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Methodology for the definition of the mixing zones of punctual discharges in coastal waters

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Abstract

The European Union allows the designation of mixing zones (MZ) adjacent to a punctual discharge. Concentrations of one or more substances could exceed the environmental quality standards in the MZ if the rest of the water body meets the standards. However, the European Union does not explain how to define it. In this paper, the simulation of the MZ is done through two free software, Discharge Test and Visual Plumes. Discharge Test is used to perform a first analysis of the proposed scenario and to assess the discharge effects on the receiving environment. Visual Plumes is used to analyze in more detail these effects and the behavior of the polluting plume. The results obtained, for a mercury discharge from an outfall that discharges into the coastal zone of the Mediterranean Sea, show that these models could be a useful tool for the determination of MZ. A methodology is defined to delimit the MZ for an outfall. This MZ will be the circular surface, with a radius equivalent to the estimated distance, around the discharge point.

A mixing zone (MZ) is the area of an aquatic ecosystem where pollutants from punctual discharge mix with cleaner water. In this zone, dispersion occurs in all directions until the contaminants achieve uniform concentrations in the receiving ecosystem. The MZs are defined in Article 4 of Directive 2008/105/EC, "Member States may designate mixing zones adjacent to points of discharge. Concentrations of one or more substances listed in Part A of Annex I may exceed the relevant Environmental Quality Standards (EQS) within such mixing zones if they do not affect the compliance of the rest of the body of surface water with those standards."

The main objectives for defining a MZ are:

- It is allowed to exceed the EQS in an area near the point of discharge if the water body complies with the EQS.
- The areas where EQS can be exceeded are accepted and registered. The MZ that are designated must be included in the river basin management plan. Thus, it is possible to control more exhaustively the areas where the EQS are exceeded.
- Efforts are focused on compliance with the EQS. If the area where the standard is not met is limited and is relatively small, it will be much easier and more feasible to search for and find solutions.

In December 2010, the European Commission published "Technical Background Document on the Identification of Mixing Zones" (CIS-WFD 2010*a*) and "Technical Guidelines for the identification of mixing zones in application pursuant to Art. 4(4) of the Directive 2008/105/EC" (CIS-WFD 2010*b*). In these documents, several methodologies are developed for the designation of MZ in the different scenarios that can occur in a situation of discharges that exceed the norm. CIS-WFD (2010*b*) indicates "A mixing zone is designated by the Competent Authority as the part of a body of surface water which is adjacent to the point of discharge and within which the concentrations of one or more contaminants of concern may exceed the relevant EQS, provided that compliance of the rest of the surface water body with the EQS is not affected." This document helps the Competent Authorities to decide if the definition of MZ is necessary. It also helps to define its size and accessibility, through a "step strategy." This strategy will allow applying the degrees of detail and control appropriate for the scenario presented. In general, this has been the basis for the delimitation of the MZ of the existing works on the subject (Çeka 2012; Rodríguez 2016; Campos et al. 2022).

Some countries have developed their methodology based on the guidelines of the European Commission to facilitate the designation of the MZ to the competent authorities (Bleninger and Jirka 2011). Italy allows designating MZ to

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autonomous regions and provinces but does not establish concrete guidelines. In France, the MZ assignment criteria are defined, and a guide establishes to calculate it from the real data of the discharge and the receptor ecosystem with the help of software. In Portugal, the concept of MZ has been introduced but specific guidelines have not been set. In Scotland, the Scottish Environmental Protection Agency (SEPA) has established guidelines on the modeling of discharges in coastal and transitional waters (SEPA 2006), but they do not focus on MZ. In the Czech Republic, a methodology and a software tool for the calculation of the MZ called "The Czech Pollution Test" has been developed (Micanik et al. 2012).

Latvia and Lithuania manage the MZs of the transboundary territories and they have developed HOTRISK, a project focused on the harmonization of water management and the risk of pollution. In this project, based on the technical guidelines, the tool "Discharge Test" is used for transboundary waters (LEGMC and LEI 2014).

Denmark has opted for a fixed distance starting from the initial dilution zone. For coastal waters, 50–100 m are established from the point of discharge (Liefferink et al. 2011).

Netherlands defines a maximum permissible length of the MZ, for water bodies of linear type, based on the width of the water body, 10 × width, with a maximum of 1000 m. For the coastal waters, a maximum volume is defined, as a length of 150 m in deep coastal waters (Rodríguez 2016; Skorbilowicz et al. 2017).

Austria limits the length to 1000 m, for water bodies up to 100 m wide, and to 10 × Width if this is higher (Rodríguez 2016; Skorbilowicz et al. 2017).

In Spain, Real Decreto 60/2011 (now abolished) on EQS in the field of water policy, defined the concept of MZ. If an MZ is defined, the Real Decreto 60/2011 require to reflect it in the corresponding Basin Hydrological Plan, and strategies to reduce its surface area in the future must be defined. This regulation did not explain how to define the MZ. Nowadays, Real Decreto 817/2015 establishes the criteria for monitoring and evaluating the state of surface water and EQS, and it defines in Article 3.47 the MZ as the "Zone adjacent to a point of discharge where the concentrations of the different constituents may not correspond to the regime of complete mixing of the effluent and the receiving water body." Thus, in Spain, the regulation of MZ is not too delimited nor sufficiently standardized. This regulation only establishes that the designation of the MZ (which should be limited to the vicinity of the discharge point) must be included in the Basin Hydrological Plan in which the discharge is made. It should include a description of the approaches and methods applied to define such zones, as well as the measures adopted to reduce the extension of such MZ in the future. So, in Spain, only three Hydrographic Demarcations have included in some way the concept of a MZ in their Basin Plan, although without a specific definition. These Basin Plan are:

- The Miño-Sil Hydrological Plan introduces the MZ in a purely theoretical way and, for the moment, does not have any practical application.
- The Guadalquivir Hydrological Plan defines a specific value for the MZ without considering any characteristic of the receiving ecosystem: type, width, length, currents, type of priority substance or contaminant, etc. It defines MZ for the discharges in a river of wastewater treatment plants (WWTP) as 100 m downstream of the point of discharge. It does not define the methodology and focuses on WWTP discharges in a river.
- The Management Plan of the River Basin District of Catalonia defines MZ as a fixed value for emissaries discharged at the coast (circumference with a radius of 50 m) and for direct discharges into rivers (50 m downstream from the point of discharge).

