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Impact of the 2018 drought on pharmaceutical concentrations and general water quality of the Rhine and Meuse rivers

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HIGHLIGHTS

- Impacts of the 2018 drought on water quality of the Rhine and Meuse rivers were statistically analyzed.
- Significant increases found in water temperature (on average + 1.9 °C) and salinity levels (+11%)
- Increased pharmaceutical concentrations of carbamazepine (on average + 10%) and metoprolol (+29%)
- Exceeded water quality standards for temperature, salinity, metoprolol and ibuprofen during 2018 drought
- Larger water quality impacts for rainfed Meuse river than the mixed rain/snowmelt-fed Rhine river

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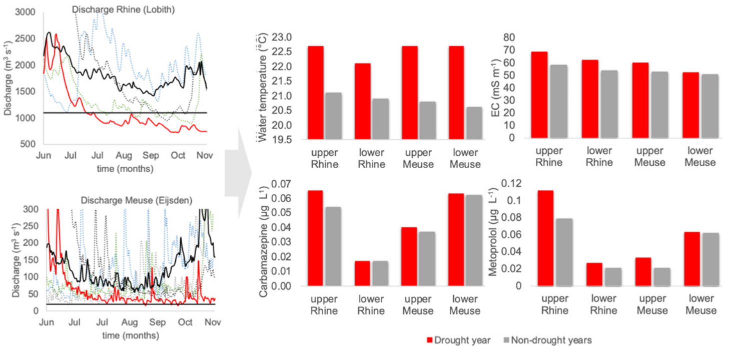
GRAPHICAL ABSTRACT



ABSTRACT

Hydrological droughts are expected to increase in frequency and severity due to changing climate in several river basins. Recent severe droughts, like the 2018 drought in northwestern Europe, have shown major challenges for water management, not only in terms of water quantity, but also water quality. However, these water quality impacts have received far less attention, and limited understanding exists, in particular regarding concentration responses of emerging chemicals, such as pharmaceutical in surface waters under droughts. This study therefore shows the impacts of the 2018 drought on the water quality of the Rhine and Meuse rivers (Western Europe) focusing on a selection of water quality parameters relevant to multiple sectoral water uses and ecosystem health, i.e. water temperature, salinity and four pharmaceuticals (carbamazepine, metoprolol, ibuprofen and sulfamethoxazole). Surface water quality data of six monitoring stations (mainly in the Netherlands) were analyzed for the 2018 drought in comparison to the reference period 2014–2017. Our results show that low flow combined with high temperatures resulted in a general deterioration of surface water quality of both the Meuse and Rhine rivers during the 2018 drought. This was reflected by significant increases in water temperatures (average of +1.9 °C) and salinity levels (+11%). While we found higher concentrations of some pharmaceuticals (carbamazepine (+10%) and metoprolol (+29%)), these increases were statistically insignificant. The decline in water quality is primarily caused by limited dilution of the chemical load derived from point sources and salinity intrusion in the lower part of Rhine-Meuse delta. A comparison of the water quality responses of the Rhine and Meuse shows larger impacts for the rainfed Meuse river with lower summer flow, compared to the mixed rain- and snowmelt-fed Rhine river. Sustainable, transboundary river water management is essential to ensure water of suitable quality for different sectoral uses during future projected droughts.

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1. Introduction

The summer and autumn of 2018 in northwestern Europe was characterized by a long-lasting drought spell, caused by a high-pressure system established over Central Europe. High evapotranspiration, due to excessive sunshine, combined with little precipitation during summer lead to extreme low flows in both the Rhine and Meuse rivers (Kramer et al., 2019; Sluijter et al., 2018). This resulted in challenges for water management not only from a water quantity, but also water quality perspective. Drinking water quality standards were temporally exceeded, and this resulted in intake stops for drinking water production and restrictions due to water pollution (Stroomberg et al., 2019).

Previous studies on impacts of droughts on river water quality have shown water quality deterioration due to a reduced dilution capacity of streams and rivers (e.g. Coppens et al., 2015; Wright et al., 2014) and an increase in the proportion of discharge that originates from polluted groundwater resources (Wright et al., 2014). Furthermore, several studies have shown increases in water temperature under droughts and compound drought-heatwave events in various river basins, mainly attributed to warmer atmospheric (higher air temperatures) conditions (Baurès et al., 2013; Delpla et al., 2009; Hanslík et al., 2016) and lower river discharge, resulting in lower thermal capacity and dilution capacity for thermal effluents from power plants and industries (Van Vliet et al., 2013, 2011).

While most studies on the impact of low-flow conditions or streamflow drought on river water quality focused on basic physical-chemical parameters, such as water temperature, chloride and nutrients (Baurès et al., 2013; Hellwig et al., 2017; Mortazavi-Naeini et al., 2019; van den Brink et al., 2019; Wright et al., 2014), limited studies focused on responses in pharmaceutical concentrations in surface waters under droughts (Mosley, 2015; Osorio et al., 2012; Palma et al., 2020). Most studies on pharmaceuticals are principally linked to the efficiency of wastewater treatment plants (Coppens et al., 2015; Lindholm-Lehto et al., 2016) or seasonal discharge fluctuations (Barbosa et al., 2018; Paíga et al., 2016) ignoring impacts under extreme events such as droughts. Research on the behavior and responses of pharmaceuticals during hydrological drought or low flow conditions is therefore scarce (e.g. Osorio et al., 2012; Sjerps et al., 2017).

Water quality impacts of the Rhine and Meuse rivers have previously been studied during major historical droughts, such as the droughts of 1976 and 2003 (Senhorst and Zwolsman, 2005; van Vliet and Zwolsman, 2008; Zwolsman and van Bokhoven, 2007). While those studies have shown an overall water quality deterioration for most of the general water quality parameters considered, limited understanding, however, exist in terms of water quality responses during the recent 2018 drought and considering impacts on pharmaceutical concentrations. Although their concentrations in the surface water are overall small and often below detection limits, the environmental impact is of concern due to the ecotoxicological effects that these low concentrations can promote in the aqueous environment (Moermond et al., 2016; Pereira et al., 2017). Moreover, public concerns regarding potential effects of unintentional exposure to pharmaceuticals is high (Houtman et al., 2013).

This study therefore evaluates the impact of the 2018 drought on the water quality of the Rhine and Meuse rivers focusing on a selected set of water quality parameters, i.e. water temperature, salinity and four pharmaceuticals (carbamazepine, metoprolol, ibuprofen and sulfamethoxazole), which are relevant for multiple sectoral water uses and ecosystem health.

The approach was based on statistical analysis of water quality monitoring records at six monitoring stations in the lower part of the Rhine and Meuse for the period 2014–2018 (Section 2). In order to assess the effects of the 2018 drought, the responses in water quality during this drought was compared to water quality under common hydrological regimes (reference years 2014–2017) with comparable chemical pollution. The concentrations of chemical substances were related to both discharge and water temperature, and exceedance of water quality threshold values were evaluated (Section 3). Additionally, the obtained results are discussed into a broader perspective by comparing the water quality responses between both rivers, and by comparing results of the 2018 drought to previous drought events, along with the implications for water management and decision making (Section 4).

