





## Article

# Sediment Quality Indices for the Assessment of Heavy Metal Risk in Nador Lagoon Sediments (Morocco) Using Multistatistical Approaches

Otman El Ouaty <sup>1,\*</sup> , Abdelmounim El M'rini <sup>1</sup> , Driss Nachite <sup>1</sup> , Elena Marrocchino <sup>2</sup> and Iliaria Rodella <sup>3</sup> 

<sup>1</sup> Research Laboratory in Applied Marine Geosciences, Geotechnics and Geohazards (LR3G), Faculty of Sciences, Abdelmalek Essaâdi University, Tetouan 93000, Morocco; aelmrini@gmail.com (A.E.M.); nachited@yahoo.fr (D.N.)

<sup>2</sup> Department of Environmental and Prevention Science, University of Ferrara, Corso Ercole I d'Este 32, 44121 Ferrara, Italy; elena.marrocchino@unife.it

<sup>3</sup> International Research Office, University of Padova, Via Martiri Della Libertà 8, 35137 Padova, Italy; ilaria.rodella@unipd.it

\* Correspondence: elouatyotman@gmail.com; Tel.: +212-626490940

**Abstract:** Heavy metals in coastal ecosystems caused by the increased expansion of urbanization, industrialization, and agricultural practices have become a significant environmental risk to human well-being. This study evaluates and compares 17 sediment quality indices to examine the possible ecological and human health risks associated with heavy metal concentrations in the sediments of the Nador lagoon in Morocco. The concentration order of the HMs and sulfurs evaluated was S > Sr > Ba > V > Zr > Zn > Cr > Rb > La > Cu > Pb > Ni > Ce > Nd > Co > Sc > Nb > Ga > Th > Y > Hf. Sulfurs, Pb, Sr, and Nd exhibited concentrations that exceeded geochemical background values. The analysis of the sediment quality indices allowed us to understand that the Nador lagoon was moderately to strongly polluted by heavy metals originating from various anthropogenic activities. Results from the Sediment Quality Guidelines indicated a toxic response in the benthic organisms within the lagoon, while the ecological hazard analysis revealed a very high risk of heavy metal contamination in the ecosystem. The Hazard Index for non-carcinogenic values was below the limit, suggesting a lack of non-cancerous effects. However, Cu and Pb concentrations surpassed the Lifetime Cancer Risk range, indicating a potential cancer risk with prolonged exposure. Integrating our research into coastal management frameworks can contribute to the preservation and enhancement of these coastal ecosystems for future generations.

**Keywords:** heavy metals; sediment quality indices; numerical sediment quality guidelines; potential ecological risk; potential human health risk; Nador lagoon



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## 1. Introduction

The Nador lagoon (Northeast, Morocco) is assessed as a pollution hotspot in the Mediterranean basin [1]. This area has a valuable ecological level that encompasses protected habitats for several species, such as the marbled goby, *Pomatoschistus marmoratus* [2]. However, this prestigious site has been recently affected by anthropogenic issues, especially due to poor environmental management in industrial and agricultural areas [3]. In general, heavy metals (HMs) are critical environmental pollutants that impact coastal wetlands such as the Nador lagoon, due to their toxicity, persistence in the environment, and bioaccumulative nature. These pollutants derived from natural and anthropogenic sources are considered hazardous and persistent chemicals that can accumulate in soil matrices and sediments, and can contaminate water bodies [4], as well as green seaweed (e.g., *Ulva lactuca* as reported by [5]). Mobilization of these elements in the environment and disruption of their biogeochemical cycles have increased in the recent past in correlation with the increase in industrial activities, especially in developing countries where the population

is more sensitive and affected by pollutants [6]. HM pollution in wetlands and terrestrial environments, such as lagoons, rivers, lakes, and streams, induces their accumulation in living organisms (fish, clams), agricultural lands, crops, etc. Therefore, we can observe both environmental contamination and a public health problem because of HM accumulation in the food chains. Consequently, monitoring and analysis of HM concentrations and the assessment of the ecological state of coastal lagoon environments, such as Nador, are mandatory for evaluation and management [7]. From this perspective, several studies have used these analyses to examine the sedimentological and geochemical characteristics of the sediments in the Nador lagoon and assess the ecological framework of this basin. For example, the sediment and water requalification management plans in 1992 and 2011 were studied in [8].

An initial assessment of lead (Pb) pollution was carried out by [9], while simultaneously [10,11] identified multiple instances of human-induced pollution in the lagoon. These investigations consistently implicated anthropogenic activities, particularly urban effluents, as the primary contributors to the presence of HMs in lagoon sediments. In a more recent study, ref. [12] examined the ecotoxicological state of sediments, taking into account seasonal variations in HM concentrations. Despite some restoration initiatives, ref. [11] noted a progressive increase in concentrations of lead (Pb), zinc (Zn), and copper (Cu) over time. Furthermore, ref. [11] conducted an analysis of the spatial distribution of heavy metals and sulfur in the lagoon, revealing a noteworthy enrichment of strontium (Sr).

The current research focused on evaluating the extent of HM pollution in the sediments of the coastal Nador lagoon in northeastern Morocco. The choice of this study area was motivated by the various industrial and agricultural activities that occur, which have a potential impact on pollution levels. These activities pose a risk of releasing toxic substances, which induce potential hazards to both the environment and the health of nearby populations. Therefore, this study aims to comprehensively assess HM pollution in the sediments of the Nador lagoon and discern the influence of various sources of toxic metals. Understanding the dynamics of heavy metal pollution in the Nador lagoon is imperative for effective coastal management strategies. This study aims to investigate the distribution of HM contamination and pollution levels and assess their implications for coastal environmental management.

This study on heavy metal contamination can play a crucial role in assessing the health of coastal ecosystems and facilitating the development of monitoring programs aligned with the Mediterranean Action Plan—Barcelona Convention System. It helps identify pollution sources, pathways, and hotspots, enabling policymakers to implement targeted measures for pollution prevention and control in accordance with the Convention's protocols. Additionally, such studies contribute to understanding environmental risks and vulnerabilities in coastal zones, informing decision-making processes related to land-use planning, industrial activities, and waste management within the framework of Integrated Coastal Zone Management (ICZM).

The main objective of this work is (i) to identify HM concentrations in sediments; (ii) to estimate contamination levels in sediments using sediment quality indices and compare them with different classifications of contamination degrees; (iii) to measure the ecological and health risks of HMs; and (iv) to identify polluted areas in the Nador basin, comparing the distribution of HMs with previous studies, discussing the ecological state of the lagoon. Overall, the analysis of sediment quality indices constitutes basic information for monitoring and protecting wetlands and coastal areas. This management approach allows us to interpret the accumulation and origin of pollutants and to understand the distribution of pollutants associated with their potential risks. This Nador overview can be useful in recovering the lagoon substrate, providing helpful information to structure a sediment and water management strategy for policymakers and managers in this region.

## 2. Overview of the Lagoon

The Nador lagoon, called the Sebkhha of Bou Areg or 'Marchica', constitutes one of the most important lagoons in the Mediterranean due to its size, and is the second largest lagoon in North Africa. The Nador lagoon, located in northwest Morocco, represents a meeting zone between the Rifain and Atlas geological systems, characterized by small mountains that dominate large almost flat depressions, open in the Mediterranean (Figure 1). The Nador lagoon is the most extensive water body of Morocco (115 km<sup>2</sup>), its shape is semi-elliptical elongated semi-elliptical, and its depth varies between 0.50 and 4 m around the perimeter and reaches 7 m in the center. The lagoon is separated from the Mediterranean Sea by a 25 km long sand spit that also allows water exchanges from and to the sea due to artificial channels (see old and new passes in Figure 1) [13]. The tide is semi-diurnal [14], varying between 0.5 m (open water) and 0.1 m (dead water). The lagoon catchment area covers several small independent and juxtaposed hydrographical networks that open at different points of the lagoon; however, a large number of thalwegs are lost in the plains and their waters never reach the lagoon.

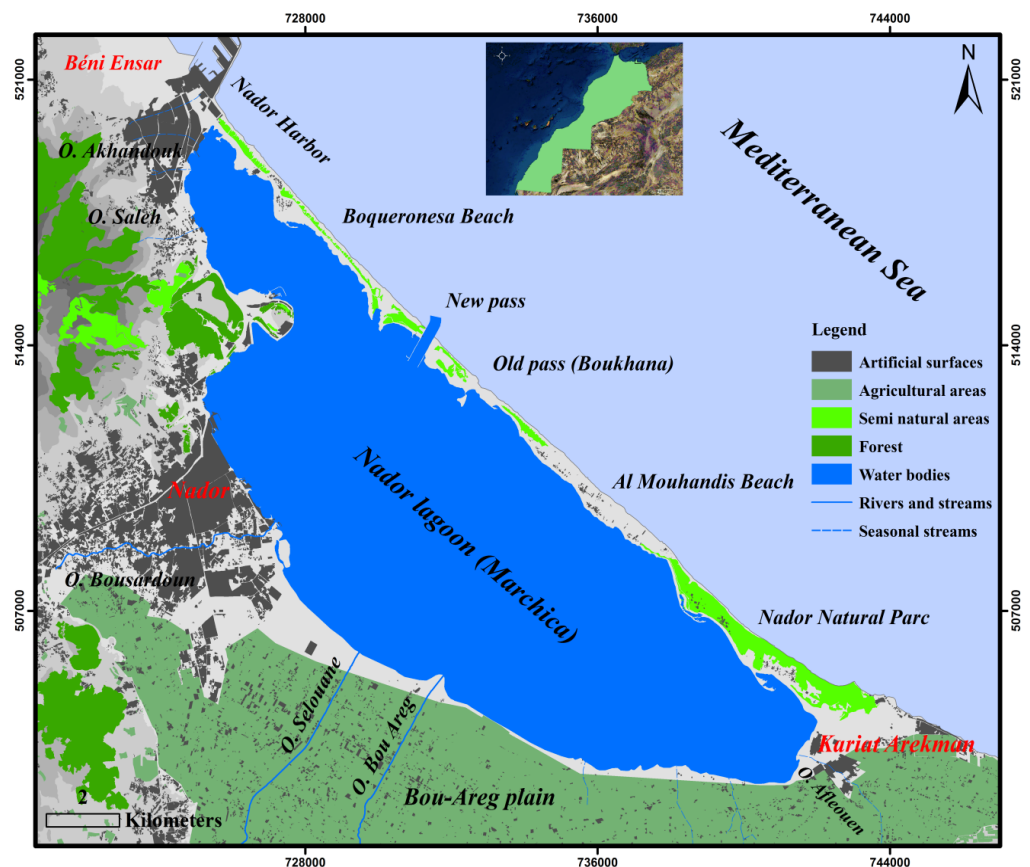


Figure 1. The geographic framework of the Nador lagoon.