However, these definitions or designations of MZ are not adapted to Directive 2008/105/EC. There are not descriptions and/or explanations of the applied approaches and methods to delimit it. Furthermore, the definition of MZ with fixed values is inaccurate, and it may not make sense because it may be too large or insufficient. Really, the study of submarine outfalls cannot focus only on the area near the discharge. The area to be studied should be extended to an adjacent area of the discharge pipe, with an extension that depends on wastewater discharge, pollutant load, marine currents, and typical winds of the area (Mossa 2006).

Thus, the objective is to define a methodology to determine the MZ of a specific discharge to coastal waters, in compliance with the EQS of Real Decreto 817/2015 and based on the guidelines of Directive 2008/105/EC for MZs. For this purpose, the steps described in the European Commission document "Technical guidelines for the identification of mixing zones" (CIS-WFD 2010*b*) will be considered.

Materials and procedures

Procedures

For MZ identification, the guidelines set by the European Commission (CIS-WFD 2010*b*) are followed. First, it must be determined if MZ needs to be defined. Second, its size and admissibility must be defined. For this, the "step strategy" is used and it allows for applying the appropriate level of detail and control. It is divided into five levels:

- Level 0—Determination of the presence of risk pollutants. This level is designed to identify the presence of punctual discharges capable of causing non-compliance with the EQSs.
- Level 1—Preliminary analysis. This step determines if the discharges, identified at level 0, should be subject to new considerations. This can be done by applying simple tests, to exclude safe discharges from other more advanced studies.

- Level 2—Simple study of the MZ. Its purpose is the elimination of those discharges that belong to the categories of admissible or inadmissible.
- Level 3—Detailed evaluation of the MZ. Its objective is the evaluation in detail of the most complex cases.
- Level 4—Scientific study. This step will avoid doubts that may exist after analyzes conducted in the previous levels. A scientific study will be capable of validating the results, refining the methods applied, or describing in detail the impacts derived from the EQS.

For efficient discharge management, it is important to properly define the MZ generated. The MZ will depend on the hydrodynamic conditions, geomorphology, and bathymetry of the receiving ecosystem (Rodríguez et al. 2016). Therefore, the delimitation of the MZ must consider these factors, and the use of mathematical models is needed.

Several models can help to apply the strategy proposed in the technical guidance document (CIS-WFD 2010*b*). These could be CORMIX, VISUAL PLUMES, VISJET, MOHID, DISCHARGE TEST… (Doneker and Jirka 2002; Etemad-Shahidi et al. 2004; SEPA 2006; Etemad-Shahidi and Azimi 2007; Loya-Fernández et al. 2012; Palomar et al. 2012*a*,*b*).

In this paper, two free software are used, with complementary purposes: DISCHARGE TEST and VISUAL PLUMES.

Discharge Test is a computer program of the Ministry of Environment of the Netherlands, and it has been developed by Deltares Institute. It is based on the technical guidelines document of the European Commission (CIS-WFD 2010*b*). This evaluates the first three levels, 0, 1, and 2, and it helps to determine if a discharge is clearly admissible (i.e., other more precise or detailed analyses will not modify this conclusion) or if, on the contrary, it is inadmissible. If all the proposed zones are clearly admissible, the designation of the MZ may proceed without the need for further studies. This software determines the concentration near the discharge point. It evaluates whether the concentration at the edge of the MZ, a limited area near the discharge point, meets the EQS standard. It determines if an increase in the concentration does not lead to a significant deterioration of water quality.

Visual Plumes is a computer program developed by the United States Environmental Protection Agency. It is designed to simulate the behavior of a discharge, its movement, and the contaminant concentrations present in it. It integrates several simulation models (NRFIELD, DKHW, UM3, PSDW, FRFIELD) (Baumgartner et al. 1994; Frick et al. 2002, 2003, 2007; Frick 2004). It is useful for predicting plume dilution and physical properties, in the MZ and in the far field (Frick 2004; SEPA 2006; Hunt et al. 2010; Bottelli 2011; Loya-Fernández et al. 2012; Muhammetoglu et al. 2012).

Discharge Test could be used to perform a first analysis of the proposed scenario (levels 0, 1, and 2). It is useful for making a first assessment of the discharge effects on the receiving environment. Visual Plumes could be used to analyze in more detail the effects of the discharge on the receiving medium and the behavior of the polluting plume (level 3). This will help to understand the discharge behavior, and to analyze which are the variables that most influence the definition of the MZ, its extent, scope, concentrations, and so forth.

Study case

European Environment Agency (2018) points out that only 38% of surface waters (rivers, lakes, and transitional and coastal waters) are in good chemical status. Its report indicates that some Member States have gotten a poor chemical status due to a few priority substances, the most common being mercury. A total of 45,973 water bodies in 24 European Union Member States do not achieve a good chemical status for mercury (European Environment Agency 2018). The inputs of urban WWTP lead to the contamination of more than 13,000 water bodies with polyaromatic hydrocarbons, mercury, cadmium, lead, and nickel (European Environment Agency 2018).

Annex I of Directive 2008/105/EC and Directive 2013/39/EU, indicate the EQS values for the different priority substances, and on which it is possible to define an MZ. The priority substance chosen for this study is mercury because it is a highly polluting substance, its EQS are quite restrictive and high concentrations have been found in some analyses of effluents from different treatment plants. For mercury and its compounds (CAS number 7439-97-6) in "Other surface waters," the EQS marked in Annex I of Directive 2008/105/EC and Directive 2013/39/EU are annual average 0.05 μg L^{-1} and maximum allowable concentration (MAC) 0.07 μg L^{-1}.

For the MZ definition, it is necessary to reproduce the most adverse situation in environmental aspects. Thus, the following premises have been assumed for the choice of "the worst-case approach (discharge/concentration)":

- The contaminant presents a conservative behavior. The decay rate is considered null. Background concentrations are assumed null in the receiving medium.
- The discharge volume is the maximum expected for the WWTP. The peak flow of the design of the emissary is taken as flow.
- The ambient current in receiving ecosystem is uniform, with velocity and direction equal in depth.

During non-stratified winter conditions, the effluent concentration can climb along the water column. However, during stratified summer conditions, the released effluent can rise and become trapped beneath the pycnocline (Signell et al. 2000; Lucas and Kudela 2017). So, different annual periods are considered in the receiving environment and in the discharge:

- Summer: Stratified environment and summer discharge temperature.

- Winter: Non-stratified environment and winter discharge temperature.

Different scenarios are also considered, with different velocity ranges and current directions.

The study is conducted for a discharge coming from a WWTP located in the Valencian Community (Spain). The treated wastewater is discharged to the Mediterranean Sea, through a submarine outfall formed by 25 diffusers, of 120 mm of diameter, separated 2.5 m. It discharges at a distance of 2 km from the coast, at 17 m depth, and with an angle of 0.461° over the bottom. It has an orientation of 59°N sea-land (being 0 the north).