2. Methods

2.1. Study area

The Rhine and Meuse river basins are located in western Europe and both have their outlets in the Netherlands with a mean annual discharge of 2068 m^3 s^{-1} of the Rhine at the German-Dutch border and 228 m^3 s^{-1} of the Meuse at the Belgium-Dutch border (average over 2010–2018). The Rhine has a length of 1230 km from its source in the Alps of east-central Switzerland to the mouth of the Rhine-Meuse Delta near Rotterdam in the Netherlands (Leuven et al., 2009). The Meuse has its headwaters in north-eastern France and then flows through Belgium and the Netherlands with a total length of 935 km (Woolderink et al., 2019). The Rhine and Meuse basins host about 50 and 8.8 million inhabitants, respectively. The main water use sectors are agriculture, cooling of power plants and process water for industry (Hut et al., 2013; Zwolsman and van Bokhoven, 2007). Furthermore, the Rhine and Meuse serve as a drinking-water source for respectively 30 and 6 million people (Houtman et al., 2013; Hut et al., 2013). The Meuse has a length of 195 km in the Netherlands and crosses the Belgian-Dutch border at Eijsden. The Rhine enters the Netherlands at Lobith (Fig. 1). Downstream of Lobith, the Rhine divides into three major tributaries: the Waal (80 km), Nederrijn (54 km) and IJssel (125 km) (Klaver et al., 2014). The Waal and Nederrijn connect with the Meuse river in the Rhine-Meuse delta plain, before debouchment into the North Sea through two major outlets (Brevé et al., 2014; Klaver et al., 2014; van den Brink et al., 2019), while the IJssel tributary discharges into the lake IJselmeer.

The regime of the Rhine is controlled by both rainfall and meltwater from the Alps. As a result, the summer discharge of the Rhine is relatively high due to meltwater pulses (e.g. Middelkoop et al., 2001). In contrast, the Meuse is a fully rain-fed river and has a smaller river basin, causing the discharge of the Meuse to respond quickly to seasonal variations in precipitation-evapotranspiration in the river basin (De Wit et al., 2007). These factors lead to a less buffered hydrological system, which results in a lower discharge in the Meuse compared to the Rhine during droughts (e.g. Zwolsman et al., 2014). During periods of low flow, the contribution of tributaries and groundwater inflow to the total discharge of the Meuse river is relatively high (Pyka et al., 2016). Additionally, weirs are used to manage a minimum water level during droughts. This results in very long residence times with almost stagnant flow conditions of the Meuse in the southern part of the Netherlands (van Vliet and Zwolsman, 2008).

The high population density, extensive agriculture, and point source emission by wastewater effluents and the many industries located along the rivers contribute to water quality deterioration (Houtman et al., 2013). Under current circumstances, over 10% of the measurements in the Meuse exceed the target value as set by the European River Memorandum (ERM) for pharmaceuticals (Sjerps et al., 2016). Most pharmaceuticals enter the water column by wastewater effluents. In addition, other sources are unintentional discharges from hospitals, the pharmaceutical industry, and runoffs from landfills or agriculture (Lindholm-Lehto et al., 2016; Palma et al., 2020). This results in already heavily polluted water entering the Netherlands, which can adversely impact drinking water production and other sectoral uses, as surface water from the Rhine and Meuse rivers accounts for 40% of the drinking water production in the Netherlands (Sjerps et al., 2017).

Fig. 1. The location of monitoring stations Lobith, Nieuwegein and Andijk along the Rhine, and the monitoring stations Luik (Belgium), Eijsden, Heel and Keizersveer along the Meuse in the Netherlands.

2.2. Water quality parameters

Our analysis focuses on six water quality parameters relevant for drinking water production, agriculture and ecological status of rivers (Sjerps et al., 2017; van der Aa and Meijers, 2016; van der Velden-Slootweg and Bannink, 2018). General parameters such as water temperature and salinity, as indicated by electrical conductivity (EC), are included. These parameters have been widely studied (e.g. Mosley, 2015; van Vliet and Zwolsman, 2008) and are well sampled also in the Rhine and Meuse rivers. Furthermore, four pharmaceuticals are included in the data analyses: carbamazepine (anti-epileptic), metoprolol (ß-blocker), ibuprofen (painkiller) and sulfamethoxazole (antibiotic) (Table 1). This selection of pharmaceuticals is based on the following criteria: the consumption volumes, previous detection (Moermond et al., 2016; ter Laak et al., 2010), ecotoxicological relevance (such as antibiotics and nonsteroidal anti-inflammatory drugs) (Palma et al., 2020), their classification as substance of attention (according to Association of River Water companies) and representation of different therapeutic classes including one representative for anti-epileptics, painkillers, β-blockers, and antibiotics (Houtman et al., 2013).

Table 1
The selected parameters for water quality and their corresponding European River Memorandum (ERM) target value.

General parameters		ERM target value
Water temperature		25 °C
Electrical conductivity (EC)		70 mS m^{-1}
Pharmaceuticals	Type	ERM target value
Carbamazepine	Anti-epileptic	0.1 μg L^{-1}
Metoprolol	β-Blocker	0.1 μg L^{-1}
Ibuprofen	Painkiller	0.1 μg L^{-1}
Sulfamethoxazole	Antibiotic	0.1 μg L^{-1}

The pharmaceuticals carbamazepine, metoprolol and sulfamethoxazole are poorly removed in wastewater treatment (Moreno-González et al., 2014; Palma et al., 2020; ter Laak et al., 2010). In addition, the water quality parameters have low degradation rates, insignificant sorption capacities and poor biotransformation (Lindholm-Lehto et al., 2016; Mandaric et al., 2019; Palma et al., 2020; ter Laak et al., 2010). These characteristics cause the pharmaceuticals to be highly present and persistent in the environment (Moreno-González et al., 2014; Palma et al., 2020). However, the concentration of ibuprofen is overall below the detection limit at most monitoring stations. The low occurrence of ibuprofen in the rivers is favored by several factors, such as the high elimination of ibuprofen during the wastewater treatment (Houtman et al., 2013; Lindholm-Lehto et al., 2016; Osorio et al., 2012; ter Laak et al., 2010). Other factors include low solubility, which favors the adsorption to the sediments, and the short degradation time in water (Coppens et al., 2015; Lindholm-Lehto et al., 2016; Palma et al., 2020).

Considering ecotoxicological and drinking water perspective, it is undesirable to exceed the target values for water quality parameters set in the European River Memorandum (ERM) by the Internationale Arbeitsgemeinschaft der Wasserwerke im Rheineinzugsgebiet (IAWR) et al. (2020). The ERM is a covenant in which 170 European drinking water companies in the major European river basins Rhine, Danube, Elbe, Meuse, Scheldt and Ruhr collectively set minimum quality requirements for river water (Table 1). It differs from mandatory water quality standards set by the Water Framework Directive (WFD), because exceedance of the ERM target value has no legal consequences. However, the ERM target values are more stringent and meet precautionary aspects and sustainability principles. In addition, the effectiveness of natural treatment methods is taken into account. River water, the composition of which remains below the ERM target values, makes it possible to prepare drinking water by natural purification methods (IAWR et al., 2020). This study therefore uses ERM target values (Table 1) in evaluating the impact of the drought.

