



A strategy for the implementation of water-quality-based discharge limits for the regulation of hazardous substances

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Abstract

Many developing countries apply technology-based discharge standards that set quantitative limits on pollutant discharges. These standards do not inherently consider ambient constraints and, therefore, cannot guarantee to protect aquatic life from hazardous pollutants. It is a challenge for developing countries to enforce water-quality-based limits for wastewater discharges and guarantee the intended use of water. This study aims to develop a strategy that suits the needs of developing countries for a transition from technology-based discharge standards to water-quality-based discharge limits. To this end, a pilot monitoring program was carried in the Gediz River Basin in Turkey. Surface water, industrial, and urban wastewater samples were collected and analyzed for 45 priority pollutants identified by the European Union and 250 national river basin specific pollutants. The monitoring results revealed that the environmental quality standards (EQSs) were exceeded for 8 priority, and 28 specific pollutants. This finding indicated that the existing technology-based discharge standards are not satisfactory to guarantee the intended water quality, and there is a need for adopting a new strategy for the implementation of water-quality-based discharge limits in Turkey. As a widely applied approach for determining water-quality-based discharge limits, firstly, conservative mass balance with and without consideration of mixing zone was evaluated. The results indicated that this approach was not applicable due to the receiving environment concentrations being higher than the EQSs. As an alternative approach, the dilution methodology, which considers the level of dilution occurring at the immediate discharge point, was tested. The results proved that the dilution methodology is the most appropriate strategy for developing countries with relatively poor surface water quality to improve the water quality to the level where the conservative mass balance approach can be applicable.

Keywords Dilution factor · Discharge Test Software · Hazardous substances · Environmental monitoring · Environmental quality standards · Water Framework Directive · Water-quality-based discharge limits

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Introduction

Preserving the quality of freshwater is vital for protecting aquatic ecosystems due to the biodiversity they support and the social and economic services they provide to communities. In this framework, it is vital to establish and implement adequate legislation that governs how freshwater quality is managed. Although developing countries have been making considerable progress in improving water quality, they are still in need of increased efforts to identify and strengthen the appropriate institutional, legal, and financial mechanisms for the control of wastewater discharges (Sikder et al. 2013; Haider et al. 2015) containing hazardous materials.

In recent years, hazardous substances, which are either used in or produced by many different industrial activities, have started to present a new global water quality challenge with potentially dangerous environmental and health risks (Tricker and Tricker 1999; Schwarzenbach et al. 2006; European Commission 2008). In addressing pollution from hazardous substances, the European Union (EU) uses the Water Framework Directive (WFD 2000/60/EC) that requires compliance with all the environmental quality standards (EQSs) established for priority substances at European level and river basin specific pollutants at national/local level. Therefore, the primary strategy in the EU regarding the control of wastewater discharges in compliance with EQSs at receiving environments.

There are two common approaches in the regulation of wastewater discharges: technology-based and water-quality-based standards. In a general sense, the technology-based standards are sector-specific and refer to the effluent standards that are economically achievable for industry, regardless of the quality of the receiving body. On the other side, water-quality-based discharge standards are specific to polluters and set the maximum concentration of pollutants a water body can receive while still meeting its quality standard. In Turkey, since 1988, the technology-based discharge standards defined in the Regulation on Water Pollution Control (RWPC 2004) have been applied for conventional. Nevertheless, the recently revised regulation entitled “Regulation on Surface Water Quality (RSWQ 2016)” has defined EQSs for 45 priority substances and 250 national river basin specific pollutants and dictated applying EQSs as the main approach.

In the EU countries, WFD 2000/60/EC adopts a “combined approach” for discharges from point sources into surface waters (European Commission 2010a). Firstly, it requires the specification of technology-based discharge standards, and secondly, the definition of water quality objectives for the water bodies themselves. In the USA, discharge limits are specific to polluters and determined considering both technology and water-quality-based standards. First, technology-based discharge standards are applied based on the best available techniques to provide fewer raw materials and energy use

and less waste generation in the production process. If technology-based standards are insufficient to protect the quality of the receiving environment, then water-quality-based standards are applied (USEPA 2008).

Several other countries also apply water-quality-based standards. In Japan, discharge limits are set according to the dilution of the discharge in receiving water body (Wako 2012). The discharge standard for a pollutant is defined as 10 times its annual average EQS. (AA-EQS) value for all discharges more than 50 m³/day. Similarly, in Finland, a fixed dilution factor of 10 is applied to determine discharge standards for industrial effluents in Helsinki (ECHA 2016) and medicinal effluents in London (EMEA 2006). Nevertheless, in a recent study, using the standard dilution factor of 10 is reported to result in the underestimation of environmental concentrations for pollutants by a factor of 3–5 for about 10% of wastewater treatment plants (Link et al. 2017).

The developing countries such as Korea (Kim et al. 2010), China (Li et al. 2012), and India (Rajaram and Das 2008) apply technology-based discharge standards, but fail to guarantee the intended use of water (Massoud et al. 2009). Johnstone and Horan (1996) and Johnstone (2003) discussed the difficulties in setting up and implementing the water-quality-based standards by developing countries and indicated the common problems encountered as (i) directly taking guidelines as national standards instead of adapting to the country’s circumstances, (ii) taking guideline values as absolute (rigid) values instead of target values, (iii) copying the national standards from developed countries, (iv) some standards being stringent or relaxed that not reflecting water quality objective for its intended use, (v) being too expensive of the technologies leading to compliance of standards, (vi) inadequacy of the number of the parameters as being too excessive that not having actual regional importance or being too few that not safety sufficient for intended water uses, (vii) not having adequate infrastructure and institutional capacity for supporting and regulating the implementation of standards, and (viii) not being compatible of discharge standards with carrying capacity of the water bodies (von Sperling and de Lemos Chernicharo 2002). Based on these difficulties, a stepwise implementation of water-quality-based discharge limits is indicated to be necessary for developing countries (von Sperling and de Lemos Chernicharo 2002).

In this study, it was aimed to develop a strategy to apply in a developing country, such as Turkey, for the control of water pollution originating from hazardous substances in wastewater discharges. As being late for applying water-quality-based discharge limits to improve water quality, it was targeted to develop a road map for a transition to the implementation of water-quality-based standards from the already existing

technology-based discharge standards. To this end, a pilot study in the Gediz River Basin was conducted. A four seasons water quality monitoring program was carried out covering surface waters and all major wastewater discharges. The purpose of the implementation of this monitoring study was not only to investigate the water quality in the Gediz River but also to investigate the possible application of water-quality-based discharge limits for the point sources existing in the basin. The water quality monitoring study included 45 priority pollutants identified by the WFD 2000/60/EC and also 250 national river basin specific pollutants. All these parameters were monitored both in the river and in wastewater discharges. Based on the information collected, firstly, an assessment of the water quality in the basin was made and, then alternative approaches to implementing EQS-based discharge limits that could be applied for point source pollution control in a developing country were adopted. As alternative approaches, firstly, the strategy suggested by the EU in the guidance document entitled “Technical Guidelines for the Identification of Mixed Zones” was tested. Then, setting discharge limits based on the level of dilution that occurs at the immediate discharge point was considered. Based on the results obtained, the best scenario for a transition from technology-based discharge standards to water-quality-based standards in a developing country was developed.