This aquatic fauna (invertebrates and fish) represents approximately 7% of the Moroccan marine fauna, about the inventory carried out by [15], and 91 species of water birds. In addition to this aspect related to comprehensive biodiversity, this lagoon constitutes a complementary background to the marine environment with respect to the maintenance of the populations of migratory coastal fish, knowing that it offers them a biotope for the growth and fattening of fry [14]. Therefore, this lagoon has huge ecological value (Ramsar site), but it has suffered and is still suffering from anthropogenic stress linked to population growth, urban development, and industrial and agricultural discharges, in addition to threats due to economic activities carried out at the lagoon site (e.g., recreational activities, tourism).

In general, one of the main environmental issues related to the Nador lagoon is water and sediment pollution. Indeed, recent studies have highlighted that HMs such as, e.g., lead, zinc, copper, chromium, and cadmium, are concentrated in sediments [11,14,16], especially clay minerals such as kaolinite, smectite, and chlorite [16]. Furthermore, the concentrations of certain HMs such as Cu, Pb, and Zn identified in the Nador lagoon exceed those observed in other Mediterranean lagoons (Malaga Bay, Lake Burullus, Bizert Lagoon, and Manzala Lagoon), underscoring the imperative for a comprehensive investigation and restoration of the environmental quality at this site.

### 3. Materials and Methods

The sampling methodology, as described in [8], was tailored to fit the ecological zones delineated within the lagoon. These ecological zones were classified into four main areas: (I) and (II) representing confined regions, (III) indicating a continental-influenced area, and (IV) indicating a marine-influenced area (Figure 2). Four sediment sampling campaigns were conducted from March to June 2011, employing scuba diving and a Van Veen grab (10 cm × 20 cm) in the four zones. A comprehensive set of 50 sediment samples was collected, as described in [11].

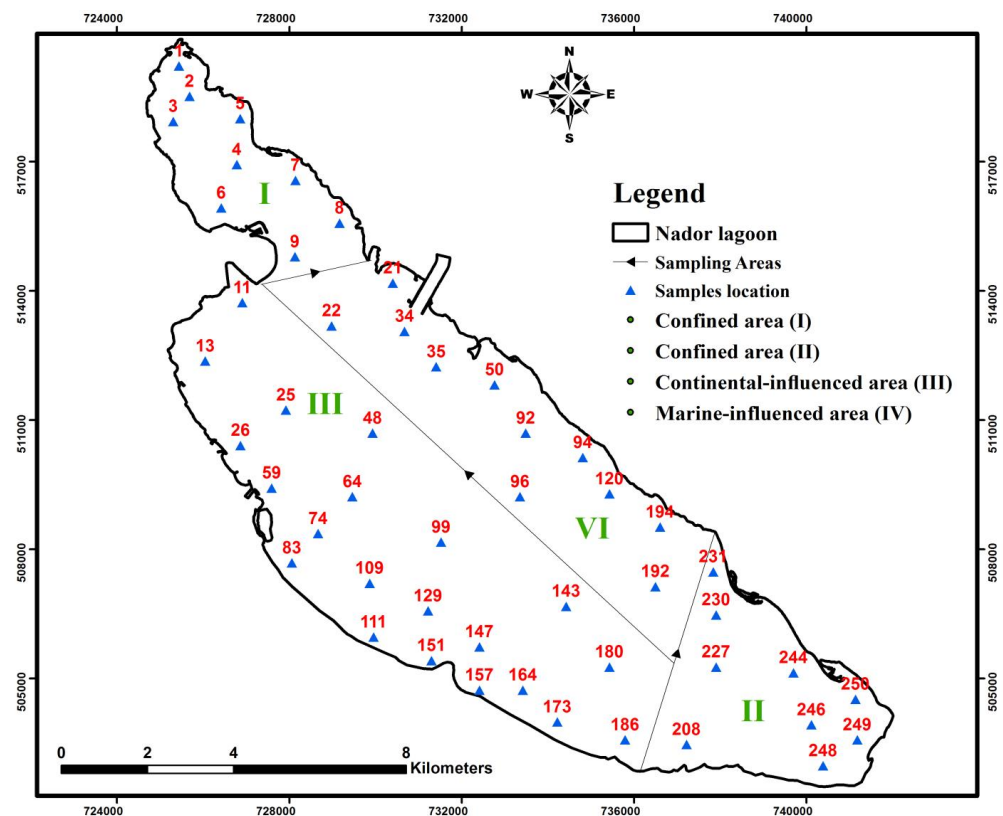


Figure 2. Sampling area network [10].

Heavy metal analysis was performed using (XRF) in powder pellets with an automated ARL Advant-XP automated X-ray Fluorescence (XRF) spectrometer (Waltham, MA, USA), achieving precision and precision exceeding 10% for heavy metals above 10 mg kg<sup>-1</sup>. An aqua regia (AR) extraction test, following official Italian soil analysis methods, involved specific steps such as wetting the powdered sample, acidification, addition of hydrogen peroxide, evaporation, and further treatment with aqua regia. The subsequent analysis, conducted with a Thermo-Scientific spectrometer at the University of Ferrara (Italy), incorporated internal standards (Rh, In, and Re) to mitigate instrument drift, achieving accuracy and precision exceeding 10% for all elements. The E.P.A. The reference standards SS-1 and SS-2 were examined as reference standards for cross-verification. Further extraction tests,

using less aggressive reagents such as 0.05 M EDTA, 0.005 M DTPA, 1 M  $\text{NH}_4\text{NO}_3$ , and a 'rhizosphere solution' were carried out in triplicate. For example, the extraction of EDTA involved dissolving 2 g of soil in 20 mL of 0.05 M EDTA, adjusted to pH 7.0, and shaking for 1 h. The rhizosphere-based extraction protocol included mixing 2 g of soil with 20 mL of acetic, lactic, citric, and malic acids in a molar concentration, resulting in a total molar concentration of 10 mM. After shaking for 16 h, the soil suspension was centrifuged and an aliquot of supernatant (10 mL) was withdrawn and acidified with suprapure  $\text{HNO}_3$ . All extraction procedures, including blanks, were executed in triplicate. For the extraction of  $\text{NH}_4\text{NO}_3$ , 10 g of soil was combined with 50 mL of 1M  $\text{NH}_4\text{NO}_3$  and stirred for 2 h at room temperature. The extraction involved shaking 10 g of soil with 100 mL of deionized water for 16 h, followed by centrifugation at  $300 \times g$  for 15 min, and stabilization with suprapure  $\text{HNO}_3$  [17].

The elements analyzed encompassed Cr, La, Nb, Pb, Rb, S, Sc, Sr, Th, Nd, Ba, Ni, Ce, Zr, Co, Cu, V, Ga, Hf, Y, and Zn (A1).

#### 4. Sediment Contamination Indices

The following contamination indices were calculated taking into account the list of analyses reported in Appendix A Table A1, based on the 50 samples collected.

##### 4.1. Contamination Factor (Cf)

The extent of heavy metal contamination can be quantified using the contamination factor (Cf) [18]. This factor represents the ratio of the heavy metal content in the sediment to the geochemical background value, which is the average concentration in the shale standards for the respective heavy metal [19]. The background concentration standards for Zn, Cu, Mn, Fe, and Pb are 95 mg/kg, 45 mg/kg, 850 mg/kg, 47.200 mg/kg, and 20 mg/kg, respectively [19]. This ratio serves as a means to assess the relative impact of the contaminant on the environment and pinpoint potential sources of pollution. The degree of contamination is determined by the sum of all the contamination factors, making it a valuable tool for monitoring the trends of contamination over time [20]. The calculation is expressed as follows (1):

$$Cf = \frac{C (\text{Heavy metal})}{C (\text{Geochemical background})} \quad (1)$$

where C (Heavy metal) represents the concentration of each heavy metal within the lagoon ecosystem and C (geochemical background) represents the concentration of each heavy metal background according to [19]. According to [18], the factor can be expressed after calculation as follows:

$Cf < 1$	Low degree of contamination
$1 < Cf < 3$	Moderate degree of contamination
$3 < Cf < 6$	Considerable degree of contamination
$Cf > 6$	Very high degree of contamination

##### 4.2. Pollution Load Index (PLI)

The Pollution Load Index (PLI) serves as a quantitative measure to assess overall pollution in a designated environment. It indicates the extent to which the concentration of HMs in the sediment exceeds the geochemical background concentration of the respective element. The PLI offers a comprehensive indication of the collective toxicity of HMs in a specific sample [20,21]. This index is frequently used to compare contamination levels across various environments and identify areas that require pollution control measures. It is mathematically expressed as the n root of the product of the contamination factors (Cf) (2):

$$PLI = \sqrt[n]{Cf_1 \times Cf_2 \times Cf_3 \times \dots \times Cf_n} \quad (2)$$

where CF n: the contamination factor value of the heavy metal n. Pollution load index values are interpreted into two levels: polluted (PLI > 1) and unpolluted (PLI < 1). Therefore, a PLI value of 0 indicates excellent, a value of 1 indicates the presence of only baseline-level pollution, and a value above 1 indicates progressive deterioration of the site [22].