2.3. Monitoring data

The discharge and water quality parameters of the rivers are investigated at the monitoring stations Lobith (at the German-Dutch border), Nieuwegein (Lekkanaal), Andijk (IJsselmeer) and Nieuwersluis (Amsterdam-Rijnkanaal) along the Rhine, and at the monitoring stations Luik (Belgium), Eijsden (at the Belgium-Dutch border), Heel and Keizersveer along the Meuse (Fig. 1; Table 2). Since the discharge is not measured at Nieuwegein, discharge measurements from nearby monitoring station Hagestein was used (6.5 km located upstream from Nieuwegein).

The drought in 2018 started at the beginning of June and remained till the end of autumn. Therefore, the water quality is studied for the June–November period in the drought year 2018 and compared to the June–November periods of the reference years of 2014–2017. These reference years were selected as they overall represent common hydrological conditions and similar chemical pollution. Though it is common to choose the years previous to and after a drought as reference (Hrdinka et al., 2012; Palma et al., 2020), at the time this study was conducted the data of 2019 was not yet available for inclusion in the analysis.

The database of RIWA-Rhine and RIWA-Meuse (accessed 2020), providing both water quality and discharge data for the monitoring stations, was used as basis for the analyses. Discharge is measured on 10 minute-timestep and average daily discharges are constructed from these high-temporal timestep measurements. Water temperature and EC were provided on weekly timestep at Eijsden, Keizersveer and Andijk, and every other week at Lobith, and monthly at Nieuwegein. Monitoring data of RIWA of the concentration of the pharmaceuticals

Table 2
Availability of water quality data.

	Rhine				Meuse	
	Lobith	Nieuwegein	Andijk	Nieuwersluis	Eijsden[a]	Keizersveer
Water temperature	X	X			X	X
Salinity (EC)	X	X	X	X	X	X
Carbamazepine	X	X			X[1]	X
Metoprolol	X	X			X[2]	X
Ibuprofen	X	X			X[1]	X
Sulfamethoxazole	X	X			X[2]	X

[a] For Eijsden the pharmaceutical concentrations were not measured. Therefore, the upstream locaton [1]Luik and downstream location [2]Heel were used.

were used, which are overall measured on a monthly timestep. Pharmaceutical concentrations were measured by taking samples in pre-rinsed bottles and were immediately transported in a refrigerated van to the laboratory and kept under 4 °C until processing (Sjerps et al., 2017). At the laboratory, the concentrations were determined under ISO/IEC 17025 (NEN-EN-ISO/IEC 17025, 2006).

With respect to the irregular sampling (e.g. the small sample size) of some water quality parameters, outliers are taken into account for this study. In addition, the samples which had a pharmaceutical concentration less than the detection limit were artificially set at half of the individual detection limit (Sjerps et al., 2017).

2.4. Data analysis

Streamflow drought was identified based on the number of days that discharge was lower than the 20-percentile of daily discharge calculated over the period of 2010–2018 in line with previous hydrological drought studies (Pyrce, 2004; Smakhtin, 2001). This resulted in the following threshold values for streamflow drought of 1100 $m^3\ s^{-1}$ for Lobith, 28 $m^3\ s^{-1}$ for Nieuwegein, 20 $m^3\ s^{-1}$ for Eijsden and 29 $m^3\ s^{-1}$ for Keizersveer.

The water quality responses were analysed for the monitoring stations for which water quality parameters were available (Table 2). To evaluate responses in water quality during drought, water quality of each of the parameters (i.e. water temperature, salinity and concentrations of the four pharmaceuticals) were plotted for the 2018 drought compared to the reference years of 2014–2017 and were compared to the ERM-target values (Table 1) to evaluate exceedance of these water quality target values.

In addition, it was tested whether water quality during the 2018 drought deviated significantly from water quality during the reference period. Average, median, minimum and maximum values were computed, and unpaired *t*-test and Mann-Whitney *U* test were performed to test whether differences in water quality were significant at 95% confidence level. In the case of normally distributed data (tested by the Shapiro–Wilk test (Moreno-González et al., 2014)), an unpaired t-test is carried out. If data are not normally distributed, the non-parametric Mann Whitney *U* tests are executed (Jones and van Vliet, 2018; Moreno-González et al., 2014; Mosley et al., 2012; Pereira et al., 2017). Uncertainties arise within the taken assumptions that: 1) the conditions during the drought and reference years are well represented by the measurements despite the irregular sampling and unequal amount of measurements through the years; 2) outliers are representative for the sample; 3) the artificially set values (correction for concentrations below detection limits) are representative for the sample; 4) the pharmaceuticals are emitted to surface water at a more or less constant load during the years 2014–2018.

In addition, the effects of variability in discharge and water temperature, and turbidity on water quality were estimated by fitting empirical relations between water quality, discharge and water temperature at the monitoring stations. For this, a conceptual relation describing dilution based on van der Weijden and Middelburg (1989) (Eq. (1)) was fitted for all water quality parameters and monitoring stations. Due to the conservative nature of salinity and some of the pharmaceuticals (e.g., carbamazepine), the concentration in the surface water is assumed as a simple function of river discharge and emission load (Sjerps et al., 2017). As the water quality parameter can be regarded to have stable point source emissions (both chemical load (a) and background concentration (b)) within the short time frame considered in this study (2014–2018), concentration responses of major elements are mainly determined by discharge fluctuations, which affect the dilution of the chemical input. Nonetheless, for pharmaceuticals with intermittent emissions or reactive characteristics, the concentration is also affected by processes such as the emission dynamics, transformation, decay processes and sorption. When the influence of these processes is considerable, the discharge-concentration relation may deviate from Eq. (1), which only assess dilution of the chemical loading. The goodness of fit of the relation is described by the squared correlation coefficient (R^2) and also implies which part of the variability in water temperature, EC or pharmaceutical concentration can be explained by variations in discharge or temperature.

$$C = \frac{a}{Q} + b \qquad (1)$$

where c = concentration ($\mu g\ L^{-1}$), a = chemical load ($mg\ s^{-1}$) (anthropogenic input of chemicals, mainly from point sources), Q = discharge ($m^3\ s^{-1}$), and b = background concentration ($\mu g\ L^{-1}$) (the natural concentration of the river water and the input of chemicals due to overland flow in the catchment).

3. Results

3.1. Hydrometeorological conditions

The drought of 2018 was characterized by a considerable precipitation deficit. In 2018, the annual average precipitation was 607 mm, which is 240 mm less than normal (Kramer et al., 2019), and precipitation deficits with a national average maximum of 309 mm were found. The mean recurrence time of the 2018 drought was estimated to be 30 years (Sluijter et al., 2018). In comparison, the mean recurrence times of previous severe droughts of 2003 and 1976 were estimated to be 10 years and 98 years, respectively (Beersma and Buishand, 2004). Therefore, the drought of 2018 is more exceptional than the drought of 2003, but less extraordinary compared to 1976. Apart from being exceptionally dry, two heat waves occurred during the drought of 2018. The first heatwave (air temperature > 25 °C) lasted 13 days (15 July–27 July) with 4 days of tropical temperatures (air temperature > 30 °C). The second heatwave extended for 13 days (29 July–7 August) (KNMI database, accessed 2020).