Materials and methods

Study area

The Gediz River Basin was selected as the study area because it is an urban area with industrial and agricultural activities. It is located in the western part of Turkey between 38° 04′–39° 13′ Northern Latitudes and 26° 42′–29° 45′ East Longitudes. The basin has an area of 1,697,892 ha, which is about 2.2% of Turkey. The average annual water potential of the basin is estimated at 2270 hm³. The Gediz River, which discharges into the Aegean Sea in Izmir, is with a length of 275 km. Two major land cover classes are agricultural areas (52.4% of total land) and forest and semi-natural areas (44.5% of total land) in the basin (EEA 2013). The detailed land use information is given in Online Resource 1. Textile, food and food processing, leather, construction materials, dairy, meat and poultry processing, and manufacturing of agricultural vehicles are the major industries in the basin. Most of the cultivated area is given to grape, olive, cherry, tomato, walnut, and cotton (Ministry of Forestry and Water Affairs 2017). In this study, the Gediz River Basin was split into 10 subbasins to ease the calculations and effectively present the results. The study area, subbasins, and sampling sites are shown in Fig. 1. The map of the Gediz River Basin land use is given in Online Resource 2.

Monitoring program

The water quality of the Gediz River was monitored throughout a year at 40 sampling sites (Fig. 1). In the selection of surface water sampling sites (Online Resource 3), the following points were taken into consideration: (i) representing the background/upstream condition of the river, (ii) representing conditions before major wastewater discharges, (iii) representing the effect of branches joining to the mainstream, (iv) representing the effects of dams/lakes on surface water quality.

In addition to surface water quality, 10 urban and 47 industrial wastewater treatment plant (WWTP) effluents were selected for the sampling study. The monitored urban WWTP discharges corresponded to 99% of the total urban WWTP discharge in the basin, while the monitored industrial discharges account for 94% of the total. Therefore, the point sources in the basin were adequately represented in the monitoring program. The samples from the surface waters and wastewater treatment plants were collected in four seasons: November 2015 (autumn), February 2016 (winter), May 2016 (spring), and August 2016 (summer).

The monitoring program also included flowrate measurement at each wastewater discharge and at each river quality monitoring station. Because of the large area of the river basin, each monitoring campaign could be completed in 7 days, and the collected samples were transported to the laboratory on a daily basis using cold storage.

Sample collection and preparation

The samples collected were placed in either amber glass or plastic bottles depending on the analysis procedure (Online Resource 4) and transported to the laboratory using cold storage (4 °C). Subsequently, the samples were frozen at –20 °C until analysis. The monitoring program also included flow measurements at each discharge and river quality sampling site using an “Acoustic Doppler Current Profiler (ADCP).”

Analysis of the samples and the evaluation of the results

The samples were analyzed for 45 priority and 250 specific pollutants listed in the Turkish Regulation on Surface Water Quality (RSWQ 2016) at the TUBITAK MRC Environment and Cleaner Production Institute’s accredited laboratories. The analyses of pollutants were performed using ICP-MS, GC-MS, GC-MS/MS, and LC-MS/MS instruments (Online Resource 5 and 6).

The analysis results were evaluated considering the average of the four seasonal measurements. All the values recorded below the limit of detection (LoD) of the analytical method

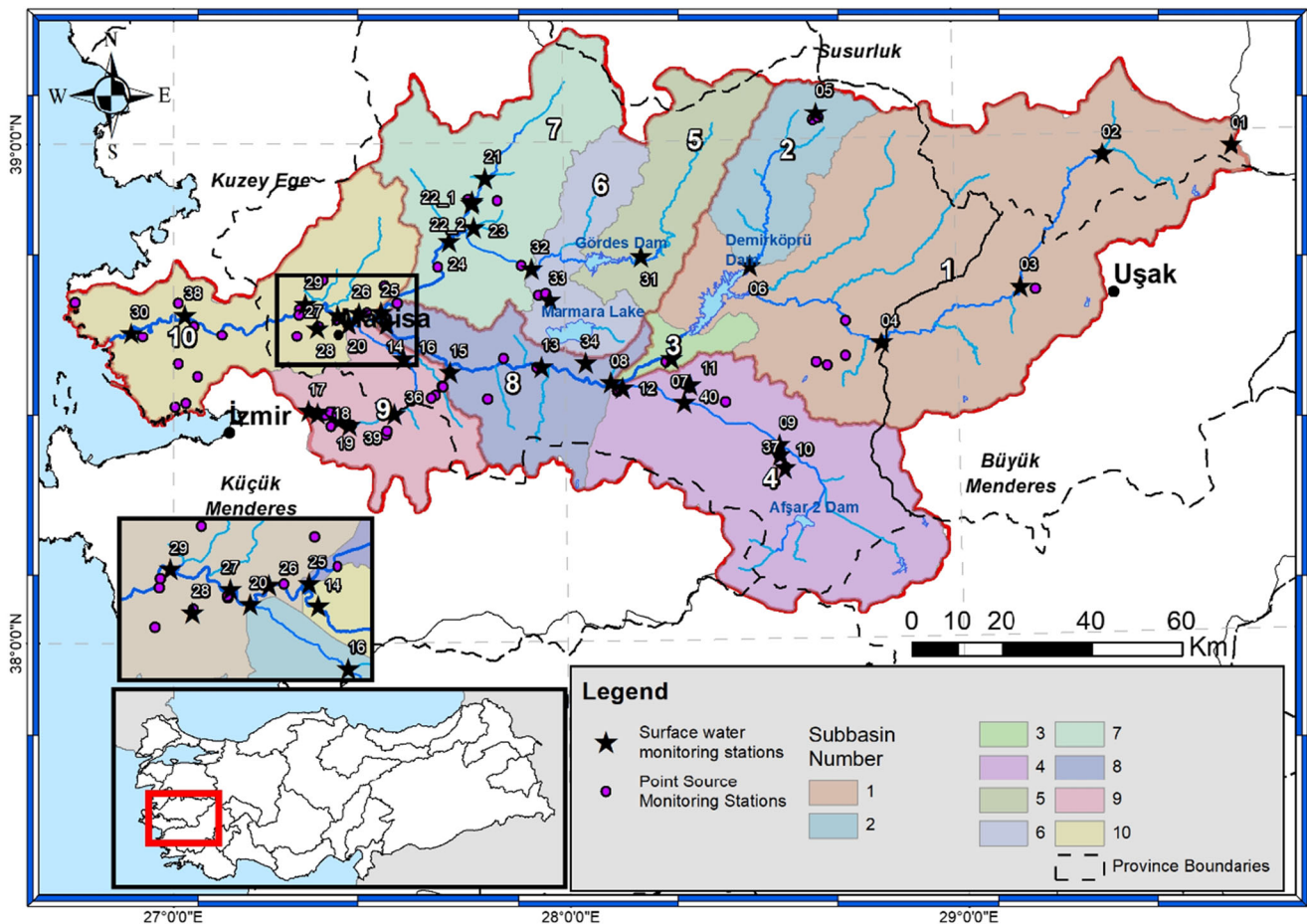


Fig. 1 Gediz River Basin, subbasins, and sampling sites

were halved and taken as measured data, as suggested in the document entitled “Technical Specifications for Chemical Analysis and Monitoring of Water Status, 2009/90/EC” (European Commission 2009). The average and maximum concentration of a pollutant measured at a given site were then compared to its AA-EQS and maximum allowable concentration (MAC-EQS), respectively. If at least one of the four measurements for a pollutant at a given site exceeded its MAC-EQS, that pollutant was considered to exceed MAC-EQS.

Methodology followed in the discharge limit determination

The methodology given by the EU in the “Technical Background Document on Identification of Mixing Zones (European Commission 2010b)” was considered in the determination of water-quality-based discharge limits. According to the WFD 2000/60/EC, EQSs are to be met at the end of the “mixing zone” where pollutants from a wastewater discharge are mixed by natural means with the receiving water. As the calculation of the extent of a mixing zone is rather complicated, the document suggests a tiered approach and to focus on

wastewater discharges that might have a significant impact on the receiving environment. The tiered approach consists of five tiers. In tier 0, wastewater discharges where pollutant concentration is below the EQS are ignored. In tier 1, selected wastewater discharges are checked if the discharge might have a significant impact on the receiving water body based on the initial mixing characteristics. Tier 2 is applied for significant discharges, which pass tier 1, and involves the estimation of the mixing zone’s size and the concentration of pollutants at the checkpoint (the maximum length of the zone) where the EQS has to be met. Tier 3 is applied for the pollutants, which exceed the EQSs at the checkpoint targeted in tier 2. For this purpose, a complex 3D model that requires extensive input data to describe the situation in a reliable way is used. Tier 4 involves running detailed investigative studies that may also contribute to any of tiers 0–3.