#### 4.3. Modified Degree of Contamination (mCd)

The modified degree of contamination (mCd) was engendered to estimate by and large the degree of contamination at a given site according to the formula of [23] (3):

$$mCd = \frac{(\sum_{i=1}^{i=n} Cf)}{n} \quad (3)$$

In this equation, 'n' represents the number of elements analyzed, and 'i' denotes the specific heavy metal. Adjustments made to the contamination degree formula (Cd\_deg), which involves summing all contamination factors (Cf) for a specific sediment location, which involves dividing the sum by the number of pollutants analyzed, enables the inclusion of an unlimited number of HMs in the study [20]. This modification provides flexibility in analyzing a diverse range of HMs, as classified below.

mCd < 1.5	Very low degree of contamination
1.5 ≤ mCd < 2	Low degree of contamination
2 ≤ mCd < 4	Moderate degree of contamination
4 ≤ mCd < 8	High degree of contamination
8 ≤ mCd < 16	Very high degree of contamination
16 ≤ mCd < 32	Extremely high degree of contamination

#### 4.4. Potential Contamination Index (Cp)

According to [24], the potential contamination index (Cp) can be calculated by the following procedure (4):

$$Cp = \frac{(HMs)_{\text{Sample Max}}}{(HMs)_{\text{Geochemical Background}}} \quad (4)$$

In this context, "HM sample Max" denotes the highest concentration of HM in the sediment, while "HM geochemical background" signifies the average value of the same heavy metal at a background level [19]. Interpreting the Cp values aligns with the recommendations of [22] and is expressed as follows:

Cp < 1	Low degree of contamination
1 < Cp < 3	Moderate degree of contamination
Cp > 3	Severe degree of contamination.

#### 4.5. Geo-Accumulation Index (Igeo)

Initially introduced by [25], the Geo-accumulation index (Igeo) serves the purpose of evaluating heavy metal contamination in sediments by comparing current concentrations of HMs with their preindustrial levels. Consequently, the Igeo index is applied to gauge the existence and magnitude of anthropogenic contaminant deposition in surface sediment, and its calculation is expressed as follows (5):

$$Igeo = \log_2 \left[ \frac{Ci}{(1.5 \times C_{GB})} \right] \quad (5)$$

In this equation, Ci represents the measured concentration of the specific heavy metal 'i' in the sediment, and C<sub>GB</sub> denotes the geochemical background concentration or the reference value for the same heavy metal. Incorporation of a factor of 1.5 is justified due

to potential fluctuations in the geochemical background values for a given heavy metal within the ecosystem and the presence of minimal anthropogenic influences. According to [25,26], the Geo-accumulation index (Igeo) is classified into seven classes:

Class 0	$I_{geo} \leq 0$	Unpolluted
Class 1	$0 < I_{geo} \leq 1$	Unpolluted to moderately polluted
Class 2	$1 < I_{geo} \leq 2$	Moderately polluted
Class 3	$2 < I_{geo} \leq 3$	Moderately to strongly polluted
Class 4	$3 < I_{geo} \leq 4$	Strongly polluted
Class 5	$4 < I_{geo} \leq 5$	Strongly to extremely polluted
Class 6	$I_{geo} > 5$	Extremely polluted

4.6. Pollution Index (Pi)

For the evaluation of the levels of heavy metal pollution, the pollution index was calculated following [27]. The pollution index (Pi) is determined by dividing the concentration of each heavy metal in the sample area by its respective background value, as expressed in Equation (6):

$$P_i = \frac{C_i}{C_{GB}} \tag{6}$$

$C_i$  represents the HM concentration, and  $C_{GB}$  denotes the geochemical background value corresponding to each HM. The pollution index for each heavy metal in the lagoon was classified as follows: non-pollution ( $P_i < 1$ ), indicating that the concentration of the heavy metal was below the threshold level. It is important to note that a  $P_i$  value below 1 does not necessarily imply the absence of pollution; there may still be influences from anthropogenic sources or other enrichments in the background [25]. Furthermore, low-level pollution is indicated when  $1 < P_i \leq 2$ , moderate pollution for  $2 < P_i \leq 3$ , and high pollution for  $P_i > 3$ . This classification was based on the criteria outlined by [27]:

$(P_i < 1)$	Non-pollution
$(1 < P_i \leq 2)$	Low-level pollution
$(2 < P_i \leq 3)$	moderate level of pollution
$(P_i > 3)$	High level of pollution

4.7. Nemerow Integrated Pollution Index (NIPI)

By computing the individual  $P_i$  indices, we derive the Nemerow integrated pollution index (NIPI), a metric employed for evaluating the extent of pollution in an industrial area [28,29]. In this investigation, the NIPI was utilized to evaluate the overall quality of the results of the sediments based on the  $P_i$  index. The calculation of NIPI is expressed as follows (7):

$$NIPI = \sqrt{P_{i_{max}}^2 + P_{i_{average}}^2 / 2} \tag{7}$$

Here,  $P_i$  Max represents the maximum  $P_i$  value for an individual heavy metal, and  $P_i$  Ave signifies the mean value of  $P_i$  for the same heavy metal. NIPI classification was determined based on the criteria described in [30]:

$NIPI \leq 0.7$	Non-pollution
$0.7 < NIPI \leq 1$	Warning line of pollution
$1 < NIPI \leq 2$	Low level of pollution
$2 < NIPI \leq 3$	Moderate level of pollution
$NIPI > 3$	High level of pollution

4.8. Nemerow Pollution Index and Modified Pollution Index (PI Nemerow—MPI)

The modified pollution index (MPI), introduced by [31], was designed to evaluate sediment and soil for multiple elements. This modification addresses the constraints

associated with single-element pollution indices by incorporating the enrichment index in its computation. The Nemerow Pollution Index, developed by Nemerow [30–32], considers the results of the contamination factor calculation. Both factors can be calculated using the following equations (Equations (8) and (9)):

$$PI = \sqrt{\frac{(Cf_{(average)})^2 + (Cf_{(Max)})^2}{2}} \quad (8)$$

$$MPI = \sqrt{\frac{(Ef_{(average)})^2 + (Ef_{(Max)})^2}{2}} \quad (9)$$

In these equations, Cf average, Ef average, Cfmax, and Ef max denote average contamination factors, average enrichment factors, maximum contamination factors, and maximum enrichment factors, respectively. The Pollution Index (PI) is then classified into five categories:

PI < 0.7	Unpolluted
0.7 < PI < 1	Slightly polluted
1 < PI < 2	Moderately polluted
2 < MPI < 3	Severely polluted
MPI > 3	Heavily polluted

Six thresholds are employed to categorize sediment quality based on the modified pollution index (MPI).

MPI < 1	Unpolluted
1 < MPI < 2	Slightly polluted
2 < MPI < 3	Moderately polluted
3 < MPI < 5	Moderately–heavily polluted
5 < MPI < 10	Severely polluted
MPI > 10	Heavily polluted

#### 4.9. Nemerow Multifactor (Pc)

As described in [33], the Nemerow multi-factor method was used to evaluate the contamination index, focusing on the impact of elevated concentrations of HMs on the quality of environmental sediments [34]. The calculation is expressed through the following Equation (10):

$$Pc = \left\{ \frac{\left[ \left( \frac{Ci}{Si} \right)_{average}^2 + \left( \frac{Ci}{Si} \right)_{max}^2 \right]}{2} \right\}^{1/2} \quad (10)$$

In this equation, Pc represents the comprehensive contamination index for sediment contaminants,  $\left( \frac{Ci}{Si} \right)_{average}$  is the average value of the pollution index for sediment contaminants, and  $\left( \frac{Ci}{Si} \right)_{max}$  is the maximum value of the single contamination index. According to [33], the standard for the sediment contamination scale is derived from the trial implementation outline for the Assessment of Environmental Quality in green food growing areas, compiled in 1994. The classification criteria for the comprehensive sediment assessment Pc are delineated as follows:

PC	Contamination level	Contamination degree
PC ≤ 0.7	Safe	Clean
0.7 < PC < 1	Alert	Still clean
1 < PC ≤ 2	Light contamination	Sediments slightly contaminated.
2 < PC ≤ 3	Moderate contamination	Sediments moderately contaminated
PC > 3	Severe contamination	Sediments seriously contaminated

#### 4.10. Sediment Quality Guidelines (SQGs)

The evaluation, protection, and management of aquatic ecosystems place a significant emphasis on sediment quality concerns [35]. Sediments are recognized as potential reservoirs and sources of both inorganic and organic contaminants, particularly during changes in environmental conditions [36]. Numerical values known as sediment quality guidelines (SQGs) play a crucial role in the evaluation of sediment quality, often referred to as “guidelines”, “criteria”, or various other terms. SQGs for freshwater ecosystems have been developed through various approaches, each with distinct advantages and limitations that influence their application in the sediment quality assessment process. Notably, consensus-based SQGs have been established for 28 chemicals in freshwater sediments. To streamline this variety, two SQGs were derived from the existing guidelines: threshold effect concentration (TEC) and probable effect concentration (PEC) [37]. These SQGs offer a framework for interpreting comprehensive sediment chemistry data by pinpointing concentrations of potentially harmful chemicals capable of causing or significantly contributing to adverse effects on organisms that dwell in sediment [38].