WWTP data have been obtained from the Spanish reports to the European Commission in compliance with the Urban Wastewater Treatment Directive (European Environment Agency 2017). It is designed for 280,000 inhabitants-equivalents with an inflow of 269,921 inhabitants-equivalents. The project flow is 60,000 $m^3 d^{-1}$.

Based on the historical data available on the effluents of different treatment plants in the Valencian Community (own works), the characteristics of the discharge have been selected. A mercury concentration (Hg) of 8 $\mu g\ L^{-1}$ (maximum of all the concentrations of the WWTPs) has been chosen. Effluent salinity is 1.4 psu, and the temperature is considered 25.20°C in summer and 19.67°C in winter (averages of all WWTPs).

The direction of propagation of the current in the study area with real data from state ports between 2005 and 2018 fluctuates in the entire range from 0°N to 359°N, being 0 the north (Puertos del Estado 2018). The minimum velocity of the current found in the study area is 0.01 $m\ s^{-1}$. The monthly maximum velocity between 2005 and 2018 in the study area ranges from 0.14 $m\ s^{-1}$ in February 2008 to 1.0 $m\ s^{-1}$ in October 2010. The monthly average velocity between 2005 and 2018 in the study area ranges from 0.05 $m\ s^{-1}$ in February 2008 to 0.33 $m\ s^{-1}$ in August 2017.

For the environment, two scenarios have been proposed, stratified in summer, and not stratified in winter. In the area of the discharge point, the water temperatures at each depth, for both scenarios, are shown in Table 1, with a thermocline at 10–12 m depth in summer. Salinity is considered constant in depth (37.5 psu).

Table 1. Temperature (°C) profile for each scenario.

Depth (m)	Stratified	Non-stratified
0	28.5	17
1	28.3	17
5	28	17
10	28	17
12	20	17
15	20	17
18	20	17

In each of these scenarios, different cases are studied according to the current (velocity and direction). The current velocity is defined from 0.002 to 1.0 $m\ s^{-1}$ (0.002, 0.01, 0.05, 0.1, 0.15, 0.25, 0.3, 0.5, 0.7 and 1.0 $m\ s^{-1}$). The current direction will cover all possible directions (0°N, 90°N, 180°N, 270°N, 59°N, 149°N, 239°N, and 329°N). All these parameter combinations yield 80 cases for each scenario.

Assessment

As indicated in the Technical Guidelines of the European Commission (CIS-WFD 2010*b*), the first step is to know if the discharge falls into the category of admissible or completely inadmissible. To do this, Discharge Test is used. This application performs basic calculations. It determines if the concentrations in the calculated MZ are acceptable or if on the contrary a more advanced study is needed to determine this aspect. Discharge Test does not consider the directions of the current or the discharge direction of the outfall.

It is important to highlight that, in Discharge Test, multiport diffusers are not taken into consideration and a single round discharge opening is therefore assumed. If the outfall discharges with a multiport diffusor, the software recommends an approximation, in which the total surface area of the ports (Opp) is used. The diameter can be derived from 2 × sqrt (Opp/pi). In our case, the diameter of the discharge pipe will then be 0.60 m.

To define if it is possible to delimit an MZ in a specific case, Discharge Test checks two aspects:

- Concentration at a distance from the discharge point of 0.25 × depth of the water body (m) (4.70 m in this case). This is compared with MAC (0.07 $\mu g\ L^{-1}$). These concentrations are shown in columns 2 and 5 of Table 2.
- Concentration at a distance from the discharge point of 10 × depth of water body (m) (188.06 m in this case). This is compared with EQS (0.05 $\mu g\ L^{-1}$). These concentrations are shown in columns 3 and 6 of Table 2.

If the first concentration is higher than MAC or the second one higher than EQS, defining MZ would be ruled out.

Discharge Test allows the user to redefine the distance to study. So, for this case studied, the distance can be modified, and it can be checked where the concentration is lower than the EQS. (0.07 $\mu g\ L^{-1}$). This distance is shown in columns 4 and 7 of Table 2.

As the current velocity increases, the mercury concentration at 188.06 m decreases in both scenarios, and the distance necessary to reach concentrations below 0.07 $\mu g\ L^{-1}$ is smaller. However, for a velocity higher than 0.7 $m\ s^{-1}$ in a stratified environment and 0.5 $m\ s^{-1}$ in a non-stratified one, the distance increases when the velocity is increased. Furthermore, there is a slight increase in concentration at the highest velocity (1.0 $m\ s^{-1}$) in the non-stratified environment.

Table 2. Concentrations and distances reached for the different current velocities in both scenarios (Discharge Test).

	Stratified			Non-stratified		
	4.70 m	188.06 m	<0.07 μg L^{-1}	4.70 m	188.06 m	<0.07 μg L^{-1}
Velocity (m s^{-1})	Conc (μg L^{-1})	Conc (μg L^{-1})	Distance (m)	Conc (μg L^{-1})	Conc (μg L^{-1})	Distance (m)
0.002	2.49	2.077	> 25,000	1.96	1.652	> 25,000
0.01	1.21	0.696	> 25,000	0.86	0.502	> 25,000
0.05	0.87	0.317	5112	0.58	0.231	1520
0.1	0.77	0.219	1440	0.51	0.119	455
0.15	0.68	0.143	670	0.68	0.092	257
0.25	0.90	0.081	224	0.90	0.052	131
0.3	0.96	0.067	178	0.93	0.042	102
0.5	1.11	0.042	106	1.15	0.028	74
0.7	1.27	0.033	92	1.27	0.025	85
1.0	1.41	0.029	105	1.37	0.032	108

Hg concentration is lower than 0.05 μg L^{-1}, at 188.06 m, at velocities higher than 0.5 m s^{-1} in a stratified environment, and at 0.3 m s^{-1} in non-stratified. However, at 4.70 m, the Hg concentration is always greater than 0.07 μg L^{-1} in both scenarios. Therefore, in no case, the concentration of mercury is less than the MAC, so it is determined that the MZ is not acceptable and that appropriate measures must be taken or asked for advice in this regard.

When an effluent is discharged, a turbulent mixing process occurs, and that contributes to the dilution of pollutants. In the zone closest to the discharge (near field, zone of initial dilution or zone of hydrodynamic mixing) the effluent is diluted relatively quickly, due to its initial momentum (i.e., its discharge velocity) and to the mixing induced by the buoyancy of the discharge. The zone of initial dilution ends when the plume reaches the surface or reaches a depth at which its density equals that of the medium and it becomes trapped.