In the present study, the tiered approach was followed in determining water-quality-based discharge limits by limiting it to tiers 0 to 2. At tier 0, priority substances and specific pollutants that were present in a given wastewater discharge at concentrations lower than AA-EQSs were ignored. For tier 1, selected pollutants were listed, and then the significance of

their impacts on the receiving water was evaluated according to the process contribution (PC) described in the above-mentioned document. The PC, which is defined as the increase in concentration after complete mixing due to the effluent discharge, was calculated according to Eq. 1.

$$PC = \frac{C_{\text{effluent}} * Q_{\text{effluent}}}{(Q_{\text{upstream}} + Q_{\text{effluent}})} \quad (1)$$

where;

C_{effluent} Pollutant concentration in the effluent discharge
 Q_{effluent} Effluent flow rate
 Q_{upstream} Stream flow rate upstream of the effluent discharge

Q_{upstream} can be further described as the stream flowrate just before the discharge point of the wastewater effluent.

In the next step, the PC as a percentage of the AA-EQS was compared with the proposed allowable increase (Eq. 2).

$$\frac{PC}{EQS} * 100\% = \% \text{increase} \quad (2)$$

If the PCs calculated for all the selected pollutants in a given wastewater discharge were less than the proposed allowable increase—which was determined as 2% for all surface waters in the Gediz River Basin by considering the net flow—this discharge was ignored (European Commission 2010b). Otherwise, the discharge was considered to have a significant impact on the river, and the water-quality-based discharge limits were calculated for the relevant pollutants by following tier 2.

In tier 2, “Discharge Test Software” (European Commission 2010c) was used for the evaluation of mixing zone and calculation of river concentration of relevant pollutants based on conservative mass balance at the checkpoint where AA-EQS was to be met. The software is a macro-based MS Excel Workbook provided as part of the “Technical Background Document on Identification of Mixing Zones” (European Commission 2010b). It provides a mechanism for the simple estimation of the dimensions of the mixing zone (European Commission 2010b). The estimation of pollutant concentration at the end of the mixing zone was done for all pollutants that have a significant effect on the river quality, and the calculated pollutant concentrations were compared to the relevant AA-EQS. Finally, the effluent concentrations that met AA-EQS at the checkpoint or at the end of the mixing zone were defined as an EQS-based discharge limit for the relevant pollutant in the relevant wastewater discharge.

In this tier (tier 2), EQS-based discharge limits were also calculated at the discharge point in the river for all the pollutants passing tier 1 without considering the mixing zone by directly applying conservative mass balance (Eq. 3).

Equation 3 takes into account pollutant loading and dilution but did not incorporate elements of transport or environmental reactions. In other words, Eq. 3 assumes pollutants are not affected by any process along the river (such as biofouling, biodegradation, oxidation, adsorption, evaporation, etc.) and remained stable in the water:

$$C_{\text{effluent}} * Q_{\text{effluent}} + C_{\text{upstream}} * Q_{\text{upstream}} = (Q_{\text{effluent}} + Q_{\text{upstream}}) * EQS \quad (3)$$

where;

C_{upstream} Stream pollutant concentration upstream of the effluent discharge
 Q_{upstream} Stream flow rate upstream of the effluent discharge
 C_{effluent} Pollutant concentration in the effluent discharge
 Q_{effluent} Effluent flow rate

The mass balance equation (Eq. 3) was applied using the maximum observed effluent concentration under critical stream conditions (lowest river flow during sampling studies), and the water-quality-based discharge limit was calculated based on AA-EQS. This approach does not take the mixing zone into account. It only evaluates whether the EQS is exceeded at the immediate discharge point.

A second methodology based on the degree of dilution occurs in the receiving environment applied in tier 2. This approach considers the primary physical process of dilution that reduces the concentration of discharged pollutants downstream from the discharge point. The receiving water body has a certain natural dilution capacity. Based on this capacity, the effluent concentration that would result in a receiving stream after dilution was calculated. The dilution capacity was expressed by DF and calculated based on effluent and stream flow rates:

$$\text{Dilution Factor (DF)} = \frac{Q_{\text{effluent}} + Q_{\text{upstream}}}{Q_{\text{effluent}}} \quad (4)$$

The flow of a receiving stream is not constant and considerably varies throughout the seasons. Therefore, dry season flow was used to consider worst-case water quality in calculating discharge limits. In the USA, some of the regional authorities compute DFs that would be protective as dilutions computed for streams at the 7-day-average 10-year-recurrence annual low flow ($7Q_{10}$) (Colman et al. 2016; USEPA 2019). Considering the fact that flow data is only available at limited locations and not available for the long term, modifications were performed to the general DF formula (Eq. 4). Minimum upstream flow rate from the seasonal monitoring campaigns and average flow rate of wastewater discharges were used to represent the worst-case scenario in the determination of dilution. Four different scenarios were developed (Table 1) to represent the conditions in the basin better.

Table 1 Scenarios for dilution factor calculation

	Discharge flow rate*	Stream flow rate**	Dilution factor
Scenario 1	$Q_{\max, \text{effluent}}$ Maximum flow of all wastewater discharges in the water body	Q_{upstream} Stream flow rate at the upstream of the water body that receives the discharge	$\frac{Q_{\text{upstream}} + Q_{\max, \text{effluent}}}{Q_{\max, \text{effluent}}}$
Scenario 2	$Q_{\text{effluent, total}}$ Total flow rate of all wastewater discharges in the water body	Q_{upstream} Stream flow rate at the upstream of the water body that receives the discharge	$\frac{Q_{\text{upstream}} + Q_{\text{effluent, total}}}{Q_{\text{effluent, total}}}$
Scenario 3	$Q_{\text{effluent, i}}$ Flow rate of i^{th} point source	$Q_{\text{upstream, i}}$ Stream flow rate upstream of the effluent discharge, i	$\frac{Q_{\text{effluent, i}} + Q_{\text{upstream, i}}}{Q_{\text{effluent, i}}}$
Scenario 4	$Q_{\text{effluent, i}}$ Flow rate of i^{th} point source	$Q_{\text{upstream, i}}$ Stream flow rate upstream of the effluent discharge, i	$\min\left(\frac{Q_{\text{effluent, i}} + Q_{\text{upstream, i}}}{Q_{\text{effluent, i}}}\right)$
Scenario 5	Irrelevant	Irrelevant	10

*Average flow rate of wastewater discharges was used

**Minimum upstream flow rate from the seasonal monitoring campaigns was used

As summarized in Table 1, the maximum and total flow of wastewater discharges located in predefined subbasins were used for the determination of DFs in the first and second scenarios, respectively, to represent the worst-case condition. A common DF was calculated for each point source located in the same subbasin. In scenario 3, the actual dilution at the point of discharge was considered. In scenario 4, the minimum of the DFs determined in scenario 3 was taken as the DF of the subbasin. Therefore, a common DF was calculated for each point source located in the same subbasin as in the case of scenario 1, scenario 2, and scenario 4. In scenario 5, discharge limits were accepted as 10 times of the EQS regardless of flow rates. Equation 5 was applied to calculate discharge limits for each pollutant at each point source. The schematic diagram of the methodology is shown in Fig. 2.

$$\text{Discharge Limit}_{i,j} = \text{DF}_i * \text{EQS}_j \tag{5}$$

where;