Discharge values of the Rhine and Meuse were significantly lower ($p < 0.01$) during the 2018 drought (June–November) (Fig. 2) with median discharges at upstream station Lobith of 924 $m^3\ s^{-1}$ for the 2018 drought compared to the discharge for the June–November

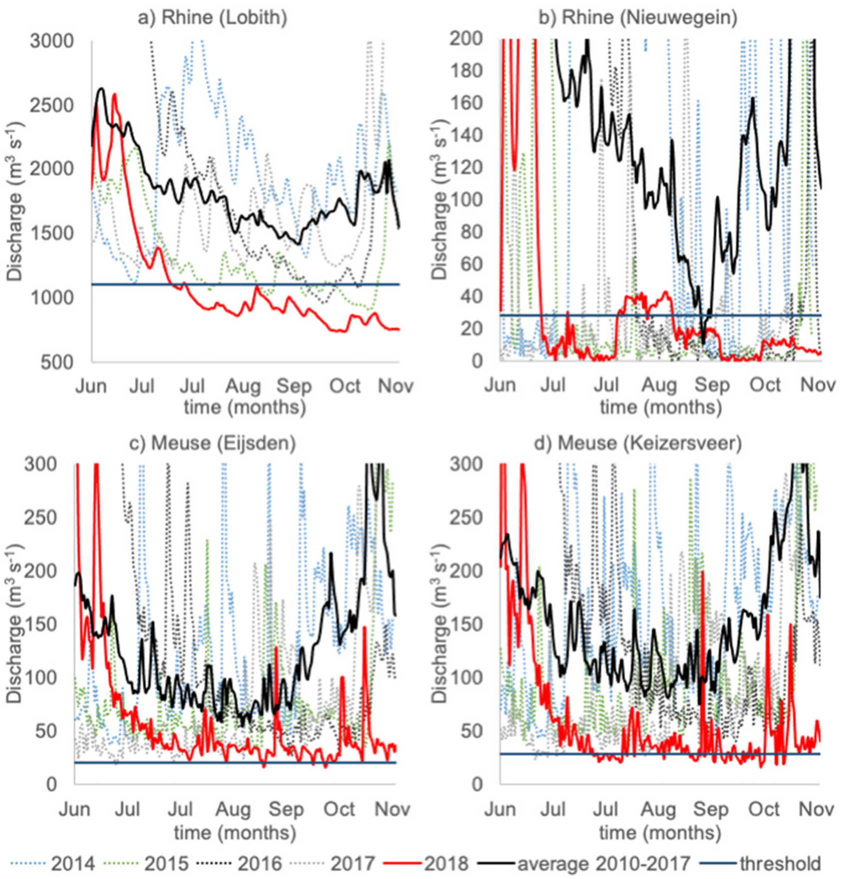

Fig. 2. Discharge of the Rhine at the monitoring stations Lobith (a) and Nieuwegein (b) and discharge of the Meuse at Eijsden (c) and Keizersveer (d) during the drought of 2018 (June–November), the reference years 2014–2017 and the average discharge of the period 2010–2017. The horizontal lines represent the threshold of 1100 m^3 s^{-1} for Lobith and 28 m^3 s^{-1} for Nieuwegein, 20 m^3 s^{-1} for Eijsden and 29 m^3 s^{-1} for Keizersveer.

periods of the reference years of 1878 m^3 s^{-1} (2014), 1209 m^3 s^{-1} (2015), 1757 m^3 s^{-1} (2016) and 1499 m^3 s^{-1} (2017). After the snowmelt period of mid-June 2018, strong declines in summer discharge of the Rhine started (Kramer et al., 2019) and with extremely low discharges from August 2018 (below 1100 m^3 s^{-1}), which remained almost continuously below that level until November. The lowest discharges of approximately 750 m^3 s^{-1} occurred in October and November (Fig. 2a–b). The discharge of the Meuse river was also distinctly lower during the 2018 drought compared to the reference years. At upstream station Eijsden (Belgium-Dutch border) significantly lower ($p < 0.01$) discharges were measured during the 2018 drought (36 m^3 s^{-1}) than during the reference period (109 m^3 s^{-1}, 62 m^3 s^{-1}, 92 m^3 s^{-1} and 53 m^3 s^{-1}, for respectively, 2014–2017), and similar results were found for downstream located stations like Keizersveer (Fig. 2c–d). The Meuse followed a similar discharge pattern as the Rhine, with quick declines in discharges (below 50 m^3 s^{-1}) from June, and which overall continued until end of November.

Regarding the median discharge of June–November over a longer period of 2010–2017 (1668 m^3 s^{-1} and 24 m^3 s^{-1} in the Rhine at Lobith and Nieuwegein respectively; 67 m^3 s^{-1} and 98 m^3 s^{-1} in the Meuse at Eijsden and Keizersveer, respectively), it can be concluded that the reference years represent the average hydrological conditions fairly well. Despite the slightly lower discharges of the Rhine and Meuse of reference years 2015 and 2017, it is still considered to be appropriate for investigating the impact of the drought on water quality. This as a result of the extremely low discharges during the 2018 drought. The 2018 drought had, relatively, larger impacts on the Meuse river than compared to the Rhine (median discharge reduction of 54% and 44%, respectively). The more extreme response of the Meuse compared to the Rhine can be explained by the fundamentally different hydrology of these rivers. The Meuse is a typical rain-fed river and its response to rainfall in the river basin is fast, whereas the Rhine is a mixed rainfed-snowmelt river.

3.2. Water temperature

Increased water temperatures were found during the summer months (June–August) of the 2018 drought in the Rhine and Meuse rivers compared to the summers of reference years 2014–2017 (on average by +1.7 °C and +2.0 °C for the Rhine and Meuse, respectively) (Fig. 3). The water temperatures exceeded the ERM target value of 25 °C once with a maximum value of 25.5 °C measured for the Rhine at Lobith. Water temperature at both monitoring stations along the Meuse exceeded the ERM target value twice, with maximum values of 25.4 °C at Eijsden and 26.5 °C at Keizersveer during the heat waves of the 2018 summer. Water temperature increases were highest for the Meuse stations (Fig. 3c–d) and were statistically significant ($p < 0.05$) as indicated by Mann-Whitney *U* test results (Supplementary Table S1). Average water temperature increases were +1.9 °C and +2.2 °C for stations Eijsden and Keizersveer (Meuse) (Fig. 3c–d) compared to +1.8 °C and +1.4 °C for stations Lobith and Nieuwegein (Rhine) (Fig. 3a–b). The elevated water temperatures corresponded with warm atmospheric conditions as indicated by the strong positive relation with air temperature (Fig. 4a). In addition, water temperature increases were also exacerbated by lower discharge due to a reduced thermal capacity and limited dilution capacity of thermal effluents from power plants and industries in both rivers, as reflected by the inverse relation between water temperature and discharge (Fig. 4b).