#### 4.11. Mean ERM Quotient (*m-ERM-Q*) and Mean PEL Quotient (*m-PEL-Q*)

Both serve as valuable tools for summarizing extensive chemical data from sediments containing mixtures of contaminants into a single numerical representation [39]. To evaluate the potential biological effects of combined toxicant groups based on the results of the numerical sediment quality guidelines (SQGs), we calculated the mean quotient for a wide range of contaminants. The mean ERM quotient and the mean PEL quotient are associated with the probability of toxicity, and two factors were calculated using the following equations (Equations (11) and (12)) [40]:

$$m - PEL - Quotient = \frac{\sum_{i=1}^n \left( \frac{C_i}{PEL_i} \right)}{n} \quad (11)$$

$$m - ERM - Quotient = \frac{\sum_{i=1}^n \left( \frac{C_i}{ERM_i} \right)}{n} \quad (12)$$

In this context,  $C_i$  represents the concentration of the contaminant (HM) in the sediment, while  $PEL_i$  and  $ERM_i$  denote the respective screening levels based on the sediment quality guidelines (SQGs). The variable ‘ $n$ ’ signifies the number of contaminants under consideration in the study area. The evaluation of the potential ecological impact of HMs has led to the characterization of four relative priority levels of contamination, as delineated in previous studies [37–39]:

M – PEL – Quotient		M – ERM – Quotient	
$m - PEL - Q > 2.3$	High	$m - ERM - Q > 1.5$	High
$2.3 > m - PEL - Q > 1.51$	Medium High	$1.5 > m - ERM - Q > 0.51$	Medium High
$1.5 > m - PEL - Q > 0.11$	Medium low	$0.5 > m - ERM - Q > 0.11$	Medium low
$m - PEL - Q < 0.1$	Low	$m - ERM - Q < 0.1$	Low

#### 4.12. Modified Hazard Quotient (*mHQ*)

The modified hazard quotient introduces a novel index for assessing sediment pollution, focusing on the individual levels of contamination levels of specific HMs. This innovative approach facilitates the evaluation of contamination by comparing the concentration of HMs in sediment with the comprehensive distributions of adverse ecological effects corresponding to slightly varied threshold effect concentrations, probable effect concentrations, and Severe Effect Levels (TEL, PEL, and SEL). The assessment of HMs through the modified hazard quotient (*mHQ*) emerges as a crucial tool, shedding light on

the level of risk posed by each heavy metal to the aquatic environment and the biota [40]. The calculation of mHQ is performed using the following Formula (13):

$$mHQ = \left[ Ci \left( \frac{1}{TELi} + \frac{1}{PELi} + \frac{1}{SELi} \right) \right]^{1/2} \quad (13)$$

In this equation, Ci denotes the measured concentration of trace elements in the sediment samples from the study area, while TELi, PELi, and SELi represent acronyms for the threshold effect level, probable effect level, and severe effect level, respectively, for each specific heavy metal. As proposed by [41], a classification system for contamination by individual HMs has been introduced using the newly developed index, described as follows.

mHQ	Degree of risk
mHQ > 3.5	Extreme severity of contamination
3.0 mHQ < 3.5	Very high severity of contamination
2.5 mHQ < 3.0	High severity of contamination
2.0 mHQ < 2.5	Considerable severity of contamination
1.5 mHQ < 2.0	Moderate severity of contamination
1.0 mHQ < 1.5	Low severity of contamination
0.5 mHQ < 1.0	Very low severity of contamination
mHQ < 0.5	Nil to very low severity of contamination

#### 4.13. Evaluation of Potential Ecological Risk Index (PERI)

The Potential Ecological Risk Index (PERI), introduced by [18], offers an approach for evaluating ecological risk in aquatic environments, presenting a quick and straightforward quantitative value for the potential ecological risk at a given contamination site. The effectiveness of this method has been validated by testing on 15 Swedish lakes, covering a diverse range in terms of size, pollution status, and trophic conditions [42]. PERI comprises three fundamental modules: Contamination factor (Cf), Toxic-Response Factor (Tr), and Potential Ecological Risk Factor (Er). Calculating the potential ecological risk factor for an individual heavy metal (Er) and the comprehensive potential ecological risk index (PERI) is achieved using the following equations (Equation (14)) [18]:

$$PERI = \sum_{i=0}^n Er^i = \sum_{i=0}^n Tr^i \times Cf^i \quad (14)$$

where

PERI = the requested potential ecological risk index for the basin/lake.

Er<sup>i</sup> = the potential ecological risk factor for the given heavy metal (i).

Tr<sup>i</sup> = the toxic-response factor for the given heavy metal.

To quantitatively determine the potential ecological risk of a given contaminant in a given ecosystem, [16] defines the risk factor (Er<sup>i</sup>) accordingly (15):

$$Er^i = Tr^i \times Cf^i \quad (15)$$

The following terminology is used to describe the ecological risk factor (Er) [18]:

Er <sup>i</sup> < 40	Low potential ecological risk
40 < Er <sup>i</sup> < 80	Moderate potential ecological risk
80 < Er <sup>i</sup> < 160	Considerable potential ecological risk
160 < Er <sup>i</sup> < 320	High potential ecological risk
Er <sup>i</sup> > 320	Very high ecological risk

Analogous to the previous calculation regarding the potential ecological risk factor (Er), [16] identifies the requested potential ecological risk index (PERI) as the sum of the potential risk factors (16):

$$PERI = \sum_{i=0}^n Er^i \quad (16)$$

The following terminology is used to describe the potential ecological risk index (PERI) [16]:

PERI < 150	Low ecological risk
150 < PERI < 300	Moderate ecological risk
300 < PERI < 600	Considerable ecological risk
PERI > 600	Very high ecological risk

#### 4.14. Assessment of Potential Human Health Risk (HHRA)

The trophic transfer of HMs within aquatic ecosystems, such as coastal lagoons, has significant implications for coastal life and human health. Numerous studies have investigated the human health risks associated with heavy metal pollution in sediments in recent years [43–47]. Given the persistent nature of HMs in the environment, they accumulate in living organisms and undergo transfer from one trophic level to another within food chains [48]. Exposure of humans to HMs can occur via ingestion, inhalation, and dermal contact, confirming potential health risks. The following equations were utilized to estimate the chronic daily intake (CDI) values resulting from exposure to HMs through these two distinct pathways (Equations (17) and (18)):

$$CDI_{Ingestion} = \frac{C_{sed} \times IngR \times EF \times ED \times CF}{BW \times AT} \quad (17)$$

$$CDI_{Dermal} = \frac{C_{sed} \times CF \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \quad (18)$$

$C_{sed}$  represents the concentration of HMs in sediment samples (ppm),  $IngR$  denotes the sediment ingestion rate (mg/day),  $EF$  stands for exposure frequency (days/year),  $ED$  represents the exposure duration (years),  $BW$  is the average body weight (kg),  $AT$  indicates the averaging time (days),  $CF$  is the conversion factor (kg/mg),  $SA$  is the surface area of the skin in contact with the soil (cm<sup>2</sup>),  $AF$  sediment is the skin adherence factor for sediment (mg/cm<sup>2</sup>), and  $ABS$  is the dermal absorption factor. The exposure factors used to estimate the Chronic Daily Intake (CDI) are detailed in the following (Table 1):

**Table 1.** Values of parameters used for non-carcinogenic risk assessment [48].

Symbol	Parameter	Value
IngR	Ingestion rate	100 mg/day (adult), 200 mg/day (children)
EF	Exposure frequency	350 days
ED	Exposure duration	24 years (adult), 6 years (children)
BW	Body weight	70 kg (adult), 15 kg (children)
AT	Averaging time	365 × ED adult/children
CF	Conversion factor	1 × 10 <sup>−6</sup> kg/mg
SA	Surface area	5700 cm <sup>2</sup> event <sup>−1</sup>
AF	Adherence factor for sediment	0.07 mg/cm <sup>2</sup>
ABS	Gastrointestinal absorption factor	0.001

In the Health Risk Assessment (HHRA), the calculated bioavailability concentration of HMs in the lagoon was used to assess both carcinogenic and non-carcinogenic risk exposures for both children and adults [49]. The Hazard Index (HI), indicative of the cumulative

non-carcinogenic/cancer risk, is determined by summing all the Hazard Quotients (HQ) as outlined in Equations (19) and (20) [49]:

$$HQ = \frac{CDI}{RfD} \quad (19)$$

$$HI = \sum HQ = HQ_{Ingestion} + HQ_{Dermal} \quad (20)$$

The reference dose (RfD) utilized in the health risk follows the directives established by the United States Environmental Protection Agency [50], as detailed in (Table 2). A Hazard Index (HI) value below 1 suggests an absence of significant risk for non-carcinogenic effects. On the contrary, if the HI exceeds 1, there is a potential for non-carcinogenic effects to manifest.

**Table 2.** The reference dose (RfD) and the Cancer Slope Factor (CSF) values of HMs [50].

HMs	RfD (mg/kg/day)	CSF
Cr	0.003	0.5
Pb	0.0035	0.0085
Cu	0.0371	-
Co	0.02	-
Ni	-	0.84
Zn	0.3	-

Assessment of health risks associated with carcinogenic metals involves calculating the total lifetime cancer risk (LCR). This value is derived from Equations (21) and (22), representing the total value of cancer risk for each heavy metal in the lagoon ecosystem. The cancer slope factor (CSF) values for Cr, Pb, Ni, Cu, Co, and Zn are provided in (Table 2) [50]. The tolerable threshold value for cancer risk is  $1.0 \times 10^{-4}$ , while the acceptable LCR range is between  $1.0 \times 10^{-6}$  and  $1.0 \times 10^{-4}$  [51].

$$LCR = CDI \times CSF \quad (21)$$

$$\sum LCR = (LCR_{ing} + LCR_{derm}) \quad (22)$$

## 5. Results

### 5.1. Contamination Factor and PLI

Metal contamination factors in Nador lagoon are shown in (Figure 3). The highest Cf values were found in S, Ce, sulfur and Pb of 5.34, 4.70, and 3.69, respectively, which indicates a considerable degree of contamination. Otherwise, Nb, Nd, Zn, Co, V, Sc, Cu, Th, Zr, Hf, Ba, and Ga showed a moderate degree of contamination ( $2.04 > 1.96 > 1.90 > 1.88 > 1.82 > 1.80 > 1.74 > 1.73 > 1.36 > 1.28 > 1.13 > 1.11$ , respectively), while La, Ni, Rb, Ce, Y, and Cr expressed the lowest contamination with  $0.98 > 0.96 > 0.87 > 0.83 > 0.61 > 0.23$ , respectively.