- Discharge limit_{*i,j*} discharge limit determined for i^{th} point source for pollutant j ($\mu\text{g/L}$)
- DF_i dilution factor calculated for i^{th} point source
- EQS_j AA-EQS for pollutant j ($\mu\text{g/L}$)

Results and discussion

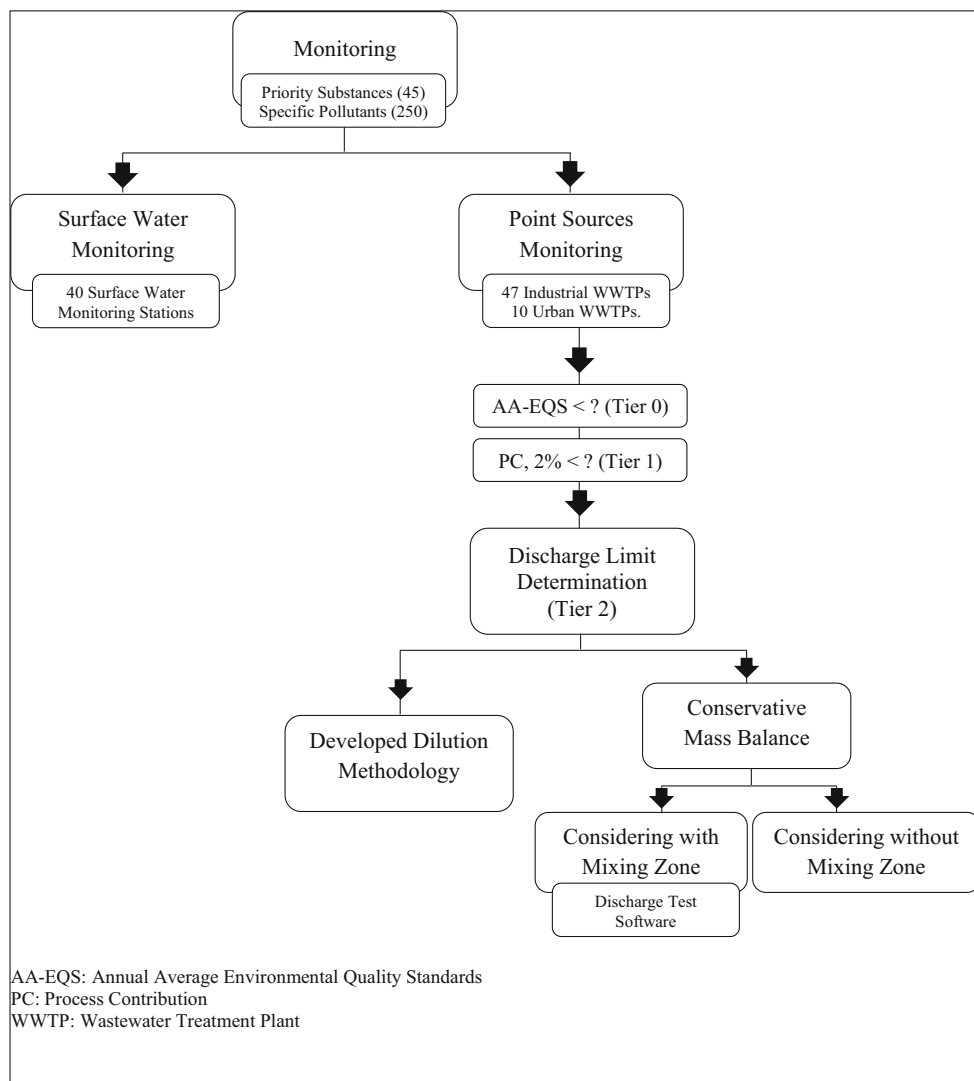
Monitoring results

Surface water monitoring

Seasonal monitoring data from 40 surface water stations between 2015 and 2016 were evaluated to determine Gediz River water

quality. In general, the concentration of pollutants in less industrialized/populated sections of the river (subbasins numbered 2, 3, 5, and 6) was significantly lower than those in highly industrialized/populated sections (subbasins numbered 4, 7, 8, 9, and 10) (Fig. 3). The results revealed 22 out of 45 priority substances present in the river. The concentrations of eight priority substances (18%) (benzene, dichloromethane, benzo(a)pyrene, fluoranthene, Cd, Ni, Pb, Hg) exceeded the AA-EQS, and six exceeded (benzene; acclonifen, Cd, Ni, Pb, Hg) the MAC-EQSs. One hundred six out of 250 river basin specific pollutants were detected in the river. The average concentrations of only 28 pollutants (11%) were higher than their AA-EQSs, and the maximum concentrations of only 29 pollutants were higher than the MAC-EQSs. The pollutants that were detected in the subbasins at concentrations higher than the AA-EQS are presented in Fig. 3. The detailed results from the monitoring program are presented in the Online Resource 7. The EQS was needed to be revised depending on the background concentrations for the assessment of metallic substances. Background concentrations are the concentrations of chemical substances (metals) resulting from only natural and geological processes (European Commission 2011). Background concentrations for metals were determined (Gursoy-Haksevenler et al. 2019), and the EQSs for 12 metals (B, Ti, Cd, Ni, Pb, Al, Co, Cr, Cu, Fe, V, Zn) were revised within the scope of this study. The priority pollutants at concentrations exceeding AA-EQS were Cd, Ni, and Pb. Among the specific pollutants, Al, As, B, Co, Cr, Cu, Fe, Zn, Si, and total petroleum hydrocarbons (TPHs) were detected in most of the stations at higher concentrations than the AA-EQSs. Imidacloprid, tolfenpyrad, nicosulfuron, fenarimol, cyfluthrin, and epoxiconazole were detected as the significant diffuse source pollutants (biocides from agriculture). These diffuse source pollutants were high mostly in the subbasins 4, 7, 9, and 10. Subbasins 4 and 7 contain agriculturally intensive areas, while the subbasins 9 and 10 contain both industrially and agriculturally intensive areas. Subbasin 9 was the

Fig. 2 Description of general methodology



most polluted subbasin with the highest number of EQS exceedance (Fig. 3). Twenty-four of these detected pollutants were only monitored in the dry season (November 2015), and seven of them, benzo(a)pyrene, 2,4-d, azoxystrobin, chlorantranilprole, chlorfluazuron, propiconazole, and tolfenpyrad, exceeded the AA-EQS. Ten of these detected pollutants were only detected in the wet season (May 2016), and only aconifen and thiabendazole exceeded the AA-EQS. In addition to cyfluthrin and imidacloprid, metals (Cd, Ni, Pb, Al, As, B, Co, Cr, Cu, Fe, V, Zn, Si, Ti, and free cyanide) exceeded the AA-EQS in all seasons. These results demonstrate that diffuse source pollutants (mainly pesticides) were more dominant in the dry season due to low flow conditions, and metal pollution was at significant levels throughout the year.

