0		
<i>n</i>						6	7	7			7	7	7		
Se															
Mean						0.51	0.34	0.33							
Median						0.46	0.32	0.34							
Min						0.01	0.02	0.06							
Max						1.20	0.74	0.90							
<i>n</i>						7	7	7							

Table 7 Partition coefficients (K_p) of metals (Guchte et al., 2000) and percentage in the dissolved phase for suspended solid concentration of 10 mg/l

Metal	K_p (l/g)	Dissolved (%)
Selenium	0.59	99.4
Barium	1.35	98.7
Nickel	8	92.6
Arsenic	10	90.9
Copper	50	66.7
Zinc	110	47.6
Cadmium	130	43.5
Mercury	170	37.0
Chrome	290	25.6
Lead	640	13.5

($p \leq 0.05$) and nickel at Eijsden during the drought of 2003 (compared to the reference periods of 2002 and 2004). However, (total) lead, chrome, mercury, and cadmium showed significantly ($p \leq 0.05$) lower concentrations during the investigated droughts. This discrepancy is due to different adsorption capacities to suspended solids, indicated by different values of the partition coefficient (K_p). Selenium, barium and nickel have very low partition coefficients (Guchte et al., 2000) and are almost completely transported in the dissolved phase (Table 7). Increased concentrations of these substances during droughts can be explained by a reduced dilution of the chemical input. In contrast, lead, chrome, mercury and cadmium have very high partition coefficients, indicating high proportions adsorbed to suspended solids. Lower total concentrations of these particle-reactive metals during droughts can thus be explained by lower concentrations of suspended solids during droughts (Section 'General water quality parameters'). No distinct and consistent changes were found in total concentrations of copper, zinc and arsenic. This is to be expected for metals with moderately high partition coefficients (such as copper and zinc), as neither the effect of less dilution nor the effect of lower suspended solid concentration dominates. In contrast to Eijsden, no clear responses were observed in total concentrations of heavy metals and metalloids at Keizersveer during the drought of 2003, which is consistent with the suspended matter concentration (invariant) and the less dramatic decrease in river flow.

Comparison of the empirical relations between concentration and discharge for all heavy metals and metalloids thus clearly reflects differences in adsorption affinity. Heavy metals with high partition coefficients (lead, chrome, mercury and cadmium) show positive concentration–discharge relations, which are comparable with the relation between suspended solid concentration and discharge. In contrast, concentrations of substances with low partition coefficients (selenium, nickel, and barium), showed negative relations with discharge, clearly indicating the dominance of dilution effects. This divergent concentration–discharge response is illustrated for total lead (highest K_p -value) and total selenium (lowest K_p -value) in Fig. 10. Although there is considerable scatter of individual data points, the difference in concentration–discharge response between total selenium and total lead is obvious.

Discussion

The drought periods of 1976 and 2003 resulted in extremely low-flow conditions of the Meuse river, especially in the southern part of the Netherlands (measurement station of Eijsden). Apart from being exceptionally dry, both drought periods were characterized by high air temperatures and occurrences of heat waves during the summer. The combination of low river flows and high temperatures resulted in a general deterioration of the water quality of the Meuse during the droughts of 1976 and 2003, as reflected by high water temperatures, eutrophication, increased concentrations of major elements and some metals or metalloids (selenium, nickel and barium). However, concentrations of nitrate and some heavy metals with high affinity for adsorption onto suspended solids (lead, chrome, mercury and cadmium) decreased, which positively affected chemical water quality during droughts.

Warming of surface waters, and decreases in dissolved oxygen concentrations during droughts were also demonstrated in previous studies (e.g. Caruso, 2002; Murdoch et al., 2000; Mulholland et al., 1997). Decreases in dissolved oxygen can be explained by a lower oxygen saturation concentration under higher water temperatures, higher rates of organic matter decomposition, and lower re-aeration rates under low-flow conditions (Mimikou et al., 2000). This explanation is confirmed by the empirical relations between oxygen concentration and water temperature, and between