Outside this initial zone, the mixing process of the discharge will continue at a rate determined mainly by advective and turbulent mechanisms, characteristic of the medium (and therefore of the velocity) and independent of the discharge parameters. It is the far field (Schnurbusch 2000; Suh 2001).

Although the simulation of the behavior of a discharge in the near field can be done in a relatively simple way, the modeling of the far field requires more detailed information of the receiving medium (e.g., realistic fields of currents and wind, …) and more sophisticated analysis tools (e.g., physical models, or 3D numerical models).

The results obtained with Discharge Test (levels 0, 1, and 2), raise the need for a detailed evaluation of the MZ (level 3) and later perhaps a Scientific study (level 4). To do this, and to define more specifically the MZ, Visual Plumes (with the UM3 simulation model) is used.

Discharge Test calculates the concentrations at a distance from the discharge point of 0.25 × depth of the water body (m) and at a distance from the discharge point of 10 × depth of water body (m). The user can calculate the concentration at a specific and selected distance, too. But this software is only used for a preliminary study. Instead, Visual Plumes provides concentrations across the entire continuum (vertical and horizontal) and facilitates the spatial (geographical) study of the plume. It is very useful software for level 3.

The discharge concentration is 8 μg L^{-1}, so to reach the maximum allowed (0.07 μg L^{-1}) the minimum dilution must be higher than 114. In Visual Plumes, the current direction is included in the modeling. Therefore, there are 80 cases for each scenario (stratified and not stratified).

The Froude number is the ratio of the momentum to the buoyancy of the discharge. It gives information about the buoyancy or momentum of a plume on the elevation. If the Froude number is less than 1, buoyancy will dominate. If the Froude number is greater than 1 (between 10 and 100), the momentum will dominate, and saltwater intrusion into the diffusers will be prevented. Froude number is 13.38 for the stratified environment and 13.51 for the non-stratified, therefore the intrusion of seawater in the diffusers does not occur. The effluent velocity when leaving through the diffusers in both environments is 2.456 m s^{-1}, so it is not expected that the effluent will obstruct the diffusers.

When the effluent leaves the diffuser and enters higher-density waters, an initial dilution of the effluent is produced by mixing. The difference in velocity and density between the effluent and the receiving waters gives rise to turbulence and mixing, while the plume rises to the surface. However, the mixture is not only vertical (the phase in which we will consider that the discharge is in the "near field"). The action of the marine currents causes a dispersion of the discharge, producing its secondary dilution and transport in the "far field" (Schnurbusch 2000; Suh 2001). Stratification reduces dilution, the height of the plume, and the possibility of reaching the surface. Even in extreme conditions, stratification can suppress the mixing (Roberts and Tian 2003).

In the stratified environment, the plume does not reach the surface and it is retained at a depth between 10 and 12 m depth depending on the velocity. Two typical cases, for 0.05 and 0.7 m s^{-1} are shown in Fig. 1.

Stratification causes the plume to be retained at the thermocline, a depth where there is a sudden change in temperature (Table 1). When the plume reaches that depth, it begins to move horizontally. Figure 1 clearly shows that, when the current velocity is small, the plume rises rapidly and is retained at the thermocline, at a point remarkably close to the discharge point (10 m distance for velocity 0.05 m s^{-1}). However, when the current velocity is higher, the plume moves horizontally while ascending, and it is retained at the thermocline at a distance from the furthest discharge point (80 m for the velocity of 0.7 m s^{-1}).

The current direction is a crucial factor, and it marks where the plume moves (Fig. 1). However, when the current velocity is too small, there is no difference in the direction in which the plume moves, as shown in Fig. 1 for the velocity of 0.05 m s^{-1}. In this case, the direction in which the submarine outfall is discharging is more important. Thus, for all the modeled current directions, the plume moves mainly in the direction of 59°N. On the other hand, for higher velocities (0.7 m s^{-1}) the plume moves in the current direction.

The point where the concentration reached is less than 0.07 μg L^{-1} will delimit the MZ. However, Visual Plumes does not provide the MZ dimensions, but it does provide the "horizontal distance from the source" (distance) and the diameter of the plume at that point (P-dia). Thus, the MZ will be given by the sum of the distance and radius of the plume (distance + P-dia/2). MZ dimensions can be determined for each velocity and direction.