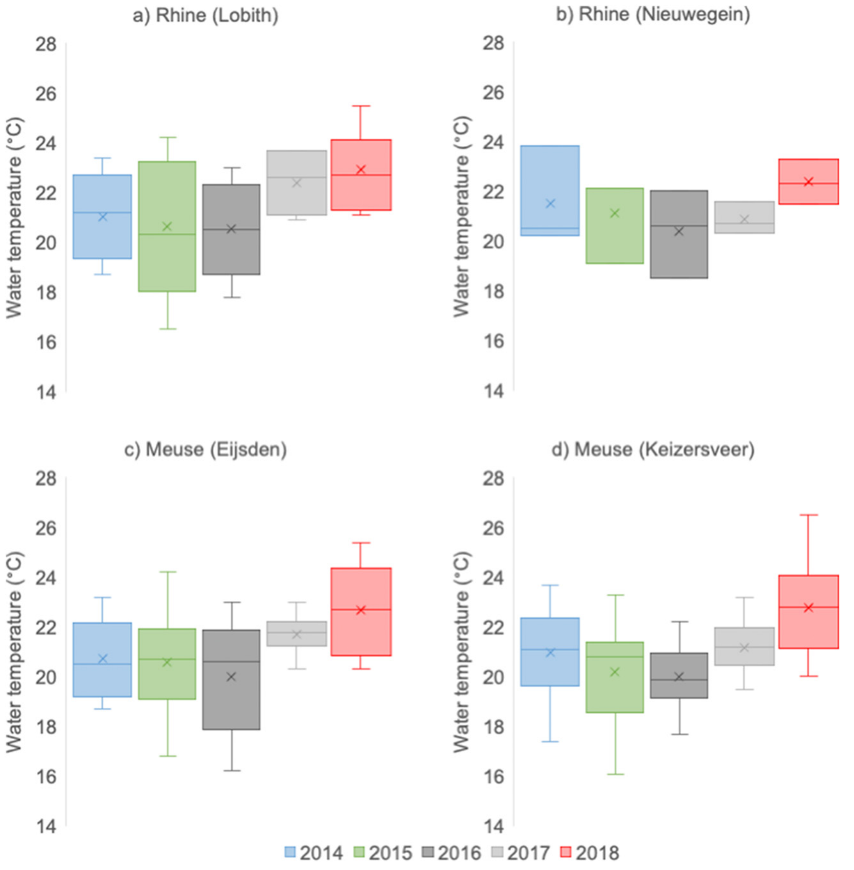

Fig. 3. The boxplots summarizing the distribution in water temperature of the Rhine at the monitoring stations Lobith (a) and Nieuwegein (b), and the Meuse at the monitoring stations Eijsden (c) and Keizersveer (d) during 2014–2018 (June–August).

Water temperatures are more strongly related to discharge in the Meuse than the Rhine, because of the higher warming rates in the Meuse (lower discharges). Also, generally higher water temperatures are observed upstream (Eijsden) rather than downstream (Keizersveer) in the Meuse. This can be explained by the cooling of the river water downstream from Eijsden (despite the presence of some cooling water discharges from power plants and industries), and by the inflow of tributaries with lower water temperatures along the stretch Eijsden-Keizersveer (Pyka et al., 2016).

3.3. Salinity

High EC values were detected during of the 2018 drought in the Rhine and Meuse rivers compared to the reference period 2014–2017 (on average by +17.3% and +8.2%, respectively). Furthermore, the ERM target value for EC (70 mS m^{-1}) was exceeded several times during the drought: six times at Lobith, two times at Nieuwegein and one time at Eijsden. The difference in average EC between the drought and reference periods was for most monitoring stations significant

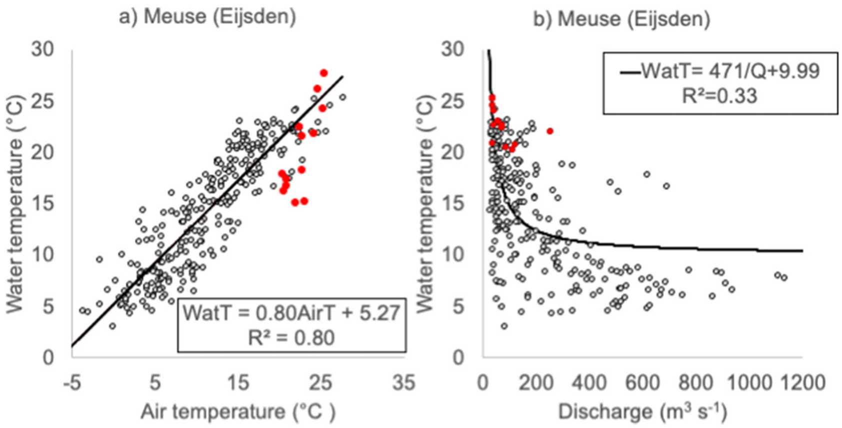

Fig. 4. The relation between water temperature at monitoring station Eijsden and air temperature (AirT) at meteorological station Maastricht (a) and between water temperature (WatT) and discharge (Q) at monitoring station Eijsden (b) for 2014–2018. The red dots represent the measurements of the 2018 summer drought (June–August).

($p < 0.05$; see Supplementary Table S1) indicated by Mann-Whitney *U* test except for 2015. The highest increases in EC were detected at Lobith (Rhine), Nieuwegein (Rhine) and Eijsden (Meuse) (on average by +18.0%, +15.6% and +13.2%, respectively) while more moderate increases were found at Keizersveer (Meuse) (+3.0%) (Fig. 5). The elevated EC values during the 2018 drought is also reflected by the strong negative relation between EC and discharge exhibiting strong determination coefficients ($R^2 = 0.70$ and 0.74), and mainly showing the limited dilution capacity of salts during low flow conditions (Fig. 6). Overall larger responses in salinity levels during the drought are found for the Rhine compared to the Meuse river. This could be the result of the already high salinity concentrating entering the Netherlands at Lobith in the summer of 2018 (Stroomberg et al., 2019) and inflow from less saline tributaries of the Meuse (Pyka et al., 2016).

To assess the influence of salinity intrusion during low flows, two more stations downstream of the Rhine located close to the open river-sea connection in the estuary were studied. The stations Andijk and Nieuwersluis show high EC during the drought of 2018, suggesting salinity intrusion in the lower part of the Rhine (Fig. 5). However, no significant difference in EC has been found at Keizersveer (Fig. 5), which indicates no influence of salinity intrusion in the lower part of the Meuse. The lower EC downstream of the Meuse could be due to the inflow of less saline tributaries and groundwater.

3.4. Pharmaceuticals

Lastly, the concentration responses of the pharmaceuticals carbamazepine, metoprolol, ibuprofen and sulfamethoxazole were studied during the drought of 2018 relative to the reference period (June–November 2014–2017). Overall, increases of the pharmaceutical concentrations were found during the summer and fall of 2018 (Fig. 7, Supplementary Figs. S1–S3, Supplementary Table S1), when the