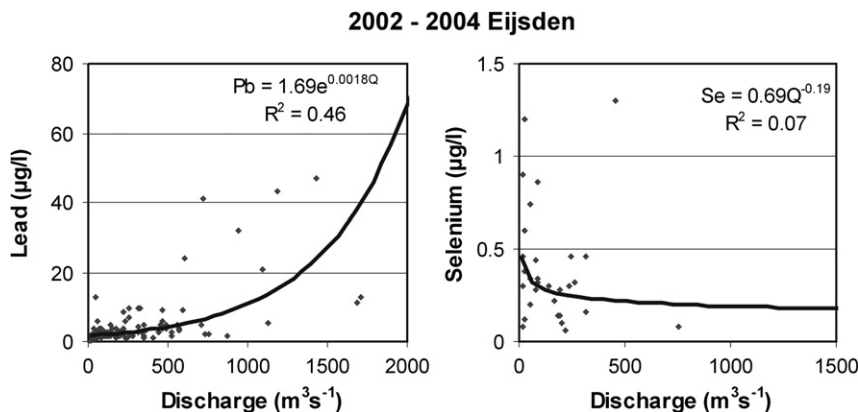


Figure 10 Concentration–discharge relations for total lead and selenium at Eijsden for 2002–2004.

oxygen concentration and discharge, established for the Meuse. Excessive algae blooms in the Meuse at Eijsden during a drought, as indicated by increases in pH and chlorophyll-*a* concentration and supersaturation of dissolved oxygen (day-time), may be less typical for other river systems. The occurrence of algae blooms in the Meuse during droughts is favoured by a combination of factors, such as extremely low discharges and long residence times, high water temperatures and high nutrient concentrations. Common patterns of increases in concentrations of chemicals released by point sources (e.g. fluoride, chloride, ammonium) were generally consistent with findings of other studies for river systems in New Zealand and North America (Caruso, 2002; Murdoch et al., 2000; Schindler, 1997), reflecting a decrease in dilution during droughts. Additionally, decreases in concentrations of substances primarily released by diffuse sources (e.g. nitrate) and substances adsorbed to suspended solids (e.g. heavy metals) were also mentioned by Mulholland et al. (1997) for freshwater systems in the South-Eastern United States. They also explained their findings by less supply by soil leaching and overland flow, and lower suspended solid concentrations under drought conditions.

From these results, it appears that the main processes affecting water quality during droughts are similar for different rivers. However, the magnitude of water quality changes depends on river regime, catchment characteristics and human activities in the catchment, which is system specific. Still, some generalizations can be made. Water quality of rivers with commonly low summer discharges and with a predominant chemical input by point sources, like the Meuse river, is expected to be most sensitive to drought conditions due to limited stream capacity for dilution and high warming rates of river water. In contrast, for rivers with a relatively high summer discharge (e.g. rivers fed by snowmelt), the magnitude of water quality changes due to reduction of the dilution capacity may be generally lower. For instance, water quality effects of droughts on the Rhine river were less pronounced compared to the effects observed in the Meuse river (Zwolsman and Van Bokhoven, 2007). Water quality of rivers with a high relative contribution of diffuse sources, might be stable or even become better during droughts, as a result of less supply of pollutants by soil leaching and overland flow. Positive effects of reduced diffuse source input on water quality were demonstrated by e.g. Caruso (2002), with respect to nitrogen.

Conclusion

The detailed assessment of the impacts of the 1976 and 2003 droughts on the water quality of the Meuse river, shows significant adverse effects with respect to water temperature, dissolved oxygen concentration, eutrophication, concentrations of major elements, and some heavy metals and metalloids (selenium, nickel and barium). Positive effects on water quality occur for nitrate and some particle-reactive heavy metals (lead, chrome, mercury and cadmium). By and large, the negative effects on water quality, especially the increase in eutrophication, outweigh the positive effects. Possible increases in the frequency and intensity of droughts due to climate change are thus expected to result in an increased temporal deterioration of water qual-

ity and, consequently, a decrease in the ecological and recreational potential of the river. Furthermore, functions depending on river water quality are expected to be hampered more often under increased drought risks. Increases in water temperatures during summer periods will limit the release of cooling water discharges by power plants, and the availability of surface water of sufficient quality for agriculture and domestic use may decrease significantly. Especially for drinking water supply, possible increases in droughts may become a serious threat, as thresholds for water temperatures and concentrations of relevant parameters such as chloride, fluoride, bromide, and ammonium are expected to be exceeded more frequently for prolonged periods. Emission reduction of point sources during low-flow conditions will be needed in order to reduce the adverse effects of droughts on surface water quality.

For the Meuse and other surface water systems in Europe, the Water Framework Directive will have an important role in reducing point source and diffuse source inputs into the water system. Regarding a possible increase in frequency and intensity of droughts in the future due to climate change (Christensen et al., 2007; EEA, 2004), emission reductions are even more necessary, especially for effluents of point sources under low-flow conditions in catchments sensitive to droughts. An obvious implication of the concentration–discharge responses found in this study would be to base permits for waste water discharges on low-flow conditions, which are most critical to water quality, instead of on average flow conditions.

Acknowledgements

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