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# Estimation of the Effect of Sewage Nitrogen Discharges on Coastal Waters: Case Study from the Mediterranean Sea

Regina Temino-Boes<sup>1\*</sup>, Inmaculada Romero<sup>1</sup>, Rabindranarth Romero-Lopez<sup>2</sup>, María Pachés<sup>1</sup> and Remedios Martinez-Guijarro<sup>1</sup>

<sup>1</sup> Institute for Water and Environmental Engineering, Universitat Politècnica de València, Valencia, Spain

<sup>2</sup> Department of Civil Engineering, Universidad Veracruzana, Xalapa, Mexico  
reteboe@doctor.upv.es

## Abstract

Sewage discharges through marine outfalls are an important source of nutrients to marine waters, which cause undesired impact such as eutrophication. However, few authors have evaluated the contribution of wastewater disposal to nutrient concentration in coastal waters. We estimated how wastewater treatment plant (WWTP) discharges alter ammonium concentrations in coastal waters of the Western Mediterranean Sea. Data obtained from the literature and from the local government was used to formulate a 1D mathematical model which predicts ammonium concentrations along coastal waters with current direct discharges. The estimations were validated by comparing them to measured data and a significant agreement was found ( $R^2=0.91$ ). Then, the simulation of a scenario with no anthropogenic direct discharges was carried out to determine how much of the excess ammonium is due to sewage inputs. The study concludes that marine outfalls are the main driver of ammonium pollution in the studied area. Near-natural conditions could be obtained by implementing tertiary treatment to reduce nitrogen in WWTP discharges. Further research should focus on the consequences of ammonium pollution for ecosystems to efficiently evaluate the ecological status of coastal waters under the Water Framework Directive and to prioritize those coastal areas in greater need of nitrogen-removing tertiary treatment.

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\* Masterminded EasyChair and created the first stable version of this document

## 1 Introduction

The Mediterranean Sea (MS) is a semi-enclosed basin, almost entirely surrounded by land. As a consequence, the water exchange with the open sea is limited and the long residence time implies an easier buildup of pollutants (Azzurro et al., 2010). The open waters of the MS are generally oligotrophic, with high nutrient loadings in coastal waters mainly due to a large population living in the coast (Powley et al., 2017). Although phytoplankton growth in the Eastern MS is considered to be phosphorus limited, the Western MS may present shifts between nitrogen and phosphorus limitation (Marty et al., 2002).

Many sources of nutrients have increased eutrophication of water bodies—agriculture, wastewater treatment plants, urban runoff, and fossil fuels—which has become one of the most significant problems in many coastal zones (Vollenweider et al., 1996). A growing evidence shows the impact that anthropogenic nutrient inputs is causing to natural composition of dissolved organic matter and phytoplankton (Aparicio et al., 2016; Pachés et al., 2012). Besides, ammonium concentrations alter nitrification rates in coastal waters, which leads to changes in the nitrogen cycle (Damashek et al., 2016). When nutrient addition changes natural concentrations to many times their natural levels, the impact becomes more serious, including loss of vegetation cover, depletion of oxygen and toxic algal blooms (Bricker et al., 2008).

Water policies require ongoing evaluation and revision to efficiently protect coastal ecosystems. The European Urban Waste Water Treatment Directive establishes a limit for nitrogen discharges to those areas considered as sensitive. The Water Framework Directive (WFD) requires member states to evaluate several quality elements to determine ecological status of coastal water bodies. Nutrient concentrations (a physicochemical quality element) need to be reduced in those coastal areas classified as not compliant. Tertiary treatment is not implemented in most wastewater treatment plants (WWTP) along the MS (Stamou and Kamizoulis, 2008), and secondary treatment still allows a substantial amount of nitrogen in discharged waters (Powley et al., 2016).