The Pollution Load Index (PLI) ranged from 0.9 to 1.04, with a mean value of 1.009. Overall, PLI showed values greater than one except for Ce, Cr, Rb, and Y; therefore, the results indicate that sediments are generally polluted by HMs (Figure 3).

The modified contamination degree (mCd) values expressed a high degree of contamination by Sr and sulfur with 5.34 and 4.57, respectively. Pb and Nb showed moderate contamination in the sediment with 3.51 and 2.04, respectively. A low degree of contamination was approximately by Nd = 1.96, Zn = 1.88, Co = 1.87, V = 1.82, Sc = 1.79, Cu = 1.736, and Th = 1.735, and a very low degree by Zr, Hf, Ba, Ga, La, Ni, Rb, Ce, Y, and Cr (Figure 4).

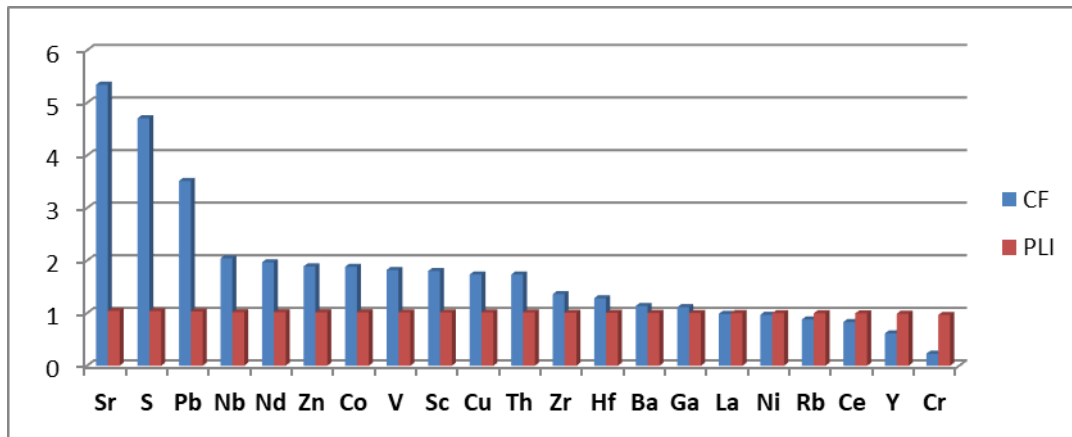


Figure 3. Contamination factor (Cf) and Pollution load index (PLI) in sediment samples of Nador lagoon sediments.

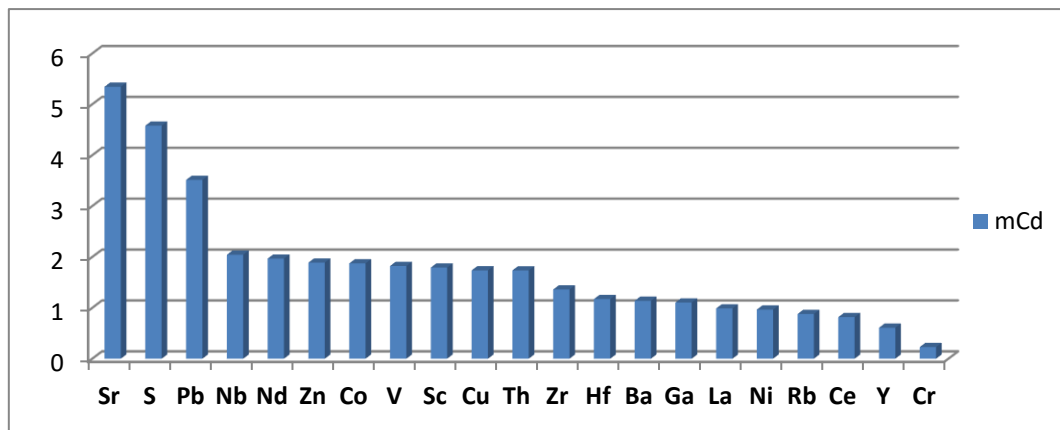


Figure 4. Variation in modified contamination degree index (mCd) in sediment.

5.2. Potential Contamination Index (Cp)

The potential contamination index (Cp) values indicated a severe contamination degree by Cr, S, Sr, Ce, Ni, Pb, Zn, and Nb with  $8.28 > 7.99 > 5.48 > 4.01 > 3.66 > 3.64 > 3.43$  and 3.13, respectively. Moderate contamination in lagoon sediment was observed by  $V = 2.67$ ,  $Th = 2.62$ ,  $Cu = 2.02$ ,  $Hf = 1.75$ ,  $Co = 1.62$ ,  $Nd = 1.44$ ,  $Sc = 1.42$ ,  $Ba = 1.41$ ,  $Zr = 1.40$ , and  $Rb = 1.04$ . At the same time, a low degree of contamination was assessed for Ga, La, and Y with  $0.87 > 0.80$  and 0.47, respectively (Figure 5).

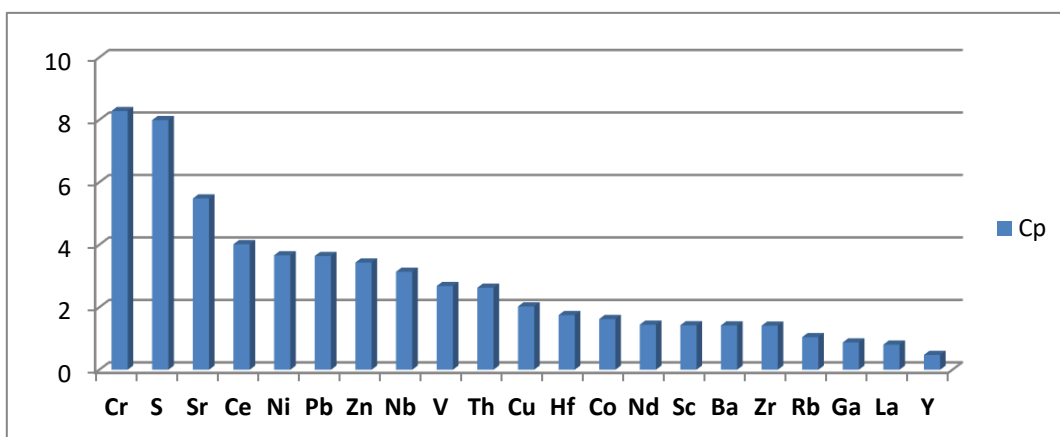
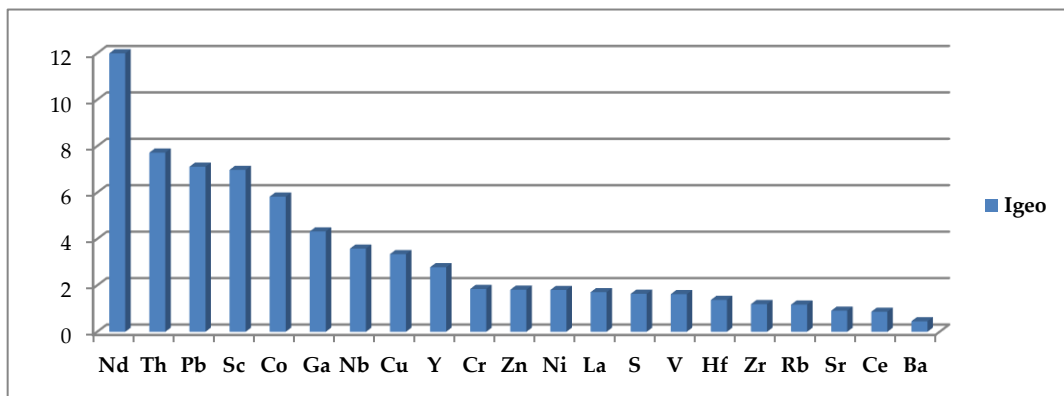


Figure 5. Potential contamination index (Cp) in the sediments of the Nador lagoon.

### 5.3. Geo-Accumulation Index (Igeo)

The results of the calculated Igeo factor show that Nd, Pb, Th, Sc, Co, and Ga ranged from strongly polluted to extremely polluted with  $11.99 > 7.71 > 7.10 > 6.97 > 5.82$  and  $4.32$ , respectively. Moderate to strong pollution is indicated by Nb = 3.58, Cu = 3.34, and Y = 2.78. Cr, Zn, Ni, La, V, S, Hf, Zr, and Rb demonstrate moderate pollution in the lagoon ecosystem with  $1.84 > 1.81 > 1.80 > 1.70 > 1.62 > 1.60 > 1.37 > 1.19$  and  $1.17$ , respectively. Sr, Ce, and Ba show a low to moderate degree of pollution with  $0.90 > 0.86$  and  $0.45$ , respectively (Figure 6).