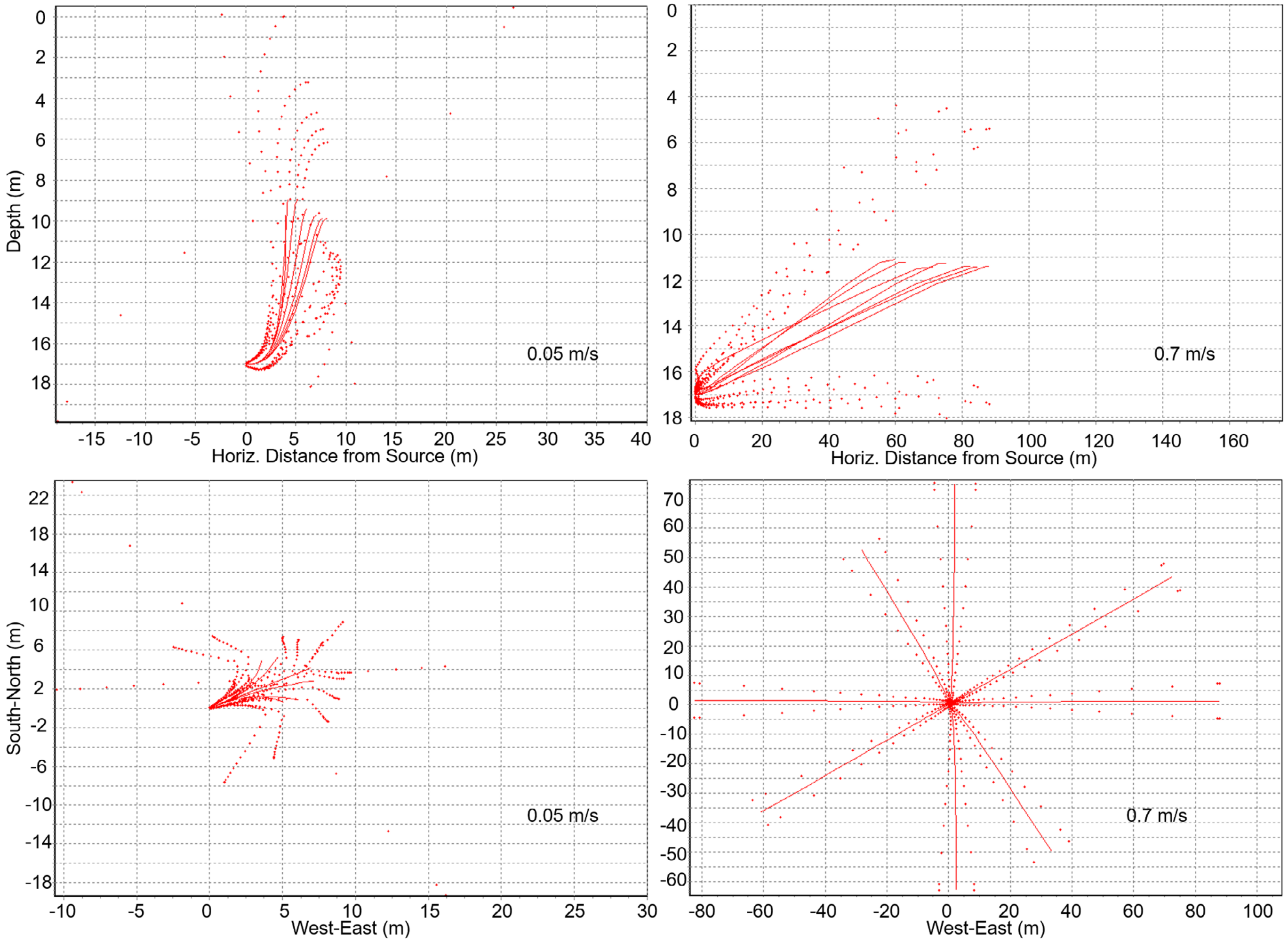

Fig. 1. Stratified environment. Centerline (solid line) and plume boundaries (dashed lines). Plume elevation and plan view for 0.05 and 0.7 m s^{-1}, for all directions. The figure shows the plume elevation and plan view for current velocities of 0.05 and 0.7 m s^{-1}, for all current directions, in a stratified environment. The centerline of the plume is shown in solid lines and the boundaries of the plume are in dashed lines.

Table 3. Mixing zone (m) for the different current velocities and directions in both scenarios (Visual Plumes).

Velocity ($m\ s^{-1}$)		Direction (°N)							
		0	59	90	149	180	239	270	329
0.002	Stratified	614.2	1152.3	987.3	423.9	614.5	1159.7	991.1	423.9
	Non-Strat	42.99	67.75	59.16	33.57	40.46	68.40	59.71	33.57
0.01	Stratified	931.6	1816.7	1481.4	662.7	941.0	1901.3	1515.6	662.7
	Non-Strat	56.8	88.95	77.92	46.30	58.47	92.55	81.38	46.29
0.05	Stratified	984.8	3091.3	1795.0	714.3	1079.4	3872.6	1877.6	714.3
	Non-Strat	13.30	13.83	13.36	151.0	339.9	6.25	533.1	151.0
0.1	Stratified	9.27	12.16	10.50	8.02	6.66	2071.2	5.10	8.02
	Non-Strat	15.26	14.66	14.26	178.8	440.7	3.82	250.1	178.8
0.15	Stratified	8.88	11.99	10.23	7.59	6.48	3.14	5.02	7.59
	Non-Strat	15.71	14.40	13.99	20.35	17.15	5.55	8.67	20.34
0.25	Stratified	9.09	12.66	10.57	7.88	6.89	4.90	5.89	7.88
	Non-Strat	15.52	14.14	13.65	20.67	17.68	6.62	9.26	20.67
0.3	Stratified	9.42	13.20	10.95	8.13	7.38	5.70	6.52	8.13
	Non-Strat	15.69	14.34	13.56	20.77	17.59	7.07	9.71	20.77
0.5	Stratified	11.14	15.83	12.90	9.97	9.32	8.84	8.97	9.97
	Non-Strat	17.32	16.16	14.53	22.66	18.86	9.10	11.64	22.65
0.7	Stratified	12.98	18.61	15.01	11.59	11.18	11.64	11.34	11.59
	Non-Strat	19.25	18.77	15.89	24.55	20.29	11.79	12.43	24.56
1	Stratified	15.67	22.76	18.12	14.12	13.68	15.68	14.65	14.12
	Non-Strat	21.77	22.98	18.46	28.13	21.97	15.87	14.76	28.13

Table 3 shows, for each case, the MZ dimensions in both scenarios. For stratified environment, the MZ is much larger for the lower velocities. Clear differences are observed between velocities lower than 0.1 $m\ s^{-1}$ and higher ones. The statistical treatment (Anova) shows that there are significant differences between the velocities 0.002, 0.01, and 0.05 $m\ s^{-1}$ and the rest ($p = 0.0000$). The only exception is 0.1 $m\ s^{-1}$ with a direction of 239°N, where the MZ is higher than for the other velocities. The reason for this exception is that, at this velocity and with this direction (contrary to the discharge of the diffuser, 59°N), the plume is retained in the vicinity of the outfall and prevents its rapid dilution. It is observed that for the lowest velocities, the highest MZs are produced at this direction of 239°N. The statistical treatment (ANOVA) shows that there are no significant differences between the directions ($p = 0.3760$).

Figure 2 shows the MZ for the lowest current velocities in different directions for the stratified environment. For these velocities, the smallest MZ is for the directions 149°N, 329°N, and 0°N and 180°N. The greatest MZ is reached in the directions 59°N, 239°N, 90°N, and 270°N. The greatest MZ, that is, the greatest distance for the concentration to be less than 0.07 $\mu g\ L^{-1}$, is for the velocity of 0.05 $m\ s^{-1}$.

Figure 3 shows the MZ for current velocity ≥ 0.1 $m\ s^{-1}$ in the different directions for the stratified environment. The smallest MZ is for 0.15 $m\ s^{-1}$, for any direction, increasing this MZ with the velocity. For these velocities, there are no such clear differences in direction as observed at the smaller velocities.