Several authors have used broad scale modelling to determine nutrient dynamics in the MS (Lazzari et al., 2016; Powley et al., 2017), or how the degree of wastewater discharges affect water quality (Stamou and Kamizoulis, 2008). There is a growing evidence of the environmental damage caused by direct sewage disposal into the coastal waters of the MS to many ecological parameters, such as benthic communities (De-La-Ossa-Carretero et al., 2012) or fish species (Azzurro et al., 2010). Nonetheless, none of the models developed to date has quantified the contribution of marine outfalls to ammonium concentrations along the surrounding coastal waters. The understanding of the consequences of anthropogenic nutrient discharges into the sea is essential for an effective coastal management.

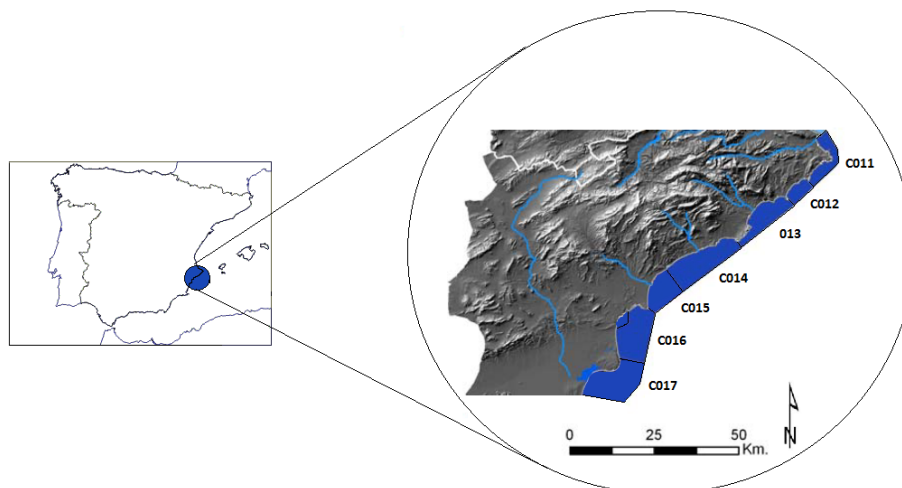
We propose the use of a simplified 1D control volume approach model to estimate to which extent ammonium disposal through marine outfalls increases natural concentrations of coastal waters. After validation with field data, we simulated a new scenario with no WWTP discharges. It is of great interest for management purposes to know how WWTPs discharges affect ammonium concentrations of coastal waters to estimate environmental damage and propose recovery measures.

## 2 Materials and Methods

### 2.1 Study Area

The Jucar River Basin District, located in the Mediterranean coast of Spain, includes 16 coastal water bodies. Among them 7 belong to the typology defined as type III-W: continental coast not influenced by freshwater input. This study focuses on these water bodies, named under the WFD as

C011 to C017 (Figure 1). As no relevant river input exists in this area, natural nitrogen concentrations are low, and the main differences among water bodies is due to anthropogenic activity.



**Figure 1:** Natural coastal waters of the Jucar River Basin District not influenced by freshwater input. Average depth: 4 m; Average length (along the coast): 25km; Width (towards the sea): 1.5km.

The average depth of the study area was approximated to 4 meters. The coastal zone covers a distance of 1500m from the coastline, which was defined as nearshore coastal water with relevant land influence on nutrients (Flo et al., 2011). Several monitoring stations were distributed all along the coast, with 3 to 5 stations in each water body, selected as to represent mean concentrations. The Commission Decision 2013/480/EU established a period of at least five years to determine the ecological status of coastal waters under the WFD. Therefore, we considered that a 5-year mean concentration was appropriated to avoid inter-annual variability. The data used was obtained from monthly campaigns who took place from January 2006 to December 2010, with samples taken from beyond the wave breakpoint (Pachés et al., 2012). Wind speed was also measured in each campaign. Monthly analysis in this area was interrupted and consequently more recent data was not available. Nonetheless, there were no relevant changes in water management since 2010 and water quality changes are not likely to have occurred subsequently.