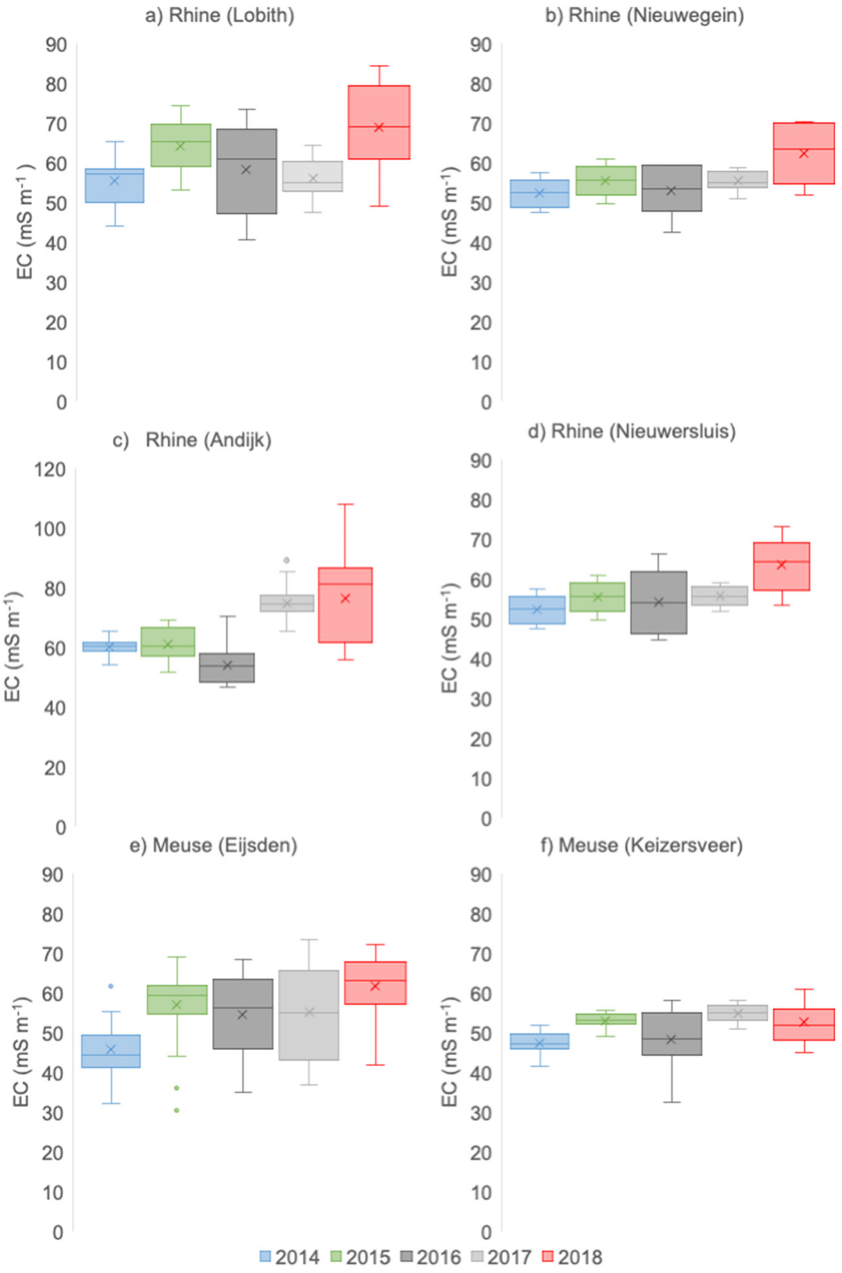

Fig. 5. The boxplots summarizing the distribution in salinity as reflected by electrical conductivity (EC) of the Rhine at the monitoring stations Lobith (a), Nieuwegein (b), Andijk (c), Nieuwersluis (d), and the Meuse at the monitoring stations Eijsden (e) and Keizersveer (f) during 2014–2018 (June–November). Note the difference in vertical axis for the Rhine (Andijk).

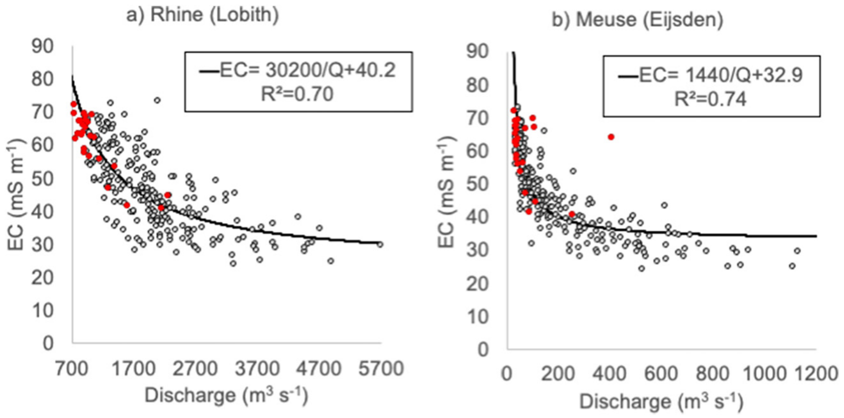

Fig. 6. The relation between salinity (EC) and discharge (Q) at the monitoring stations Lobith (a) and the Meuse at the monitoring stations Eijsden (b) for 2014–2018. The red dots represent the measurements of the 2018 drought (June–November). The black line in the EC-Q graph represents the trendline by Eq. (1).

discharge of the Rhine and Meuse is at its lowest (Fig. 2). Increased concentrations of carbamazepine and metoprolol were noted during the 2018 drought compared to the June–November periods of the reference years at almost all measurement stations (on average by +10.2% and +28.8%, respectively). Moreover, the exceedance of the ERM target value for pharmaceuticals (0.1 μg L^{-1}) occurred 3 out of 12 measurements for metoprolol in the Rhine (with a maximum concentration of 0.19 μg L^{-1} at Lobith). The Mann-Whitney *U* test and unpaired *t*-test indicated, however, that differences in average concentrations of carbamazepine and metoprolol during the drought of 2018 compared to most reference period was non-significant ($p > 0.05$) (see Supplementary Table S1). However, it should be noted that the irregular sampling, unequal number of measurements through the years, artificial correction of values below the detection limit and differences in pharmaceutical consumption through the years make these results less reliable. The ibuprofen concentrations were overall stable and showed non-significant