In the non-stratified environment (Fig. 4), when the current velocity is small, the plume rises quickly and reaches the surface (at a maximum of 15 m distance in the case of a velocity 0.05 $m\ s^{-1}$). When the current velocity is higher, the

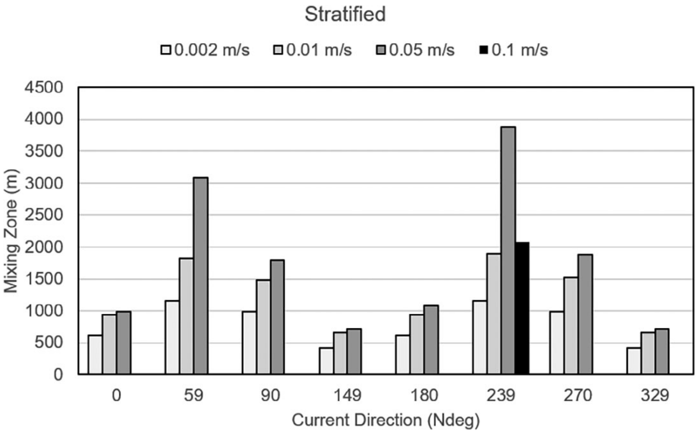

Fig. 2. Stratified environment. Mixing zone for current velocity ≤ 0.1 $m\ s^{-1}$. The figure shows the MZ for the lowest current velocities (≤ 0.1 $m\ s^{-1}$) in the different current directions for the stratified environment.

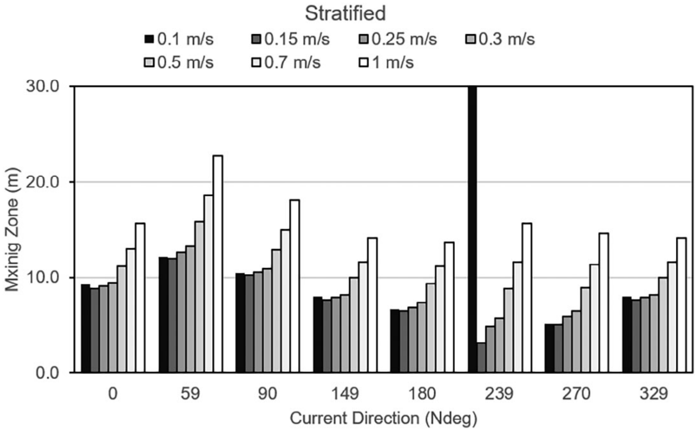

Fig. 3. Stratified environment. Mixing zone for the current velocity ≥ 0.1 m s^{-1}. The figure shows the MZ for current velocities ≥ 0.1 m s^{-1} in the different current directions for the stratified environment.

plume moves horizontally while ascending, and reaches the surface at a distance from the furthest discharge point (at a maximum of 100 m in the case of the velocity of 0.7 m s^{-1}). In addition, in this case, the direction of the current is a key factor and marks where the plume moves.

However, when the current velocity is too small, there is no difference in the direction in which the plume moves, as shown in Fig. 4 for the velocity of 0.05 m s^{-1}. In this case, the direction in which the submarine outfall is discharging is more important. Thus, for all the directions of the modeled current, the plume moves mainly in the direction of 59°N. On the other hand, for higher velocity (0.7 m s^{-1}) it is observed that the plume moves in the direction of the current.

Table 3 shows the distance from the discharge point where the concentration is less than 0.07 μg L^{-1} (MZ) for the non-stratified environment. The MZ is greater for 0.05 and 0.1 m s^{-1} with directions 149°N, 180°N, 270°N, and 329°N, but without major differences between them. The statistical

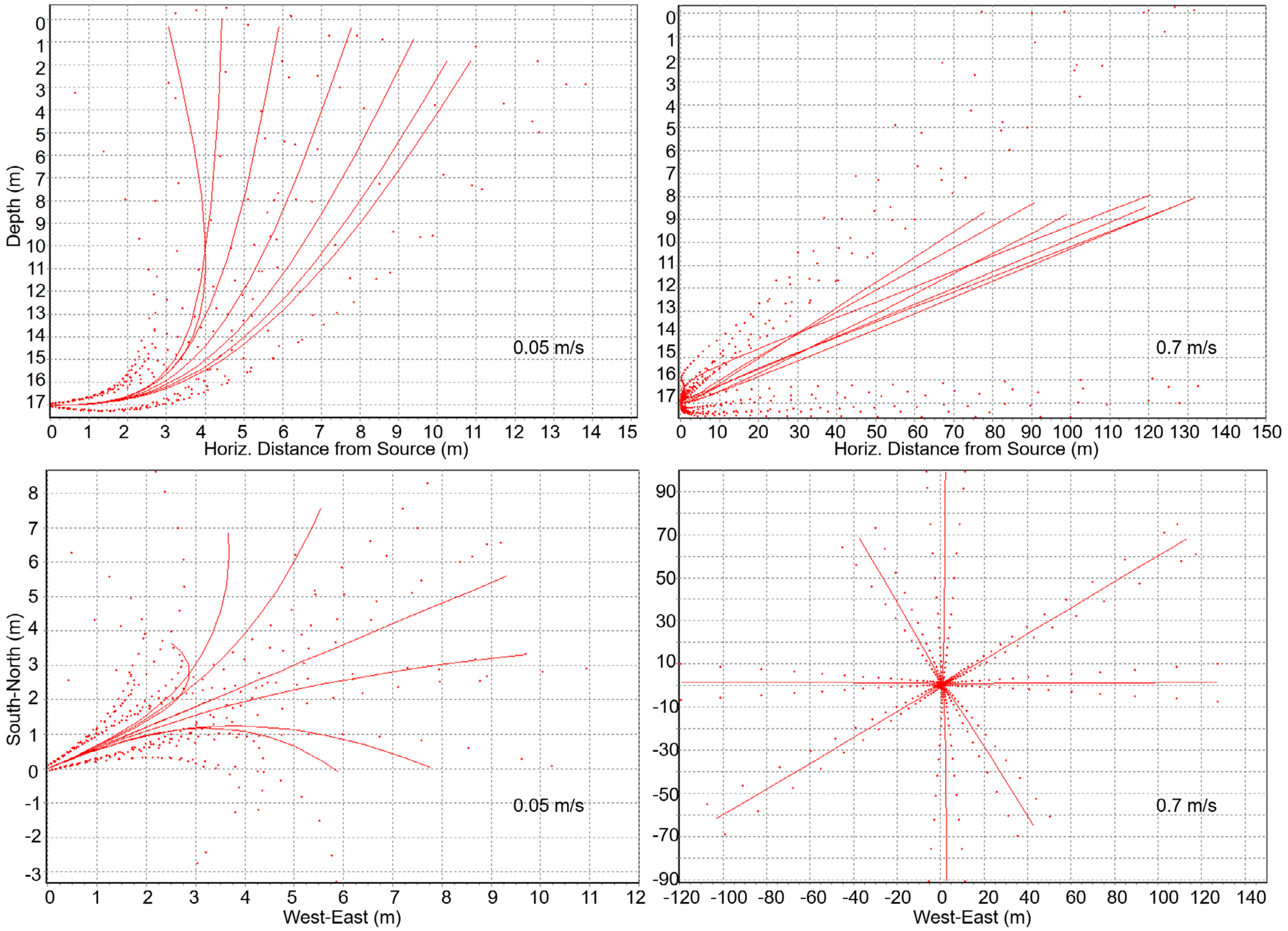

Fig. 4. Non-stratified environment. Centerline (solid line) and plume boundaries (dashed lines). Plume elevation and plan view for 0.05 and 0.7 m s^{-1}, for all directions. The figure shows the plume elevation and plan view for current velocities of 0.05 and 0.7 m s^{-1}, for all current directions, in a non-stratified environment. The centerline of the plume is shown in solid lines and the boundaries of the plume are in dashed lines.