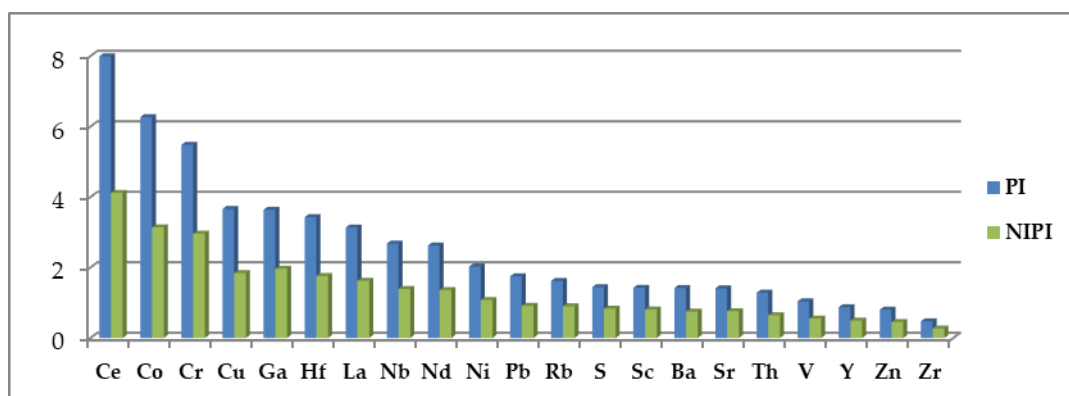


**Figure 6.** Geo-accumulation index (Igeo) in the sediments of the Nador lagoon.

### 5.4. Pollution Index (Pi) and Nemerow Integrated Pollution Index (NIPI)

A high level of pollution was detected in sediments by  $S = 7.99 > Ce = 6.2 > Sr = 5.48 > Ni = 3.66 > Pb = 3.64 > Zn = 3.43$  and Nb with  $3.13$ . V > Th and Cu confirm a moderate level of pollution with  $3.67 > 2.62$  and  $2.02$ , respectively. A low degree of pollution was detected in sediments and calculated by  $Hf = 1.75 > Co = 1.62 > Nd = 1.44 > Sc = 1.42 > Ba = 1.41 > Zr = 1.40 > Cr = 1.28 > Rb = 1.04$ .  $Ga = 0.87 > La = 0.80$  and  $Y = 0.47$ .

The NIPI results indicate that the lagoon sediments are highly contaminated by sulfurs and Ce with  $4.11 > 3.1$ , respectively. Sr detected moderate contamination with  $2.96$ , and a low contamination originated by  $Pb = 1.96 > Ni = 1.84 > Zn = 1.76 > Nb = 1.62 > V = 1.39 > Th = 1.36$  and Cu with  $1.07$ . Hf > Co > Nd > Sc > Zr and Ba recorded a quite high level of pollution and was registered by  $Hf > Co > Nd > Sc > Zr$  and Ba with  $0.91 > 0.90 > 0.83 > 0.80 > 0.76$  and  $0.74$ , respectively. No pollution level was related to the concentration of  $Cr > Rb > Ga > La$  and Y with  $0.64 > 0.55 > 0.49 > 0.45$  and  $0.26$ , respectively (Figure 7). Overall, these two indices indicate that the lagoon sediments are moderately to highly contaminated by HMs (Figure 7).



**Figure 7.** Pollution index (Pi) and Nemerow integrated pollution index (NIPI) in the sediments of the Nador lagoon.

### 5.5. Nemerow Pollution Index and Modified Pollution Indices (PI Nemerow—MPI)

Heavy contamination was detected by sulfurs > Ce and Sr with  $5.82 > 4.43$  and  $4.18$ , respectively. Pb > Ni > Zn and Nb were perceived with severe pollution in the lagoon of  $2.77 > 2.60 > 2.48$  and  $2.29$ , respectively. Moderate contamination was suggested by  $V = 1.97 > Th = 1.92 > Cu = 1.52 > Hf = 1.28 > Co = 1.27 > Nd = 1.17 > Sc = 1.13 > Zr = 1.07$  and Ba with  $1.05$ , respectively. These factors detected Cr > Rb > Ga > La and Y as unpolluted to slightly polluted elements in the lagoon sediments with  $0.91 > 0.78 > 0.69 > 0.63$  and  $0.38$ , respectively (Table 3).

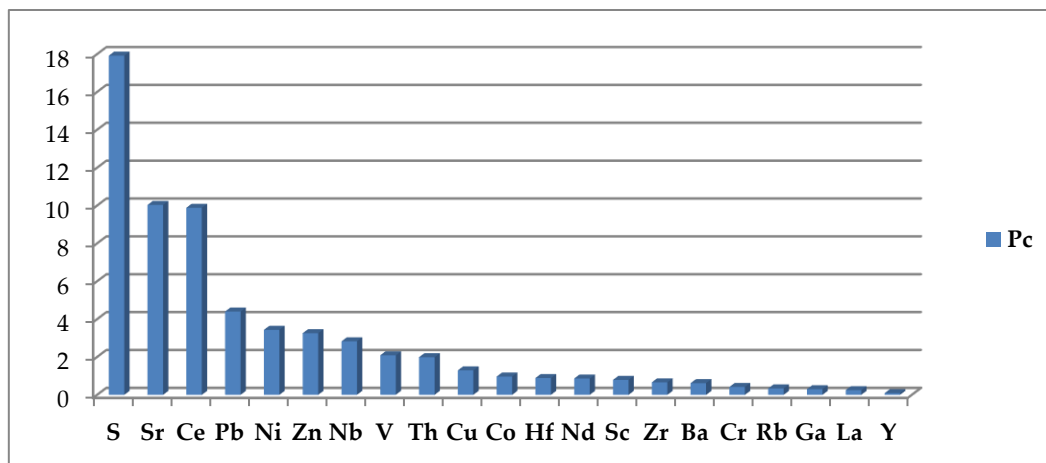
**Table 3.** Nemerow Pollution Index (PI) and Modified Pollution Index (MPI).

HMs	PI	MPI
Ba	1.06	3.71
Ce	4.44	31.29
Co	1.27	8.79
Cr	0.91	10.46
Cu	1.52	6.88
Ga	0.70	2.32
Hf	1.29	3.39
La	0.64	7.99
Nb	2.30	3.11
Nd	1.17	16.93
Ni	2.61	4.64
Pb	2.78	8.08
Rb	0.78	1.89
S	5.82	1420.02
Sc	1.14	1.00
Sr	4.19	84.24
Th	1.93	8.50
V	1.97	7.17
Y	0.38	1.72
Zn	2.49	4.52
Zr	1.08	4.90

The results of the modified pollution index show very heavy contamination of the lagoon sediments by sulfur > Sr and Ce with  $1420.06 > 84.24$  and  $31.29$ , respectively. Meanwhile, heavy contaminations by Nd and Cr were detected at  $16.92$  and  $10.45$ , respectively. Severe contamination was detected by  $Co > Th > Pb > La > V$  and Cu with  $8.78 > 8.50 > 8.08 > 7.98 > 7.16$  and  $6.87$ , respectively. Moderate to heavy pollution was checked by  $Zr = 4.89 > Ni = 4.63 > Zn = 4.52 > Ba = 3.71 > Hf = 3.39$  and Nb with  $3.10$ , respectively, and we can add Ga with  $2.3$  through moderate contamination. Finally, slightly polluted indications were given by Rb > Y and Sc with  $1.89 > 1.72$  and  $1$ , respectively (Table 3).

### 5.6. Nemerow Multi-Factor Index (Pc)

Nador lagoon sediments are moderate to severely polluted in the overall pollution classification,  $3.01$  being the average Nemerow multifactor. The severe level of contamination (severely contaminated sediments) was detected by  $S > Sr > Ce > Pb > Ni$  and Zn with  $17.91 > 10.03 > 9.88 > 4.40 > 3.43$  and  $3.25$ , respectively. A moderate level of contamination (moderately contaminated sediments) was found in the lagoon sediments by Nb and V with  $2.82$  and  $2.08$ , respectively. Light levels of contamination (sediments slightly contaminated) were detected in sediments by Th and Cu with  $1.98$  and  $1.02$ , respectively. Alert levels (still clean) were laid bare by Pc in the sediments of the lagoon by  $Co > Hf > Nd > Sc > Zr$  with  $0.96 > 0.88 > 0.86 > 0.78$  and  $0.7$ , respectively. Ba > Cr > Rb > Ga > La and Y were revealed to be safe (clean) with  $0.61 > 0.41 > 0.33 > 0.29 > 0.24$  and  $0.08$ , respectively (Figure 8).



**Figure 8.** Nemerow Multifactor (Pc) values in the sediments of the Nador lagoon.

From an integrated assessment point of view, the contamination risk of HMs in the surface sediments for the study area indicates a moderate to high-risk level, which was dominated by S, Sr, Pb, Nb, Zn, Ce, and Ni.

#### 5.7. Spatial Distribution of HMs in Sediments Based on Contamination Factors

The spatial distribution of HMs allows us to identify potentially hazardous contaminated areas following the results of the calculated contamination indices. The maps depicted in (Figure 9) illustrate areas within the lagoon characterized by elevated or reduced contamination levels in sediment samples. The spatial patterns of Cf, Cd, and Igeo PI (Nemerow) and Pc show significant similarity in detecting high contamination levels at the mouth of the Bousardoun river that crosses the agglomeration of Nador, as well as the Akhandouk river of Akhandouk of Beni ensar, and the Afelioun river of Afelioun (kariat Arekman), in addition to the mountainside of Atalayoun. This high contamination can be also spotted and highlighted by Igeo and the contamination factor (Cf) near the old pass (Figure 9). Cf, MPI, Cd, and PI (Nemerow) showed moderate degrees of contamination in the central area of the lagoon, potentially corresponding to sources of pollution derived from agricultural areas in the southeast, as well as the area of marine influence in the north-northeast.

Based on these results, the areas of similar values of contamination are summarized in Figure 10. These values allowed us to individuate three major zones: low, moderate, and high contamination with 16.75% > 49.83% and 32.53% of dominance, respectively. Low contamination areas can be observed near the new pass (Figure 10) and in the NO of Atalayoun, which is limited between two areas of high contamination (Beni Ensar), and also with a spot that follows the alignment of the Nador nature park. Moderate contamination can be observed in the central part of the lagoon, from the continental-influenced area to the littoral zone. High contamination by HMs was mainly located in the confined area (Beni Ensar) and the confined area of Kariat Arekman, in the delta of the Oued Bousardoune, in front of the purification station (STEP Nador), and in the old pass.

The comprehensive analysis of contamination indices, including Cf, Pc, and NIPI, has revealed a notable convergence in the identification and highlighting of heavy metals of concern, specifically S, Sr, Pb, Ni, and Zn, in lagoon sediments. The consistency across these indices underscores their robustness in characterizing and assessing the contamination levels. This convergence improves the reliability of the assessment, providing a more comprehensive understanding of the heavy metal contamination profile in the Nador lagoon sediments. These indices are especially valuable and practical for focused monitoring and remediation initiatives in comparable ecosystems.