Relative to their AA-EQS values, the highest concentrations in the Gediz River were recorded for the hazardous pollutants of nicosulfuron (812%), Zn (721%), and Ni (356%). Zn and Ni were quantified at all sampling locations and during all seasons at concentrations between 694 $\mu\text{g/L}$ and

56,968 $\mu\text{g/L}$ and between 1.50 $\mu\text{g/L}$ and 11,762 $\mu\text{g/L}$, respectively. Nicosulfuron was quantified at concentrations between 0.023 $\mu\text{g/L}$ and 40.623 $\mu\text{g/L}$ at approximately 16% of all samples. Additional metals (Si, Cu, Co, Cr, Pb, Al, Fe, As, V, and Ba) were also quantified at all sampling locations and during all seasons. Except for Si, the concentrations of these metals exceeded the AA-EQSs at up to 40% of all monitoring studies while Si exceeded at a ratio of 97% (Online Resource 7). Concerning priority substances, most of the subbasin surface water stations were found to be with at least one exceedance of EQS except for a few tributaries which are discharging to dams in subbasins 2 and 5, respectively and the tributary coming from subbasin 4. As per the specific pollutants, all tributaries with monitoring results were with EQS exceedance (the maps of surface water quality classification in terms of AA-EQS exceedance for priority substances and specific pollutants are given in Online Resource 8). All these findings indicated that the water quality of the Gediz River poses a high risk to both the aquatic environment and human health

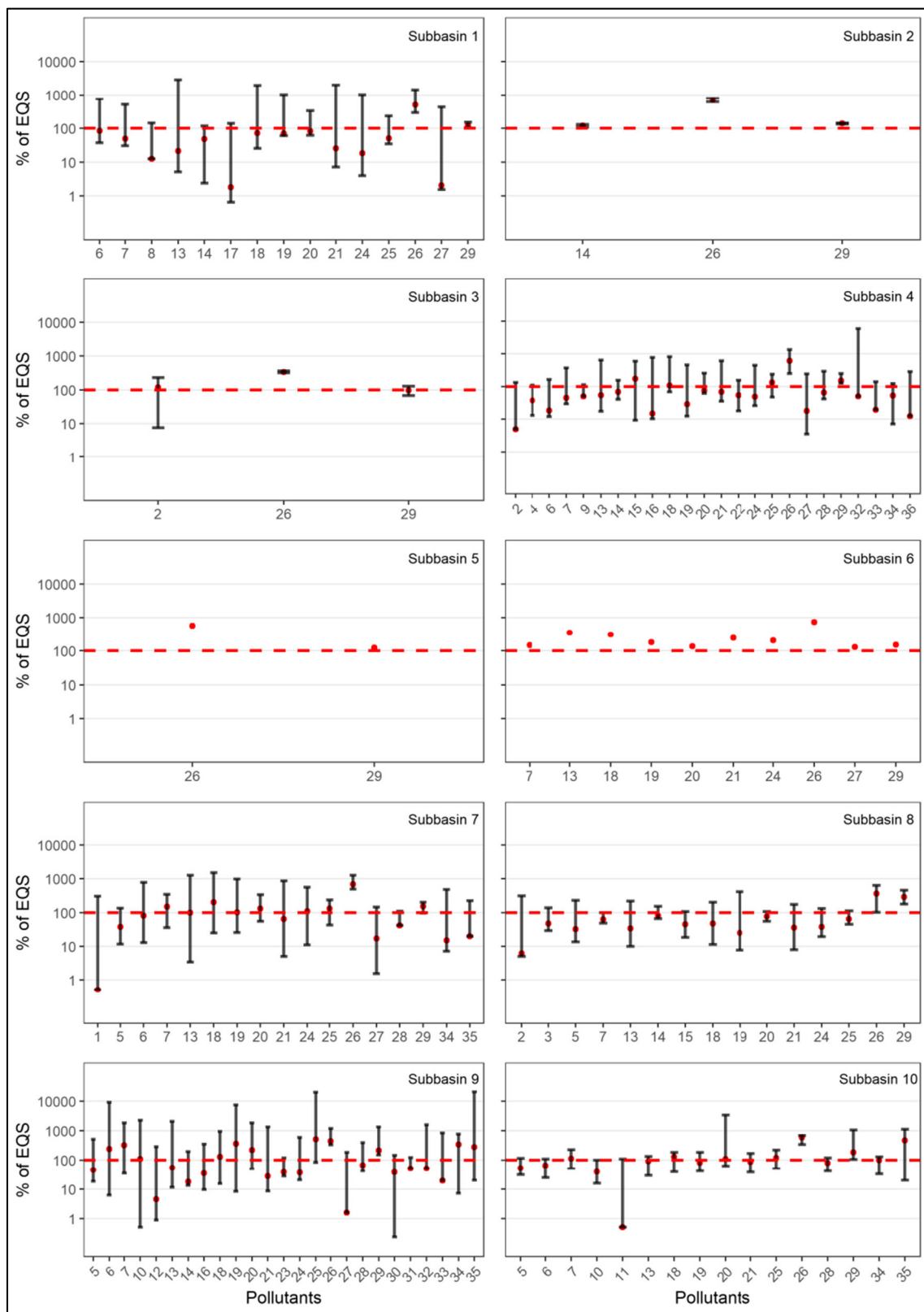


Fig. 3 Pollutants at concentrations exceeding the AA-EQS values in the subbasins of the Gediz River Basin. (1) Benzene; (2) Dichloromethane; (3) Benzo(a)pyrene; (4) Fluoranthene; (5) Cd; (6) Ni; (7) Pb; (8) Hg; (9) 4-chloroaniline; (10) Benzo(a)fluorene; (21) Fe; (22) Sb; (23) Sn; (24)V;

(25) Zn; (26) Si; (27) Ti; (28) Free cyanide; (29) TPH; (30) 2,4-dichlorophenoxy acetic acid; (31) Chlorsulfuron; (32) Cyfluthrin; (33) Diflubenzuron; (34) Imidacloprid; (35) Nicosulfuron; (36) Tolfenpyrad

due to 8 priority substances and 28 river basin specific pollutants that exceed the threshold.

Several previous studies have been conducted on the surface water quality in the Gediz River Basin aiming to investigate the presence of metals and their sources (Kucuksezgin et al. 2008; Minareci et al. 2009; Aydin and Kucuksezgin 2012; Suzer et al. 2015; Bizsel et al. 2017). A prior study done by Kucuksezgin et al. (2008) investigated the distribution of Hg, Pb, Cu, Zn, Mn, Ni, and Fe in the water samples collected from the Gediz River from five different locations in 1998 and 1999. According to their results, the highest values were observed in subbasin 10 in summer due to industrial and agricultural activities. The sampling station numbered 29 in the present study is in this subbasin, and it is the location where high metal concentrations were observed. Kucuksezgin et al. (2008) attributed high concentrations of Ni and Mn measured throughout the central part of the basin to the geochemical composition of the sediments. Another study by Kindler and Sevim (1990) confirmed that heavy metals observed in the Gediz River (Cd, Cr, Cu, Pb, and Zn) are of geomorphological origin. In our recent study, we have found out that the origin of high metal concentrations in the Gediz River waters is the geological formation of the basin (Gursoy-Haksevenler et al. 2019).

Point source monitoring

The results from the industrial wastewater monitoring part of the study indicated that dichloromethane, fluoranthene, benzo[a]pyrene, Cd, Ni, and Pb were the most common priority pollutants encountered in the industrial wastewater discharges in the basin at concentrations exceeding relevant AA-EQS (Online Resource 9). Among river basin specific pollutants, a number of organic pollutants (benzo[a]fluorine, bis(2-ethylhexyl) terephthalate and TPHs) and inorganic pollutants (Al, As, B, Co, Cr, Cu, Fe, Sb, V, Zn and Si, free cyanide) appeared as the wide-spread pollutants in the basin. When the sources discharging these pollutants were categorized, it was seen that the main industrial sectors discharging these pollutants were food processing, paper production, chemical production, textile manufacturing, and leather production industries, as well as the organized industrial zones including many different industrial sectors. Online Resource 9 shows the discharge concentrations of point sources and their sectors in each sampling study. As shown in Fig. 4, the industrial sectors that discharge the highest quantity of pollutants into the basin were in the sectors of chemical production, food processing, and organized industrial zones. The relevant industrial plants also appeared to discharge the highest number of pollutants at concentrations exceeding relevant AA-EQS.