Several WWTPs located along the coastline discharge treated waters into the sea through marine outfalls. All these plants have both primary and secondary treatment facilities. However, most of them do not include a nitrogen removal process (Table 1). Total nitrogen concentrations and volumes discharged per year was obtained from the local government (discharges for 2014). It was considered that nitrogen was mainly in the form of ammonium when it reaches marine waters. To validate this assumption and the information received, the data was compared with mean ammonium concentrations obtained from a total of 8 campaigns carried out between 2006 and 2008 at the discharge points. Even though the data do not correspond to the same years, there was no relevant changes in sewage treatment or disposal during these years. Despite small differences, the values measured during the campaigns are in line with the data obtained from the local government (no difference between means at a 95% significance level in paired t-test). We used the data obtained from the government for being more updated and because we considered that it better represents yearly average discharges.

WWTP	Location	V <sup>1</sup> (m <sup>3</sup> .yr <sup>-1</sup> )	TN <sup>1</sup> (mgN.L <sup>-1</sup> )	NH <sub>4</sub> <sup>2</sup> (mgN.L <sup>-1</sup> )	N removal
Playa Arenal	C011	1,510,369	10.5	6.5	Yes
Moreira-Teulada	C012	382,680	50.7	50.1	No
Calpe	C013	1,991,351	8.5	9.8	Yes
Benidorm	C013	9,256,785	18.8	24	No
Monte Orgegia	C016	4,585,562	29.0	25.1	No
Rincón de León	C016	10,875,207	40.6	37	No
Tabarca	C017	9,150	74.4	NA	No

**Table 1:** Sewage discharges to studied water bodies. V: volume of sewage discharged; TN: Total Nitrogen in sewage; NH<sub>4</sub>: Ammonium in sewage; N removal: existence of nitrogen removing tertiary treatment.

<sup>1</sup> Data obtained from local government (2014)

<sup>2</sup> Data from campaigns (2006-2008)

## 2.2 Mathematical Model

We used a one-dimensional formulation of the transport equation for ammonium along the coastline. y-axis was established in the direction of the coast. As tides and waves mix coastal waters constantly, we considered each water body as completely mixed. Advection is neglected over turbulent diffusion, which was considered the main driver of mixing in coastal waters. Diffusion occurs between adjacent water bodies (y-axis) and waters further into the sea (x-axis). We included a term of decay to simulate the effect of all physical and biochemical processes. A final term includes all ammonium sources or sinks. The final equation is:

$$\frac{\partial C}{\partial t} = D_y \frac{\partial^2 C}{\partial y^2} + D_x \frac{(C_{os}-C(y))}{A_x(y)} - kC(y) + \phi(y) \quad (1)$$

Where: C=ammonium concentration; D<sub>y</sub>=diffusion in y-axis; D<sub>x</sub>=diffusion in x-axis; C<sub>os</sub>=ammonium concentration in open sea; A<sub>x</sub>=cross-area in x-axis; k=decay rate.

The MS is known to have a microtidal range and therefore dispersion is dominated by the effect of wind on transport (Hernández-Carrasco et al., 2013). Considering that wind is the main driver of diffusion (wind-induced macro-eddies), we calculated wind-induced turbulent diffusion as (Leenen, 1982):

$$D = u \sqrt{\frac{\rho_a C_D}{\rho_w}} \cdot L \quad (2)$$

Where: D=diffusion coefficient; u=wind speed; ρ<sub>a</sub>=air density; ρ<sub>w</sub>=water density; C<sub>D</sub>=drag coefficient; L=characteristic length.

Coastal waters were divided into 7 finite segments (corresponding to each water body presented in Figure 1) and a mass balance for ammonium was developed for each of them. The mass balance equations were solved by means of a control volume approach. The term of sources and sinks includes a term of diffuse load S<sub>D</sub> (atmospheric load and benthic flux) and a term of direct loads Q<sub>in</sub> C<sub>in</sub>. WWTP discharges are added as direct loads. The following transport equation was established for ammonium in each segment i, and set equal to zero by assuming steady state conditions:

$$0 = \frac{D_y}{A_{y,i}} (C_{i-1} - C_i) + \frac{D_y}{A_{y,i}} (C_{i+1} - C_i) + \frac{D_x}{A_{x,i}} (C_{os} - C_i) - KC_i + \frac{S_D}{V_i} + \sum Q_{in,i} C_{in,i} \quad (3)$$