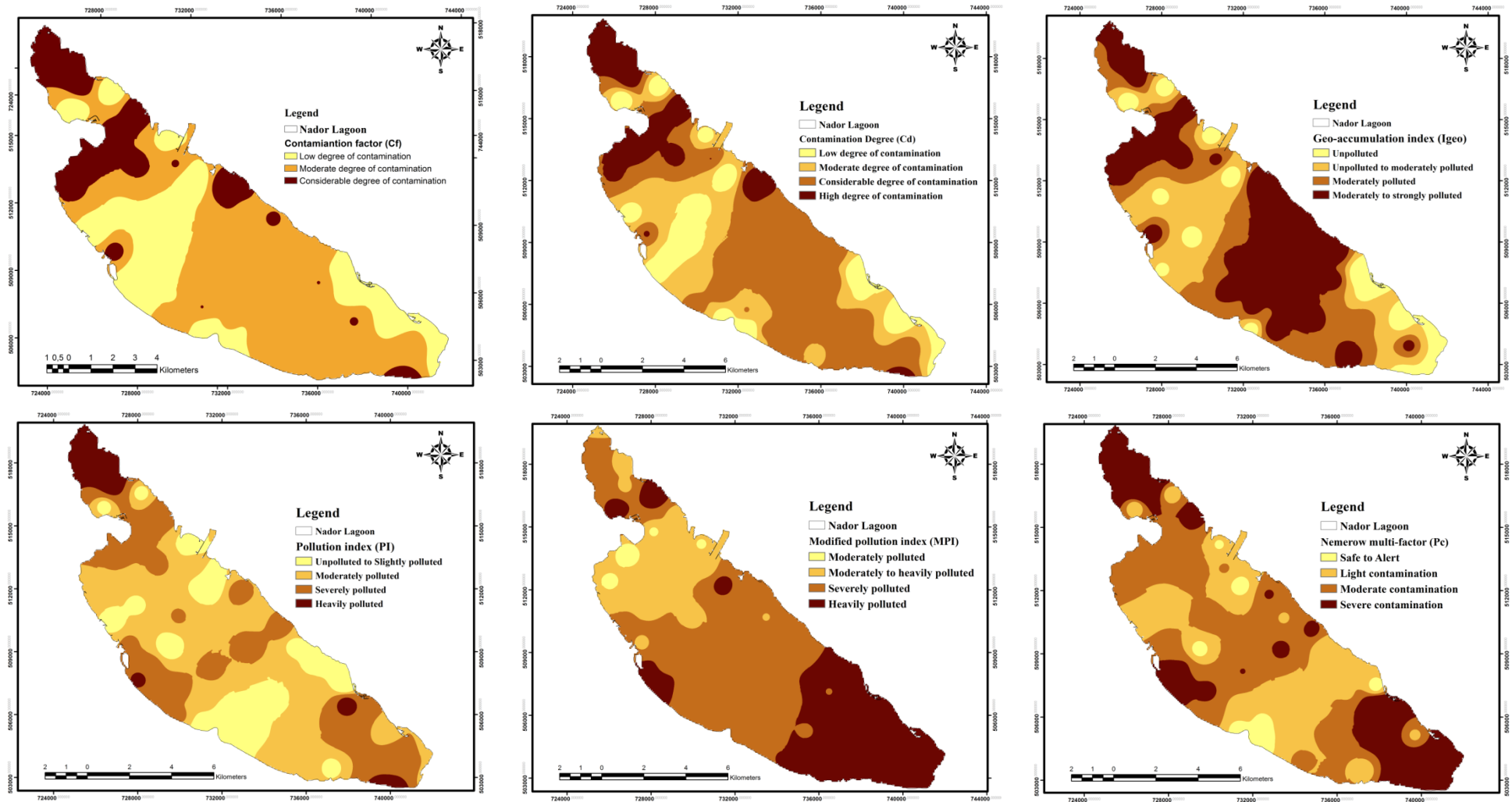
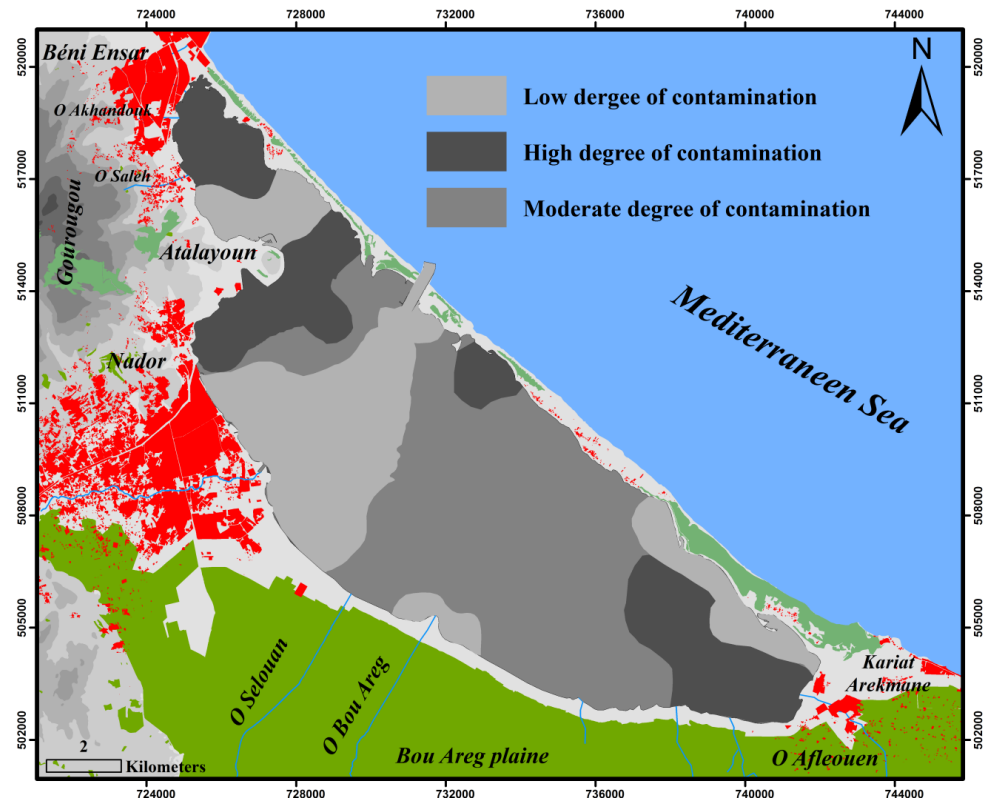


Figure 9. Spatial distribution of the calculated contamination factors (Cf, Cd, Igeo PI Nemerow—MPI, and Pc, respectively).



**Figure 10.** HMs contamination areas in the sediments of the Nador lagoon.

### 5.8. Sediment Quality Guidelines (SQGs)

The results show that the mean value of Cr was higher than TEL, LEL, and MET in 58%, 64%, and 36% of the samples, respectively. Although Cr was lower than the PEL, ERM, SEL, ERL, and TET (Table 4), only 2% of the data samples were higher than these guidelines. Cu was higher than LEL and MET with 16 and 28 in the sediment quality guidelines, and higher than TEL, LEL, and MET in 54%, 68%, and 60% of the samples, respectively. The mean concentration of Ni observed in the samples was higher than LEL, TEL, LEL, MET, ERL, and PEL in 60%, 60%, 20%, 32%, and 14% of the samples, respectively. Pb showed higher values than TEL, LEL, MET, and ERL in 42%, 48%, 32%, and 42% in all samples, respectively, and was found to be lower than the mid-range effect sediment quality guidelines (PEL and ERM) and the extreme effect sediment quality guidelines (SEM and TET). Finally, Zn was higher than TEL, LEL, and ERL in 16% of all the samples.

Cr, Ni, and Pb were higher than TEL, revealing that a toxic response has started to be observed in benthic organisms of Nador lagoon; on the other hand, the evaluated HMs were lower than PEL, which indicates that not a large percentage of the benthic population shows a toxic response [37].

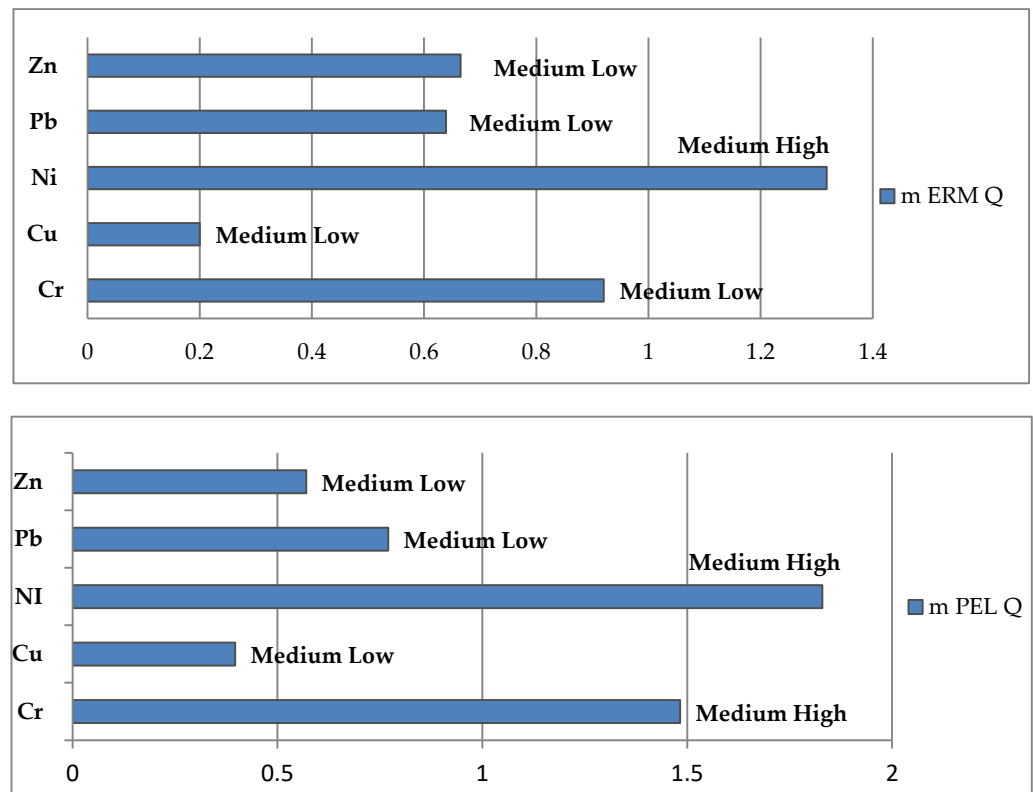
### 5.9. Mean ERM Quotient (*m*-ERM-*Q*) and Mean PEL Quotient (*m*-PEL-*Q*)