The urban WWTP monitoring part of the study was conducted in 10 urban WWTPs performing secondary treatment. The urban WWTP monitoring results indicated that 25

pollutants including metals (Pb, As, Cr, Cu, Sb, V, Zn, Si), PAHs (benzo(a)fluorine, benzo(a)pyrene, fluoranthene), industrial chemicals (bis(2-ethylhexyl) terephthalate, 4-chloroaniline, dichloromethane), biocides (permethrin, azoxystrobin, chlorantraniliprole, chlorfenapyr, cyfluthrin, imidacloprid, nicosulfuron, tolfenpyrad, azinfos-methyl), TPHs, and tridecane existed at concentrations above their AA-EQS in urban wastewaters. The most encountered pollutants in the treated urban WWTP discharges were Zn, Si, and Cu as inorganics and imidacloprid, cyfluthrin, and nicosulfuron as organic micropollutants (Fig. 5). All the results from the urban WWTPs monitoring program are presented in the Online Resource 9. Figure 5 shows the concentrations of the pollutants exceeding the AA-EQS in urban WWTPs relative to their EQS values. Among 10 urban WWTPs, 10 or more pollutants exceeded the EQS at the discharge of two urban WWTPs, while less than five pollutants exceeded the EQS at the discharge of five urban WWTPs. TPHs, Si, and Zn were the most common ones among the hazardous pollutants. TPHs are defined as a mixture of aromatic and aliphatic hydrocarbons coming from crude oil products used in industry and transportation (Green and Trett 1989; Doble and Kumar 2005; Klimek et al. 2016). The main constituents of the PHCs are hydrogen and carbon, and consist of a mixture of chemicals such as hexane, mineral oils, benzene, toluene, xylenes, naphthalene, fluorine, and jet fuels (USEPA 2017). The PHCs detected in the Gediz River Basin might be discharged from small metal processing plants and accidental spills in this context.

When the pollutants exceeding AA-EQS values in surface waters and point sources were evaluated, it was seen that priority substances and specific pollutants presented in the rivers were also detected in industrial and/or urban effluents (Online Resource 10). On the other hand, some detected pollutants and/or exceeded AA-EQS values in industrial and/or urban effluents were not observed in the river (chlorfenapyr, dichlorvos, and xylene musk). This can be explained by the fate of discharged pollutants (such as dilution, biofouling, biodegradation, oxidation, adsorption, evaporation, etc.) in the river.

Determination of discharge limits

According to the monitoring results, it is clear that the water quality of the basin is required to be improved by better control of both point and diffuse sources of pollution. For the control of diffuse pollution originated from extensive agricultural activities in the basin, best agricultural management practices need to be adopted in accordance with the Regulation on Good Agricultural Practices (RGAP 2010), which is outside the scope of the present study. However, for better control of point sources, causing the failure of water quality standards, it is necessary to adopt water-quality-based discharge limits. To

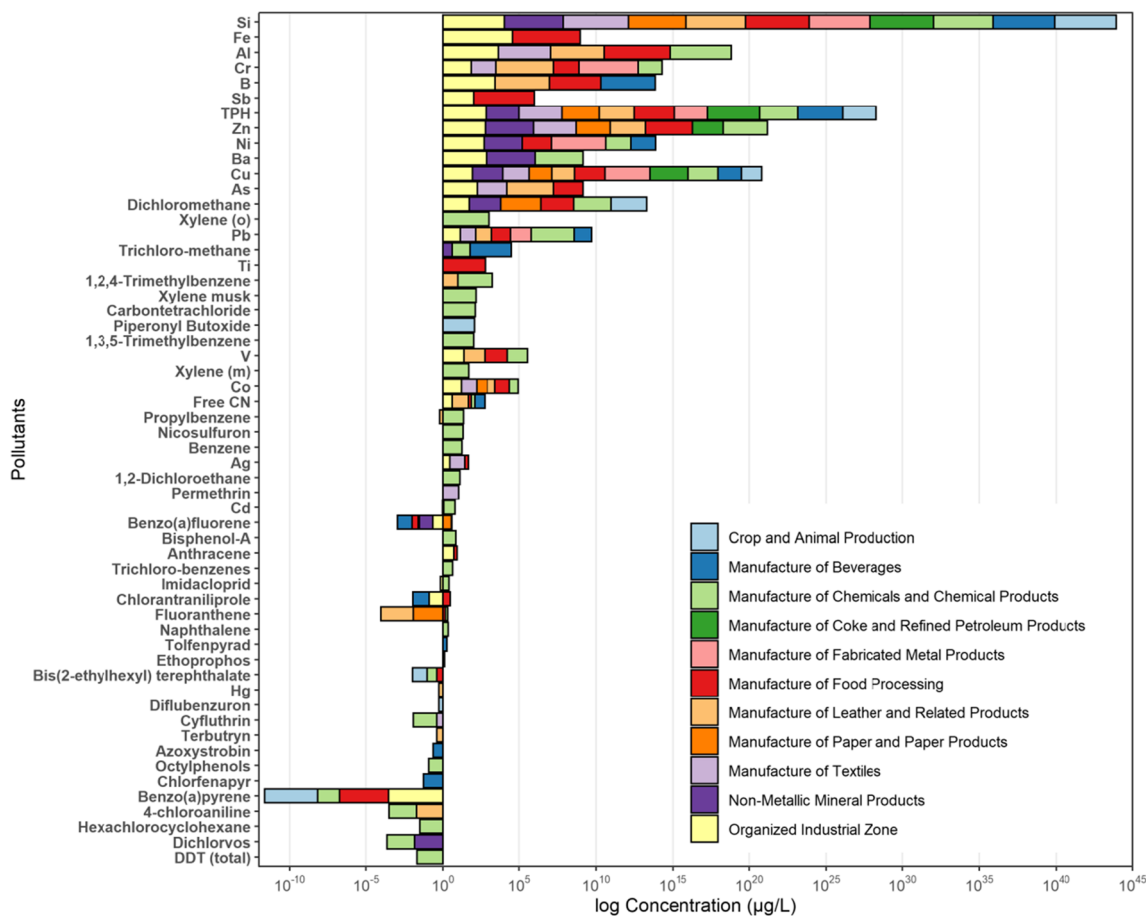


Fig. 4 Concentration of pollutants exceeding the relevant AA-EQS values in different industrial sectors of the Gediz River Basin

this end, the methodology given in Section 2.5 was applied for the pollutants exceeding the EQSs, and the optimum strategy for Turkey was tried to be developed.

The application of the tiered approach to the wastewater discharges in the basin indicated that 17 of the 45 priority substances and 41 of the 250 specific pollutants exist in concentrations exceeding relevant AA-EQS values in 57 effluents (47 industrial and 10 urban WWTP effluents). Therefore, these cases passed to tier 1. In tier 1, PCs were calculated for all pollutants that exceeded the AA-EQSs, and the cases with more than 2% PC were determined. Among the 17 priority substances and 41 specific pollutants with the AA-EQS exceedance in 57 effluents, 10 priority substances and 23 specific pollutants were found to exceed the PC limit of 2% in again 57 effluents (Online Resource 11).

Then, the “Discharge Test Software” was applied for the wastewater discharges with pollutants passed tier 1, and the water-quality-based discharge limits were calculated for 10 priority substances and 23 specific pollutants in 57 wastewater discharges. It appeared that the software could only be used if the upstream concentration of a pollutant is lower than its AA-EQS value. This limitation was due to the conservative mass balance expression used in the calculation. When subbasins

were investigated, it was observed that this condition could not be satisfied for most of the pollutants (the results are not presented as they are not acceptable) existing in 57 wastewater discharges. From this finding, it appeared that the “Discharge Test Software” could only be used after a significant improvement in the basin’s water quality. There are limited studies on the use of Discharge Test Software in the literature. One of them is the study done by Ceka (2011), who applied the WFD Mixing Zone Guidelines for the effluent discharge from a copper smelter located in the Northern Sweden to the Baltic Sea for the determination of mixing zone length for the parameters of Hg, Cd, Ni, and Pb. They concluded that AA-EQS criteria are met for these pollutants within a predefined distance of 500 m from the discharge point of the effluent. However, they did not indicate the failure of the software as the upstream concentration for the pollutants in the discharge was lower than the relevant EQSs.