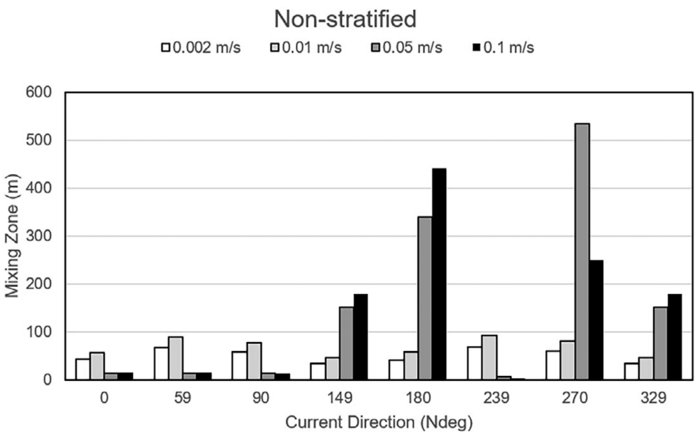

Fig. 5. Non-stratified environment. Mixing zone for current velocity ≤ 0.1 m s^{-1}. The figure shows the MZ for the lowest current velocities (≤ 0.1 m s^{-1}) in the different current directions for the non-stratified environment.

treatment shows that the distances reached for velocities 0.05 and 0.1 m s^{-1} are significantly different from the distances reached for the rest of velocities ($p = 0.0010$). However, there are no significant differences between the directions ($p = 0.2393$).

Figures 5, 6 show the MZ for each velocity and direction, for the non-stratified environment. Figure 5 shows the MZ for the lowest current velocity in the different directions. For these four velocities, the smallest MZ is reached for the directions 0°N, 59°N, 90°N, and 239°N. The greatest MZ is reached for the directions 149°N, 180°N, 270°N, and 329°N for 0.05 and 0.1 m s^{-1}.

Figure 6 shows the distance required to reach a concentration lower than 0.07 μg L^{-1} (MZ) for a current velocity greater than 0.1 m s^{-1} in the different directions. The smallest MZ is reached for 0.15 and 0.25 m s^{-1}, for any direction, increasing this distance with the velocity. For these velocities, there are no such clear differences in direction as observed at the smaller velocities. The smallest MZ are given for directions 239°N and 270°N, reaching the greatest MZ for directions 149°N and 329°N.

In the non-stratified environment, the plume reaches the surface, and the distances necessary to reach concentrations lower than 0.07 μg L^{-1} are much lower than in the stratified environment for velocities < 0.1 m s^{-1}. However, for velocities > 0.1 m s^{-1}, there are no clear differences between both scenarios, oscillating the distances between 10 and 30 m. In both scenarios, it is verified that for velocities greater than 0.1 m s^{-1}, the distance increases clearly with the velocity. However, there is no clear trend for the direction.

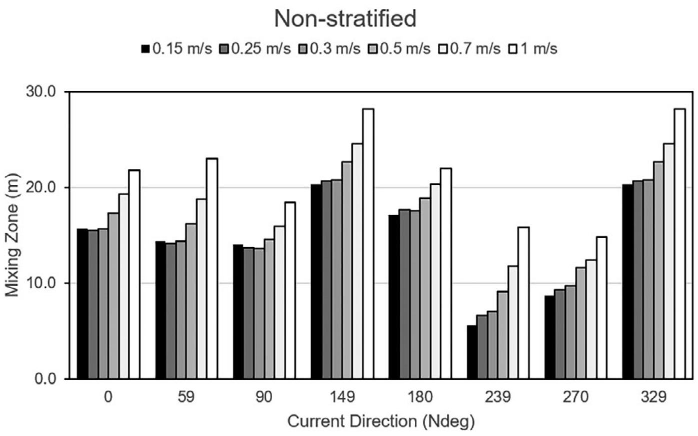

Fig. 6. Non-stratified environment. Mixing zone for current velocity ≥ 0.15 m s^{-1}. The figure shows the MZ for current velocities ≥ 0.15 m s^{-1} in the different current directions for the non-stratified environment.

Discussion

The submarine outfall generates a buoyant plume that reaches the surface when the environment is not stratified, and it is trapped in depth when there is stratification.

According to the Discharge Test, the MZ would not be admissible, and measures should be taken on the discharge to reduce the contaminant concentrations in the environment. However, Visual Plumes results conclude that concentrations lower than 0.07 μg L^{-1} can be reached at acceptable distances. There are several reasons for these differences. Both programs use quite different calculation bases. Discharge Test is based on the equations and premises presented in the Technical Background Document and in the Technical Guidelines Document of the European Commission for the identification of MZs (CIS-WFD 2010*a*,*b*). However, Visual Plumes performs the calculations based on the integration of the same equations but in the cross-section. In addition, Discharge Test does not consider the existence of diffusers and there is no possibility of indicating the current direction. And this current direction is clearly defining the distance, MZ, as it has been observed with Visual Plumes. Therefore, we can conclude that Discharge Test is useful for the study of the first levels (levels 0, 1, and 2), and it is necessary to use other models, such as Visual Plumes, to correctly define the MZ. That is, if in a specific case, the calculated concentrations were adequate (< MAC), then the MZ could be defined, and it would not be necessary to use Visual Plumes. However, in our case, the concentrations were higher than MAC, so Visual Plumes should be used for level 3.

From the analysis of the results, we can conclude that, in this case, the most suitable software for the definition of the MZ is Visual Plumes. The obtained results allow us to define the MZ. For this, the environment (stratification or not and the current, direction, and velocity) must be considered.

The MZ should not be too large. Therefore, of the 160 cases studied, those MZ greater than the 90th percentile of the 160 cases (932.49 m) are eliminated. Thus, eliminating the cases with MZ > 932.49 m, the 90th percentile of the remaining cases is 170.41 m. Therefore, the MZ for this outfall is 170 m. This MZ would represent the circular surface corresponding to a radius equivalent to the estimated distance, drawn around the discharge point.