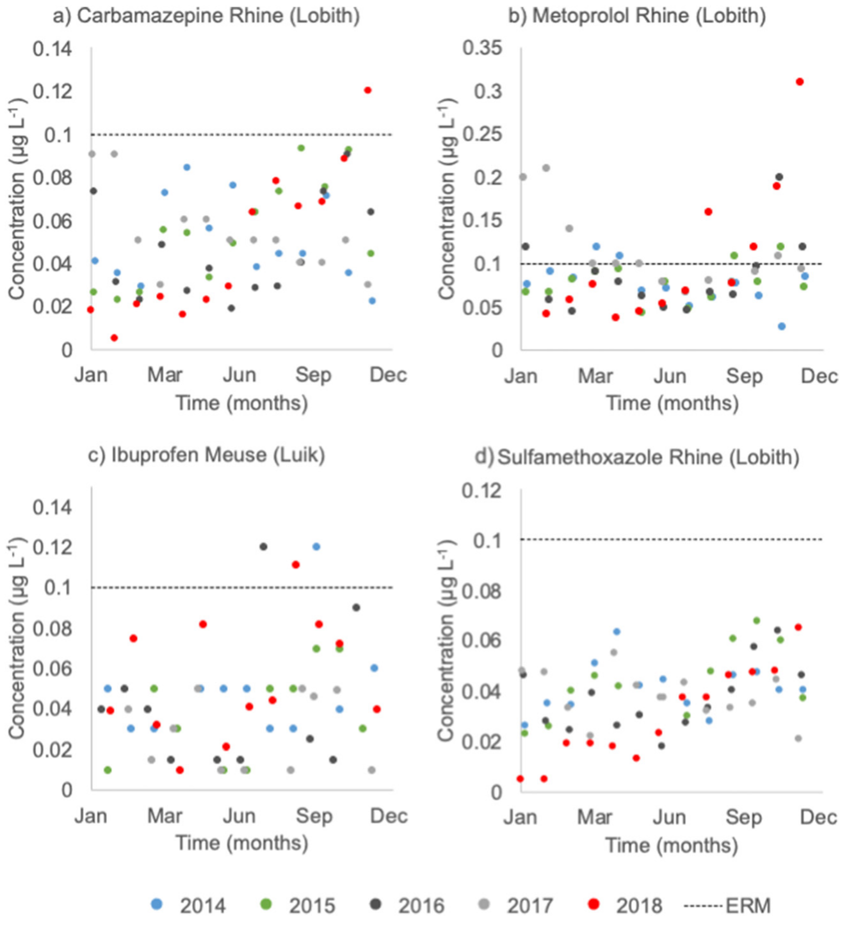

Fig. 7. Pharmaceutical concentration of carbamazepine (a), metoprolol (b), ibuprofen (c) and sulfamethoxazole (d) for the monitoring station Lobith during the drought of 2018 and the reference period 2014–2017. The black dashed line represents the ERM target value of 0.1 μg L^{-1}. Note the different values in the vertical axis of the graphs.

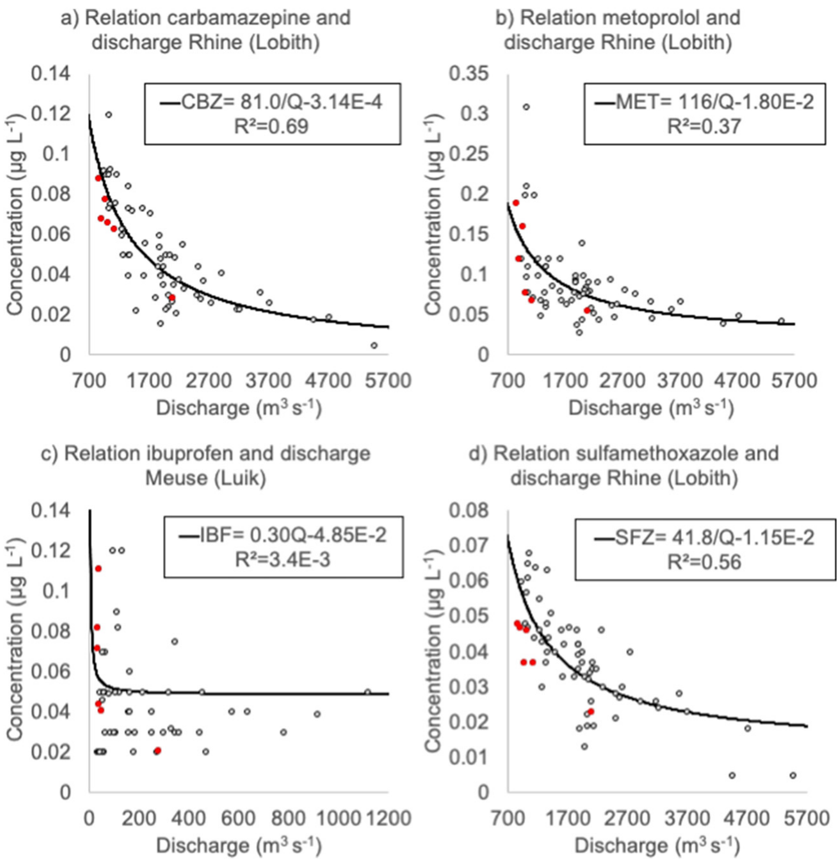

Fig. 8. The relation between the concentration of the pharmaceuticals carbamazepine (CBZ), metoprolol (MET), ibuprofen (IBF) and sulfamethoxazole (SFZ) and the discharge (Q) for a selection of monitoring stations (e.g. Lobith and Luik) the period 2014–2018. The red dots represent the measurements of the 2018 drought (June–November). The black line in the pharmaceutical-Q graph represents the trendline by Eq. (1). Note the different values in the vertical axis of the graphs.

differences compared to most of the reference years (Fig. 7). This is because the reactive properties of ibuprofen compensate for the dilution effect during the drought. The higher water temperatures and longer residence time cause ibuprofen to be more inclined to degradation. Nonetheless, ibuprofen exceeded the target value once during the 2018 drought in the Meuse with a concentration of 0.11 μg L^{-1}. Likewise, both the monitoring stations of the Rhine demonstrated similar concentrations and a non-significant impact ($p > 0.05$) with respect to the pharmaceutical concentrations of sulfamethoxazole during the period 2014–2018. The monitoring station at Heel however showed significantly higher ($p \le 0.01$) sulfamethoxazole concentrations during the 2018 drought compared to most reference years. In contrast, lower concentrations were found downstream the Meuse at Keizersveer during the drought. Nevertheless, the concentration decrease was less clear and also less significant than the increase in the concentration found upstream the Meuse. Moreover, as expected by distinct increases of the pharmaceuticals during summer and fall, distinct inverse concentration-discharge relations are observed for carbamazepine ($R^2 = 0.69$), metoprolol ($R^2 = 0.37$), and sulfamethoxazole ($R^2 = 0.56$) expect for ibuprofen ($R^2 < 0.01$) (Fig. 8). The concentration-discharge relations showed a distinct increase in concentration under lower discharges due to limited dilution. No clear relations were found between the concentrations and water temperature.

By comparing the emission loads using the fitted *a*-value of the concentration-discharge relations (Eq. (1)) of carbamazepine and metoprolol in the Meuse at Luik and Heel to those at Keizersveer, it can be concluded that there are substantial contributions of emission sources between those monitoring stations. This is reflected by an increase in emission loads by +72% for carbamazepine and by almost +210% for metoprolol (see Supplementary Table S1). Considering wastewaters as the main pathway for input of pharmaceuticals to rivers, it is common to observe higher pharmaceutical concentrations in the downstream compared to the upstream parts of rivers (Mandaric et al., 2019; Pereira et al., 2017). However, decreases in emission loads were found for all the pharmaceuticals downstream the Rhine, and ibuprofen (not detected downstream the Meuse) and also for sulfamethoxazole downstream of the Meuse based on the estimates of fitted *a*-value from the concentration-discharge relations at these monitoring stations (Supplementary Table S1). This points out the natural attenuation in the rivers by different degradation processes such as biodegradation and photodegradation (Osorio et al., 2012) and the absence of significant emission sources downstream the rivers. The pharmaceuticals in the Rhine for instance derive mostly from abroad (van der Aa and Meijers, 2016), resulting in higher concentrations upstream (at Lobith) than downstream (at Nieuwegein). In contrast, at the intake points along the Meuse, the contribution of pharmaceuticals from the Netherlands is roughly the same as from abroad. Overall higher concentrations of carbamazepine and metoprolol are therefore found downstream of the Meuse. Metoprolol is even below the detection limit at monitoring station Luik, as this pharmaceutical is mainly used in the Netherlands and hardly in Belgium (Houtman et al., 2013; van der Aa and Meijers, 2016).