In tier 2, the EQS-based discharge limits to maintain AA-EQS values were also tried to be calculated simply by applying conservative mass balance indicated in Eq. 3. This methodology does not take into account the mixing zone, whereas the above-mentioned “Discharge Test Software” evaluates the acceptability of the mixing zone resulting from the discharge.

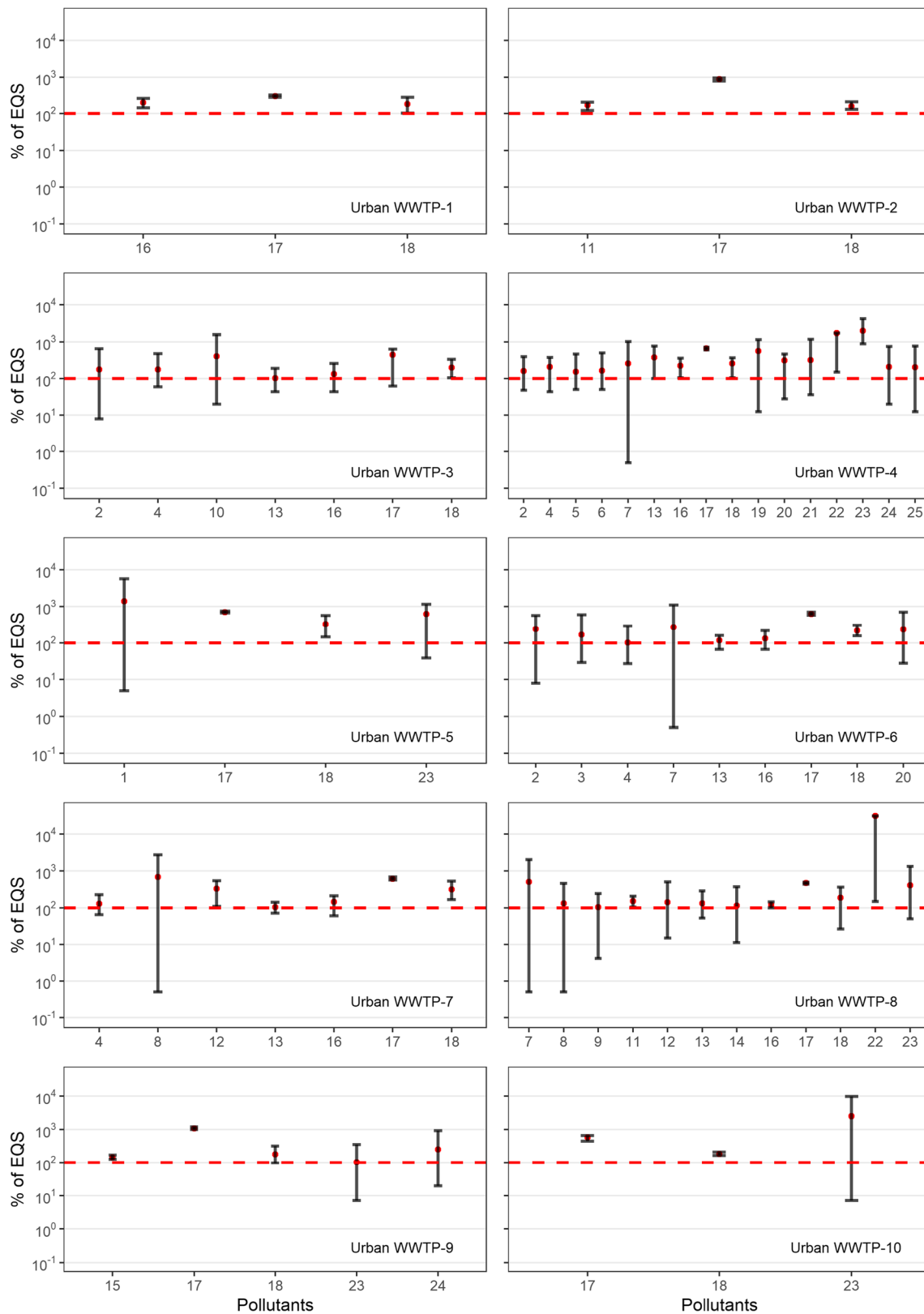


Fig. 5 Pollutants at concentrations exceeding relevant AA-EQS values in the urban WWTPs in the Gediz River Basin. (1)Dichloromethane; (2)Fluoranthene; (3)Benzo(a)pyrene; (4)Pb; (5)4-chloroaniline; (6)Azinfos-methyl; (7)Benzo(a)fluorene; (8)bis(2-ethylhexyl)

terephthalate; (9)Permethrin; (10)Tridecane; (11)As; (12)Cr; (13)Cu; (14)Sb; (15)V; (16)Zn; (17)Si; (18)TPH; (19)Azoxystrobin; (20)Chlorantraniliprole; (21)Chlorfenapyr; (22)Cyfluthrin; (23)Imidacloprid; (24)Nicosulfuron; (25)Tolfenpyrad

The result was a failure since the upstream concentration of several of the pollutants in wastewater discharges was higher than the relevant AA-EQS values. The failure was also possibly due to the flow rate data used in these calculations. For wastewater discharges, the average of the four seasonal flow rate measurements at the nearest upstream station was used as the upstream river flow rate. The flow to be added as input for the mixing zone should be median, average, Q_{90} , or Q_{95} flow. This value can only be determined by statistical evaluation of long-term flow measurements; however, in this study, only four measurements were made, which are not sufficient for assigning the average flow. Historical flow measurement is required to characterize the flow of water moving down the stream accurately. Unfortunately, there is very limited historical streamflow data in Turkey as in most developing countries (Keller et al. 2014), and our calculations were limited to the stream flow rate data collected from the field study.

Another approach tested was the determination of water-quality-based discharge limits based on a DF. This approach is a simple but useful approach that may be used in the absence of monitoring data. It assumes a conservative movement of pollutants and therefore tends to predict higher than actual concentrations for decaying pollutants. Moreover, this approach depends only upon the flow rate of the upstream river rather than both the flow rate and water quality. The DFs and discharge limits calculated by the application of five alternative DF calculation scenarios (Table 1) for all the pollutants with a PC higher than 2% are given in Online Resource 12. Table 2 presents the discharge limits calculated for Zn as an example. Zn was identified as one of the three significant pollutants in the basin with PCs higher than 2%. As shown in Table 2, the DFs calculated for Zn in ten different wastewater discharges in five different subbasins with the use of five different scenarios were different. With the applications

of scenarios 1, 2, and 4, it was seen that the DFs were almost the same for the effluents within the same subbasin. However, with scenario 3, the DFs were independent of the subbasin. The DFs calculated for the effluent discharges in subbasin 9 varied from 1.04 to 19.53. This indicated that subbasin-based DFs following scenario 3 could not be applied. On the other side, according to scenario 1 and scenario 2, the DFs are always determined based on the stream flow rate at the upstream of the water body that receives the discharge. This is, in fact, strictly conservative and would set very low discharge limits (Table 2), because relatively low upstream flowrate of the relevant water body before all the discharges is taken into account. For a comparison between scenario 2 and scenario 3, if the industrial WWTP-08 in subbasin 9 is considered as an example, it is seen that the DF values are 1.01 and 19.53, for the scenarios 2 and 3, respectively. This significant difference in the calculated DFs indicates that scenario 2 is much more conservative than scenario 1, and results in a 10 times stringent discharge standard for Zn in the industrial WWTP effluent. This is again due to its dependency on the stream flow rate at the upstream of the water body that receives the discharge. On the other hand, in scenario 3, DF is calculated based on the effluent’s flow rate and the stream flow rate upstream of the effluent. For scenario 5, all sources received a common DF of 10, which might be an oversimplified approach. Considering all these evaluations, it was considered that scenario 3 was the most appropriate for determining water-quality-based discharge limits for wastewater discharges. Since scenario 3 used the ratio of effluent flow rate to the total flow rate in the river after discharge to calculate DF, the calculated DF was different for each discharge. However, on some special occasions where the effluent flow rate was vastly lower than the river flow rate, the DF or the discharge limit calculated might have to get unreasonably high. To overcome this situation, it was