Where  $V_i$  =volume of segment  $i$ ;  $C_{i-1}$ ,  $C_i$ ,  $C_{i+1}$  and  $C_{os}$ =respectively ammonium concentration in segment  $i-1$ ,  $i$ ,  $i+1$  and the open sea;  $S_D$ =diffuse ammonium load;  $A_{xi}$ ,  $A_{yi}$  and  $A_{zi}$ =respectively cross-section of segment  $i$  in  $x$ -axis,  $y$ -axis and  $z$ -axis;  $Q_{in,i}$ =inflow to segment  $i$ ;  $C_{in,i}$ =ammonium concentration of inflow to segment  $i$ .

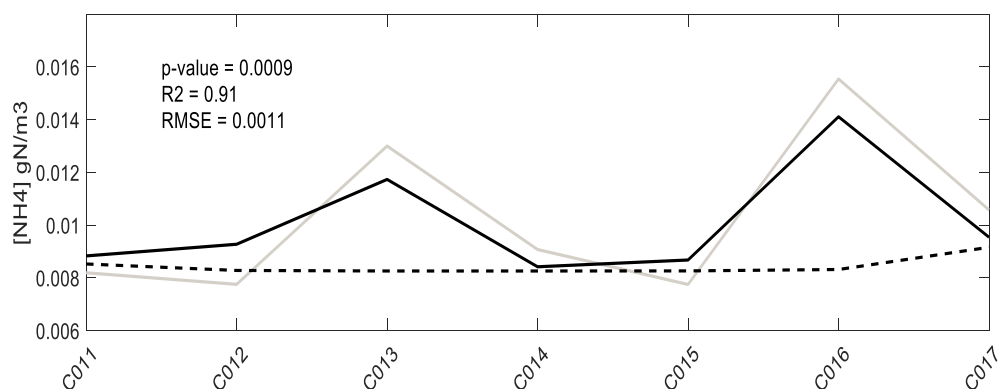
We solved the equations for each segment by establishing a matrix equation and using LU decomposition in MatLAB. We calibrated the model with literature parameters and measured boundary conditions (Table 2) and validated it with field data.

Parameter	Type	Value	Unit	Reference
$u$	constant	10.2	$m.s^{-1}$	measured
$S_D$	constant	12.91	$gN.d^{-1}.m^{-2}$	(Sospedra et al., 2015) (“The European Monitoring and Evaluation Programme (EMEP),” 2013)
$C_D$	constant	0.001	-	(Leenen, 1982)
$K$	constant	0.05	$d^{-1}$	(Bianchi et al., 1999)
$\rho_w$	constant	1000	$kg.m^{-3}$	(Leenen, 1982)
$\rho_a$	constant	1.3	$kg.m^{-3}$	(Leenen, 1982)
$C_0$	boundary condition	0.0124	$gN.m^{-3}$	measured
$C_8$	boundary condition	0.0224	$gN.m^{-3}$	measured
$C_{os}$	boundary condition	0.00406	$gN.m^{-3}$	(“Spanish Ministry of Agriculture and Fishery, Food and the Environment,” 2012)

**Table 2:** Input values to the model.  $u$ : mean wind speed;  $S_D$ : diffuse load;  $C_D$ : drag coefficient;  $K$ : decay rate;  $\rho_w$ : water density;  $\rho_a$ : air density;  $C_0$ : ammonium concentration in the limiting segment 0;  $C_8$ : ammonium concentration in the limiting segment 8;  $C_{os}$ : ammonium concentration in the open sea.

### 3 Results and Discussion

Estimated and measured ammonium concentrations are presented in Figure 2, as well as the new scenario with no WWTP inputs. Estimated ammonium concentrations are slightly lower than those measured. This may be due to diffuse nutrient loads from sources such as agriculture, aquaculture or harbours, which were not added to the model because of the difficulty in quantifying such pollution. River inputs were neglected in this study as the study area has a very low continental influence, with irregular river flows. Nonetheless, some WWTPs discharge their waters to rivers close to the mouth. This may be the case for Amadorio river, located in C014, or with Algar river in C013. C013 also has several marine farms which may represent an additional source of diffuse ammonium. Similarly, the port of Alicante, classified as a highly modified coastal water under the WFD, lies within C016.