4. Discussion and conclusions

A general deterioration of the water quality of the Rhine and Meuse was found during the 2018 drought due to extreme low flow conditions combined with high temperatures. This was first of all reflected by higher water temperatures and salinity (EC) levels, which where statistically significant and resulted in exceedance of the ERM target values. Also higher concentrations of some pharmaceuticals (carbamazepine, metoprolol and sulfamethoxazole) were found, but these changes were overall statistically insignificant (see Supplementary Table S1). In addition, the ERM target values for metoprolol and ibuprofen were exceeded during the 2018 drought, but also for parts of the reference (non-drought) period.

4.1. Comparison with other studies on drought impacts on water quality

The results obtained in our study and identified relations between water quality, discharge and water temperature overall correspond with findings of other studies covering the impacts of droughts on river water quality. Higher water temperatures during droughts or compound drought-heatwave event were also reported by Baurès et al. (2013), Hanslík et al. (2016), Hrdinka et al. (2012), Lehman et al. (2017), van Vliet and Zwolsman (2008) and Zieliński et al. (2009). In contrast, Mosley et al. (2012) did not observe a significant temperature increase during extreme low flows in the lower river Murray (Australia), which may have been due to local air temperature not increasing.

In addition, our results of increased salinity (EC) and the strong inverse salinity-discharge relations are in agreement with findings from the previous studies by Hellwig et al. (2017) for river basins in Germany and Jones and van Vliet (2018) for rivers in the Unites States. The increase in EC during droughts has been attributed to a reduction in dilution capacity (Hanslík et al., 2016; Hellwig et al., 2017; Hrdinka et al., 2012; Jones and van Vliet, 2018; Mosley et al., 2012) which increases the proportion of discharge from point sources (van Vliet and Zwolsman, 2008) and groundwater sources (Wright et al., 2014). Besides, increased evapoconcentration (Mosley, 2015) also contributed to increased salinity. However, the high EC downstream of the Rhine (Andijk and Nieuwersluis) during the 2018 drought can be explained by the reduced flushing and salinity intrusion during low flow conditions (Mosley et al., 2012; Wright et al., 2014), which results in the upstream movement of high salinity water from the sea (Stroomberg et al., 2019; van den Brink et al., 2019).

Overall, concentration responses of the pharmaceuticals during the 2018 drought varied, depending on their reactive or conservative properties. Increased concentrations and a distinct inverse relation between the concentrations of the pharmaceutical's carbamazepine, metoprolol and sulfamethoxazole and discharge correspond with the results of Osorio et al. (2012) and Sjerps et al. (2017). Considering carbamazepine and metoprolol are highly persistent in the environment and the distinct inverse concentration-discharge relations, the higher concentrations under drought might be due to reduced dilution capacity under low flow conditions, which is in agreement with previous studies (Barbosa et al., 2018; Houtman et al., 2013; Mandaric et al., 2019; Palma et al., 2020). However, the increases were statistically insignificant, which indicates that other processes, apart from dilution, affect the concentration of these pharmaceuticals (e.g. decay processes). While we explore relations with temperature-dependent decay processes, no significant relations were found between the concentrations of the investigated pharmaceuticals and water temperature. Although our additional analyses showed overall negative relations between pharmaceutical concentrations and turbidity water, indicating higher degradation of pharmaceuticals when turbidity decreases, these relations were weak ($R^2 < 0.4$). Hence, our results highlighted that pharmaceuticals in surface waters were more dependent on discharge than on temperature and turbidity, as formerly observed in other studies (Mandaric et al., 2019; Palma et al., 2020; Pereira et al., 2017). For ibuprofen, no clear relation with discharge was found in contrast to previous findings by Osorio et al. (2012) and Palma et al. (2020). Also, no significant relations were found between ibuprofen and water temperature while some previous studies (Azzouz and Ballesteros, 2013; Delpla et al., 2009; ter Laak et al., 2010) demonstrated lower concentrations under higher water temperatures, which may favor the process of degradation in the wastewater treatment and the environment.

While impacts of the 2018 drought on concentrations of pharmaceuticals were found, the identified increases were overall not significant ($p > 0.05$). It should, however, be noted that this might also be due to uncertainties associated with the limited number of measurements, irregular sampling of pharmaceuticals, or the artificial correction on below detection limit values, which makes the sample less representative to the conditions during the drought and the reference years. Next to this, there are several other reasons that can explain the low impact on water quality during the drought for instance, related to the general water quality developments over the past few decades increasing the self-cleaning capacity of the river (Stroomberg et al., 2019), changes in pharmaceutical loadings during the drought and increased degradation (Osorio et al., 2012). Moreover, the lack of precipitation during the drought overall leads to less sewer overflows and strongly reduces runoff of pharmaceuticals from livestock areas and paved surfaces (Stroomberg et al., 2019; ter Laak et al., 2010).

While the results obtained in our study for the Rhine and Meuse are generally in a comparable range with other studies covering the impacts of droughts on river water quality, differences exist in magnitude of water quality responses for each river system. This is due to differences in river regime, geographical conditions, climatic conditions, river basin characteristics (e.g. contribution of tributaries and groundwater), and human activities like water treatment, differences in distribution of point and diffuse sources in each river basin. A possible explanation for stronger water quality responses to the 2018 drought in the Meuse compared to the Rhine could be the fundamentally different hydrology of these rivers. The rainfed Meuse river showed, in contrast to the mixed snowmelt- and rainfed Rhine river, a stronger reduction in discharge and dilution capacity, and higher warming rates of river water. Overall, the water quality of rivers with a high relative contribution of point sources is expected to be more sensitive to water quality deterioration during drought. In contrast, river basins with a higher contribution of diffuse sources, are expected to show smaller impacts or even water quality improvements during droughts due to less supply of pollutants by soil leaching and overland flow (Wright et al., 2014).

4.2. Outlook on implications for water management

Considering potential increases in pharmaceutical emissions due to aging of the population, more livestock and population growth in the future (Sjerps et al., 2017) and the projected increase in frequency and severity of droughts (Prudhomme et al., 2014; Trenberth et al., 2014) pharmaceutical concentrations may potentially not be kept below the target values under future droughts. As a consequence, river functions depending on water quality are expected to be hampered more often under the increasing prolonged dry periods in the future. Especially for drinking water supply, a higher frequency of drought periods can become a serious threat when surface water quality standards are temporally not met, constraining the intake of surface water for drinking water production (Sjerps et al., 2017). Furthermore, the increased water temperatures in the summer season will limit the use of cooling water for thermoelectric power plant cooling (van Vliet et al., 2016), and salinization may decrease the availability and quality surface water for agricultural and domestic needs (Mortazavi-Naeini et al., 2019). Water quality deterioration under droughts may further exacerbate water scarcity in case sectoral water quality requirements are temporally not met (Jones and van Vliet, 2018; Van Vliet et al., 2017) This may require expansions in wastewater treatment and desalination in several river basins across the world (van Vliet et al., 2021). Next to this, emission control measures reducing pollutant source inputs into rivers may be necessary during future droughts, especially for effluents of point sources and for rivers which are mainly rainfed. International cooperation on sustainable, transboundary river water management are essential to ensure water of suitable quality for different sectoral uses during future projected droughts.

CRediT authorship contribution statement

E.W. performed the analyses and M.T.H.v.V. designed and supervised the project. Both authors equally contributed to the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.146182.

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