Table 2 Discharge limit and dilution factor values calculated for Zn discharges in the Gediz River Basin according to alternative scenarios (AA-EQS_{Zn} = 5.90 µg/L)

ID	Subbasin	C _{effluent} (µg/L)	C _{upstream} (µg/L)	Discharge limit (µg/L) (dilution factor)				
				Scenario 1	Scenario 2	Scenario 3	Scenario 4	Scenario 5
Industrial WWTP-01	9	1646	70	6 (1.03)	6 (1.01)	62 (10.54)	8 (1.40)	59 (10)
Industrial WWTP-08	9	2931	1416	6 (1.03)	6 (1.01)	115 (19.53)	8 (1.40)	59 (10)
Industrial WWTP-13	9	1320	1416	6 (1.03)	6 (1.01)	22 (3.73)	8 (1.40)	59 (10)
Industrial WWTP-18	9	199	4144	6 (1.03)	6 (1.01)	8 (1.40)	8 (1.40)	59 (10)
Industrial WWTP-21	9	906	175	6 (1.03)	6 (1.01)	22 (3.78)	8 (1.40)	59 (10)
Industrial WWTP-32	3	135	58	8 (1.38)	8 (1.35)	8 (1.38)	8 (1.38)	59 (10)
Industrial WWTP-36	7	772	105	228 (38.62)	79 (13.42)	18 (3.13)	14 (2.40)	59 (10)
Industrial WWTP-39	10	186	154	100 (16.89)	56 (9.54)	5 (1.02)	6 (1.02)	59 (10)
Industrial WWTP-42	4	1922	–	6 (1.01)	6 (1.01)	6 (1.00)	6 (1.04)	59 (10)
Urban WWTP-6	4	106	189	6 (1.01)	6 (1.01)	6 (1.04)	6 (1.04)	59 (10)

decided to modify the methodology in scenario 3 according to the value of the calculated DF. If the calculated DF is smaller than 10, the DF calculated will be directly used, and the discharge limit will be set as the DF times the EQS value. If the DF is higher than 10, the discharge limit will be independent of the DF and calculated as 10 times of the EQS.

The estimated discharge limits for Zn in the industrial effluents in the basin and a comparison with the current technology-based discharge standards are given in Online Resource 13. The Zn discharge standard given in the Turkish Regulation on Water Pollution and Control (RWPC 2004) for different sectors varies between 3000 and 5000 $\mu\text{g/L}$, and the effluent Zn concentrations measured in the industrial discharges are lower, and therefore meet the standard. However, the current technology-based discharge standards of 3000–5000 $\mu\text{g/L}$ are high when compared to the estimated water-quality-based discharge limits. In this case, the gradual reduction of Zn discharges is suggested, taking into account the applicability of the facilities such as passing to cleaner production techniques.

A similar study for the Brazilian standards focusing on water quality and effluent discharge was done by von Sperling and de Lemos Chernicharo (2000). For water-quality-based discharge limits, different dilution ratios were examined in terms of river/discharge flow as 1/10, 1/1, 10/1, and 100/1. Another study by Keller et al. (2014) investigated worldwide river concentrations of any chemicals that came from sewage treatment plants (such as pharmaceuticals) by using dilution factors (the ratio of the volume of freshwater and domestic sewage discharge). For determining dilution factors, national domestic effluent productions, and monthly and annual river flows were considered. According to their results, the DF of 40 and below was accepted as at risk. Low DFs were explained by low runoff, high population density, or their combination. Most countries having low DFs (< 10) were located in North Africa and the Middle East, while the countries having high dilution factors were in North and South America. Dilution ratio for Turkey was defined as 40 that to be recommended to prevent risk (Keller et al. 2014). However, due to our monitoring results, especially for the Gediz River Basin that is located in the Aegean Side of Turkey, the water quality is so poor even no dilution effect could be seen.

Consequences for the implementation of the proposed strategy

The strategy developed in the present study, for the implementation of water-quality-based discharge limits for the regulation of hazardous substances, has the advantage that it considers the response characteristics of the receiving environment directly due to the discharge and therefore places responsibility on the discharger. However, on the other side, as compared to the implementation of technology-based standards, it

may create an additional burden for the competent authorities because of a long-term flow rate and water quality monitoring requirement. As is well known, monitoring is often difficult and costly to perform (Jirka et al. 2004) for the competent authorities which are not adequately structured or sufficiently equipped (von Sperling and de Lemos Chernicharo 2000).

The difficulties in the setting up and implementing the water-quality-based standards by developing countries have been discussed by several studies (Johnstone and Horan 1996; Johnstone 2003; von Sperling and de Lemos Chernicharo 2002; Abbaspour 2011). The common challenges were indicated as limited financial resources and lacking institutional capacities, which were mainly linked to inadequate strategies followed in implementing water quality standards. When a strategy involves stringent water quality targets to be achieved with the implementation of water-quality-based discharge limits, the intended water quality objectives may not be met. The implementation of water-quality-based limits is complex, and consideration of all discharges in the water body is needed. Therefore, a stepwise implementation of water-quality-based discharge limits is recommended for developing countries considering these difficulties (von Sperling and de Lemos Chernicharo 2002; Ragas et al. 2005).

In our study, the major limitation for the application of the mass balance approach for discharge limits determination based on water quality was the insufficiency of long-term flow data. Moreover, the higher concentration of pollutants in the upstream than the relevant EQSs limited the application of the mass balance approach. Based on the findings presented for the determination of water-quality-based discharge limits, it is suggested for other countries addressing the similar problems, as the first step, to determine which parameters are above the acceptable concentrations in the receiving environment and to establish sustainable and correct monitoring networks across the river basins, as well as to collect long-term flow rate and water quality data. As the further step for the implementation of the water-quality-based discharge limits is proposed by starting with the dilution methodology considering dilution at the immediate discharge point. After having sufficient long-term flow rate data, a mass balance approach without considering the mixing zone can be applied. Since water quality improves after revised discharge limits application, it is recommended to use programs by considering mixing zone such as the Discharge Test Software.

Conclusions

In this study, it was aimed to develop a strategy for a transition to water-quality-based discharge limits from the existing technology-based discharge standards. Within this scope, the Gediz River Basin was selected as a pilot basin, and a seasonal monitoring study was conducted. The results indicated that the

implementation of technology-based discharge standards failed to achieve the target water quality standards in the river, and the concentrations of several priority substances and river basin specific pollutants exceeded the EQSs. In order to develop a strategy for Turkey for a transition to the water-quality-based standards from the technology-based discharge standards, alternative approaches were tested, and the following conclusions were drawn:

- The presence of inadequate long-term accurate streamflow data is one of the difficulties limiting the application of the mass balance approach in setting up water-quality-based discharge limits.
- The application of conservative mass balance with or without mixing zone is not realistic in water bodies where the concentration of a pollutant is higher than the relevant EQS.
- The recommended strategy involves
 - The establishment of sustainable and correct monitoring networks across river basins for the collection of long-term flow rate and water quality data,
 - A stepwise approach to the implementation of a water-quality-based discharge limits starting with the dilution factor methodology that sets water-quality-based discharge limits based on the level of dilution that occurs at the immediate discharge point.
 - The use of mass balance approach without considering mixing zone as the second and the use of programs such as the “Discharge Test Software” considering mixing zone as the last stage.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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