Once the MZ is defined, following the indications of the technical guidelines document of the European Commission (CIS-WFD 2010*a*,*b*), the admissibility of the MZ must be evaluated. In section 5 of these guidelines, it is stated five key issues to be considered by the Competent Authority to consider whether the MZ is admissible or not. These questions are:

1. "Proximity—Is the extent of exceedance restricted to the proximity of the point of the discharge (concept applicable to each single point discharge) under 2008/105/EC?"

In this case, the receiving medium is the coastal water body ES080MSPFC0101 defined by European Water Framework Directive and the MZ extends 170 m from the point of discharge. The area of the coastal water body is 4.41 km^2, so it can be concluded that a circle with a radius of 170 m (0.091 km^2) is negligible (2%) and, therefore, this MZ is completely acceptable.

2. "Proportionate—Is the extent of exceedance proportionate having regard to the concentrations at the point of discharge and to conditions on emissions in prior regulations? (BAT etc.) (Concept applicable to each single point discharge)"

The emission value of Hg considered, in this case, is 8 $\mu g\ L^{-1}$. This value can be compared with the emission limit value imposed in the Decision of Execution of the Commission of 09 December 2013. It establishes the conclusions on the best available technologies (BAT) to produce Chlor-alkali in accordance with Directive 2010/75/EU of the European Parliament and of the Council on industrial emissions in BAT 2: dismantling or conversion of mercury cell plants, where the mercury releases to water, at the outlet of the treatment unit of mercury during dismantling or conversion is 3–15 $\mu g\ L^{-1}$. Considering this emission value, the considered value in our case study is also proportionate. Seeing the normative emission values of texts of different legal nature and the extension of the MZ, this is proportioned.

3. "Attainment of Good Chemical Status—Does the extent compromise the attainment of appropriate chemical status for the relevant water body under 2000/60/EC (in particular Article 4), and 2008/105/EC, (in particular Annex I Part B)?"

Considering that the extension of the MZ is insignificant about the coastal water body, a priori, this will not affect the good chemical state of the rest of the water body.

4. "Attainment of Good Ecological Status—Does the extent compromise the attainment of appropriate ecological status for the relevant water body under 2000/60/EC (in particular Article 4)?"

This point is more delicate due to the high toxicity of mercury and the sensitivity of the marine population to its ingestion. The MZ can be considered small, and it should not be a problem for marine organisms unless there is a colony or species that permanently inhabit the area of excessive concentration. In this case, a much more detailed study would have to be conducted to evaluate the consequences on the ecological state of the water body.

5. "Consistency—Is the extent consistent with requirements adopted for other point source discharges under other Community legislation (e.g., 2008/1/EC) and interplay with 2000/60/EC and 2008/105/EC?"

Directive 2008/1/EC is derogated, and it has been replaced by Directive 2010/75/EU of the European Parliament and of the Council of 24 November 2010 on industrial emissions (integrated prevention and control of pollution). Neither this Directive nor the 2000/60/EC nor in 2008/105/EC has found any requirement that is incompatible with the delimited MZ.

Once these key issues are answered, it can affirm that the MZ calculated using Visual Plumes is admissible.

Conclusions

The MZ concept appeared in the European regulatory framework in 2008, and it can be defined and considered in River Basin Management Plan. It is an area where EQS can be exceeded. Thus, the definition of the MZ for a discharge can be very useful, especially in specific cases of activities with problems in the quality of the effluents discharged into bodies of water. Therefore, it is possible to work, in a much more concrete way, on reducing the extension of the area and therefore pollution, since the areas to be controlled, and therefore manage, would be well delimited.

Discharge Test has been used to perform a first analysis of the proposed scenario, and to make a first assessment of the discharge effects on the environment. For the definition of the MZ in this particular case, Visual Plumes seems more appropriate than Discharge Test, mainly because in Visual Plumes it is possible to implement the discharge using diffusers, which is not possible with Discharge Test. Probably for other types of discharges that are done through a pipe, Discharge Test is more appropriate for the calculation of the MZ or, at least, for a first approach, because it has been specifically designed for it, applying the technical guidelines set by the European Commission.

Considering the simulation results using Visual Plumes, it has been possible to delimit the MZ, covering a distance from the discharge of 170 m.

For the delimitation of the MZ, the steps to follow are:

- To determine the worst possible case of the parameters of the discharge (concentration, flow, etc.) and the outfall (length, depth, diffusers, etc.).
- To determine the most common environmental scenarios in the area, stratification, temperature, and current field (velocity and direction).
- To define the cases to be studied, cover all possible scenarios, and conduct the modeling study.
- To determine the MZ of each case.
- To eliminate cases that exceed the 90th percentile of all scenarios.
- To calculate the 90th percentile of the remaining cases (MZ).
- To evaluate the admissibility of the MZ, following the indications of the technical guidelines of the European Commission (section 5).

All calculations have been made using free-use tools, Discharge Test, and Visual Plumes. Although there is no doubt that there are much more complete payment tools than those used, the analysis of both and their comparison allows a future user or technician to make a first approximation to a specific MZ, to know which of the two tools is the most appropriate, as well as the problems and the advantages of each of them, facilitating and speeding up your task considerably.

Data availability statement

Data used in this study is available in: Romero, I. (2022): Temperature profile for each scenario. figshare. Dataset. https://doi.org/10.6084/m9.figshare.20396349.v1; Romero, I. (2022): Results of applying Discharge Test. figshare. Dataset. https://doi.org/10.6084/m9.figshare.20396952.v1; Romero, I. (2022): Mixing zone for the different current velocities and directions. figshare. Dataset. https://doi.org/10.6084/m9.figshare.20397636.v1; Romero, I. (2022): Summary of cases. figshare. Dataset. https://doi.org/10.6084/m9.figshare.20394537.v1; Results of applying Visual Plumes: Romero, I. (2022): Velocity 0.05 and no stratified environment. figshare. Dataset. https://doi.org/10.6084/m9.figshare.20397978.v1; Romero, I. (2022): Velocity 0.7 and no stratified environment. figshare. Dataset. https://doi.org/10.6084/m9.figshare.20398398.v1; Romero, I. (2022): Velocity 0.05 and stratified environment. figshare. Dataset. https://doi.org/10.6084/m9.figshare.20398545.v1; Romero, I. (2022): Velocity 0.7 and stratified environment. figshare. Dataset. https://doi.org/10.6084/m9.figshare.20398587.v1.

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Conflict of Interest

None declared.

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