**Figure 2:** Measured (grey continuous line) and estimated (black continuous line) ammonium concentrations. Scenario with no WWTP discharges (black discontinuous line). The coefficient of determination ( $R^2$ ) and relative mean squared error (RMSE) between measured and estimated values are shown.

Even so, the proposed model is in close agreement with the measured data, as indicated by the high correlation obtained between measured and estimated ammonium concentrations ( $R^2=0.91$ ). The model developed accurately simulates how direct sewage ammonium inputs affect concentrations in coastal waters. The modelled scenario with no WWTP discharges indicates that ammonium concentrations are highly influenced by marine outfall discharges, pointing out sewage inputs as the main driver of ammonium pollution in the studied area. Under the WFD, C012 was determined to be the reference water body with minor anthropogenic pressures. The most polluted site is C016 with ammonium concentrations doubling the reference C012. The model developed determined that most of its pollution comes from WWTPs. As indicated by the simulation with no sewage discharges, the near natural conditions prevailing in C012 could be re-established in all water bodies by reducing marine outfall inputs. Nonetheless, the regular monitoring of coastal waters of the studied area needs to be re-established as soon as possible to apply the model developed with updated concentrations.

These results agree with previous studies, which stated that tertiary treatment to reduce nutrient concentrations in sewage discharged would be a solution to nutrient MS pollution (Powley et al., 2016; Stamou and Kamizoulis, 2008). Nutrient anthropogenic inputs through rivers discharging into the MS were determined to be an important source of contamination (Stamou and Kamizoulis, 2008). Our study added to this work, determining that direct discharges through marine outfalls also constitute an important source of nitrogen to coastal waters. As determined previously (Powley et al., 2016) and reaffirm by our study, coastal areas where riverine inputs are minimal, direct wastewater discharges are a possible driver of eutrophication.

Clearly, an upgrade to nutrient-removing tertiary treatment is needed if natural nitrogen conditions of the MS are to be regained. However, the costs associated with the complete conversion to tertiary treatment may represent a financial challenge (Powley et al., 2016). The WFD undoubtedly implied a stepping stone to environmental sustainability of European waters. But regional authorities are concerned about water bodies not compliant with the WFD, as the palliative measures they may be obliged to take would be costly. Consequently, they are tempted to adopt methods which are too optimistic. Currently, the value being used as boundary for compliance in the study area is  $0.064 \text{ mgN.L}^{-1}$  of ammonium, a value set without scientific evidence which represents more than 8 times the amount of ammonium found in the reference site C012. Unfortunately, underestimating water pollution would carry future problems with increasing difficulty in the recovery of ecosystems. Further study of the ecological consequences of ammonium contamination should be carried out to set scientific-based reference conditions within the WFD and determine which areas have a more urgent upgrade to tertiary treatment.

## 4 Conclusion

We employed a simplified 1D model to estimate the effect of WWTP ammonium direct discharges along a coastal region of the Mediterranean Sea where river influence is not significant. Despite the assumptions used, modelled concentrations have a high correlation with field data ( $R^2=0.91$ ). We determined that marine outfalls are the source of almost all ammonium pollution in the study area. Clearly nutrient discharges from WWTPs is still an important source of ammonium in certain coastal areas of the Western Mediterranean Sea. The model developed is a good preliminary evidence of the alteration of natural conditions that direct marine sewage discharges can cause on coastal nutrient concentrations if adequate treatment is not implemented. Further research with detailed computation is required to accurately determine the consequences for coastal systems' degradation. Additionally, future research should focus on the effect that ammonium increasing concentrations have on ecosystems to correctly evaluate the ecological status of coastal waters under the WFD and to determine which areas need urgent upgrade to nitrogen-removing tertiary treatment.

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