The mean ERM and PEL quotients serve as quantifications of the likelihood of significant adverse effects on the Nador lagoon ecosystem resulting from the combined impact of HMs. The findings are depicted in (Figure 11). The *m*-ERM-*Q* indicates a moderate probability of ecosystem hazard, corresponding to a high-risk level for Ni (1.3). Simultaneously, Ni and Cr emerge as the HMs exerting the most substantial ecological impact on the biota, with values of 1.8 and 1.5, respectively. The spatial distribution of these two calculated factors exhibits a high degree of similarity, indicating a prevalent medium-to-high-risk scenario, particularly in the confined Area II. Therefore, both *m*-ERM-*Q* and *m*-PEL-*Q* affirm a moderate ecological hazard posed by HMs in the Nador ecosystem.

**Table 4.** Comparison between sediment quality guidelines (SQGs), threshold effect concentrations (TEC), and Probable Effect Concentrations (PEC) and mean of HMs concentrations in sediments. Metals are reported in ppm.

<i>Nador Lagoon Samples = 50</i>				<i>Sediment Quality Guidelines (SQGs)</i>							
<b>Metals</b>	<b>Min–Max</b>	<b>Mean–Std.Dev.</b>	<b>GB<sup>b</sup></b>	<b>Threshold Effect Sediment Quality Guidelines</b>				<b>Midrange Effect Sediment Quality Guidelines</b>		<b>Extreme Effect Sediment Quality Guidelines</b>	
				<b>TEL<sup>a</sup></b>	<b>LEL<sup>a</sup></b>	<b>MET<sup>a</sup></b>	<b>ERL<sup>a</sup></b>	<b>PEL<sup>a</sup></b>	<b>ERM<sup>a</sup></b>	<b>SEL<sup>a</sup></b>	<b>TET<sup>a</sup></b>
<b>Cr</b>	0.6–745.3	56.036–103.070	90	37.3	26	55	80	90	145	110	100
<b>Cu</b>	0.6–91.2	32.814–21.134	45	35.7	16	28	70	197	390	110	86
<b>Ni</b>	0.6–249	27.574–38.277	68	18	16	35	30	36	50	75	61
<b>Pb</b>	0.6–72.8	29.518–20.368	20	35	31	42	35	91.3	110	250	170
<b>Zn</b>	0.6–325.9	75.358–58.611	95	123	120	150	120	315	270	820	540

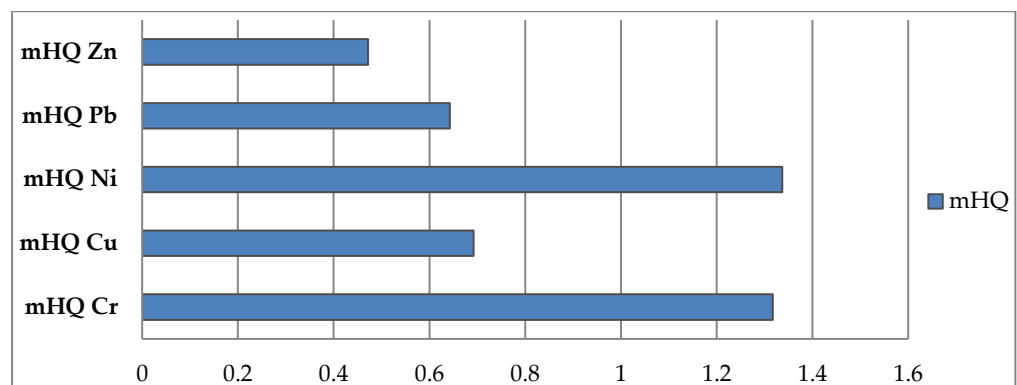
Note: <sup>b</sup> GB, Geochemical Background (average shale standard concentrations); [17]. <sup>a</sup> Threshold effect sediment quality guidelines; TEL threshold effect level, LEL lowest effect level, MET, Minimal Effect Threshold, ERL, Effects Range Low; Mid-range effect sediment quality guidelines: PEL, Probable Effects Level, ERM, Effect Range Median; Extreme effect sediment quality guidelines; SEL Severe Effect Level, TET Toxic Effect Threshold; [36].



**Figure 11.** Variation in the mean ERM Quotient (m-ERM-Q) and mean PEL Quotient (m-PEL-Q) in the Nador lagoon.

#### 5.10. Modified Hazard Quotient (m-HQ)

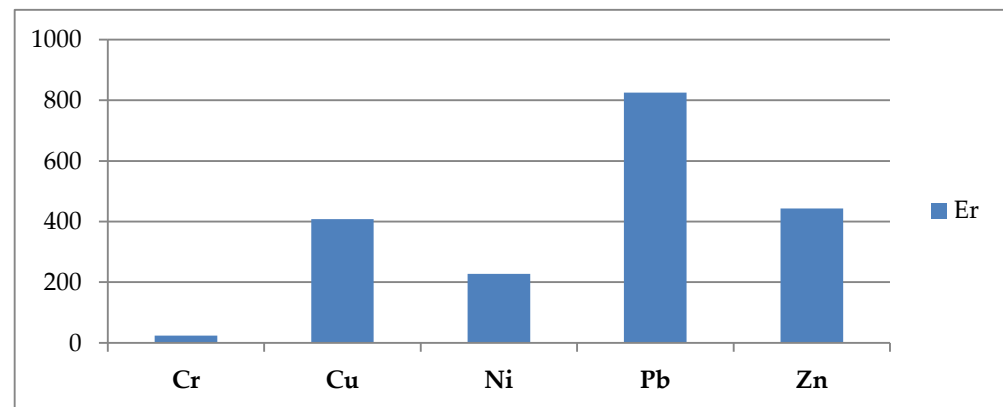
The average values of the modified hazard quotient (m-HQ) highlighted a moderate severity of contamination by HMs, dominated by Ni with 1.33 and followed by Cr with 1.31. Cu, Pb, and Zn showed a low severity of contamination with  $0.69 > 0.64$  and  $0.47$ , respectively (Figure 12).



**Figure 12.** Quantification of the modified hazard quotient of HMs in Nador lagoon.

#### 5.11. Evaluation of Potential Ecological Risk Index (PERI)

Potential ecological factor results draw attention to the important effect of HMs on the lagoon biota (Figure 13). Pb, Zn, and Cu indicated a significant (very high) ecological risk with an overall value of  $825.35 > 443.64$  and  $407.63$ , respectively. A high potential ecological risk manifested by Ni with 227.47. Furthermore, Cr confirmed a low ecological risk with 24.28. Consequently, the total study area can be assigned with very high ecological risk (PERI: 1928.43).



**Figure 13.** Variation in the potential ecological risk factor (Er) in surficial sediment.

### 5.12. Pollution Spatial Distribution

The distribution of HMs pollution indices in the sediment of the Nador lagoon had a certain spatial homogeneity (Figure 14). The spatial distribution of M-ERM-Q, M-PEL-Q, and mHQ was consistent, and the high-value areas were located near Kariat Arekmane in the southeast area, while the high-value areas of RI were mainly located in the north part of the lagoon, near the Atalayoun hill, Beni Ensar, and near the old pass. Potential moderate levels of pollution were located alongside agriculture and coastal areas controlling most of the lagoon sediment surface, indicating a relative uniqueness in the spatial distribution by M-ERM-Q, M-PEL-Q, mHQ, and RI. A relatively uniform distribution that detects a low pollution potential as a result of M-ERM-Q, M-PEL-Q, and mHQ values was located for the most part in the middle of the lagoon with a homogeneous form near the Nador agglomeration and wastewater treatment plant.

The indices were used to establish a classification zone representing the overall pollution by HMs in the lagoon (Figure 15). The high pollution potential, which accounts for 18% of the total pollution potential, is located almost in the high contamination zones, i.e., the two zones confined to the NW and SE perimeters of the lagoon (Beni Ensar and Kariat Arekmane), at Atalayoune in contact with the barrier beach, in front of the town of Nador, and the area of the old pass. The moderate potential for heavy metal pollution covers most of the lagoon (52%). Finally, the low potential pollution constitutes 30% in the lagoon; it can be detected in the SE zone of the barrier beach, at the mouth of O. Bou Areg, and can be seen following the distribution of areas with a low risk of contamination.

The evaluation of pollution indices, specifically sediment quality guidelines and the potential ecological risk index, has demonstrated their distinct utility in comprehensively understanding the pollution levels attributable to heavy metals in lagoon sediments. These indices stand out by consistently illuminating the degree of pollution, providing a nuanced perspective on the environmental impact. The robustness and reliability of these measures make them particularly valuable tools for scientifically assessing and monitoring heavy metal pollution in lagoon ecosystems. These indices can be employed to acquire a thorough comprehension of pollution dynamics, aiding in the development of focused mitigation strategies and informed decision making for sustainable environmental management.

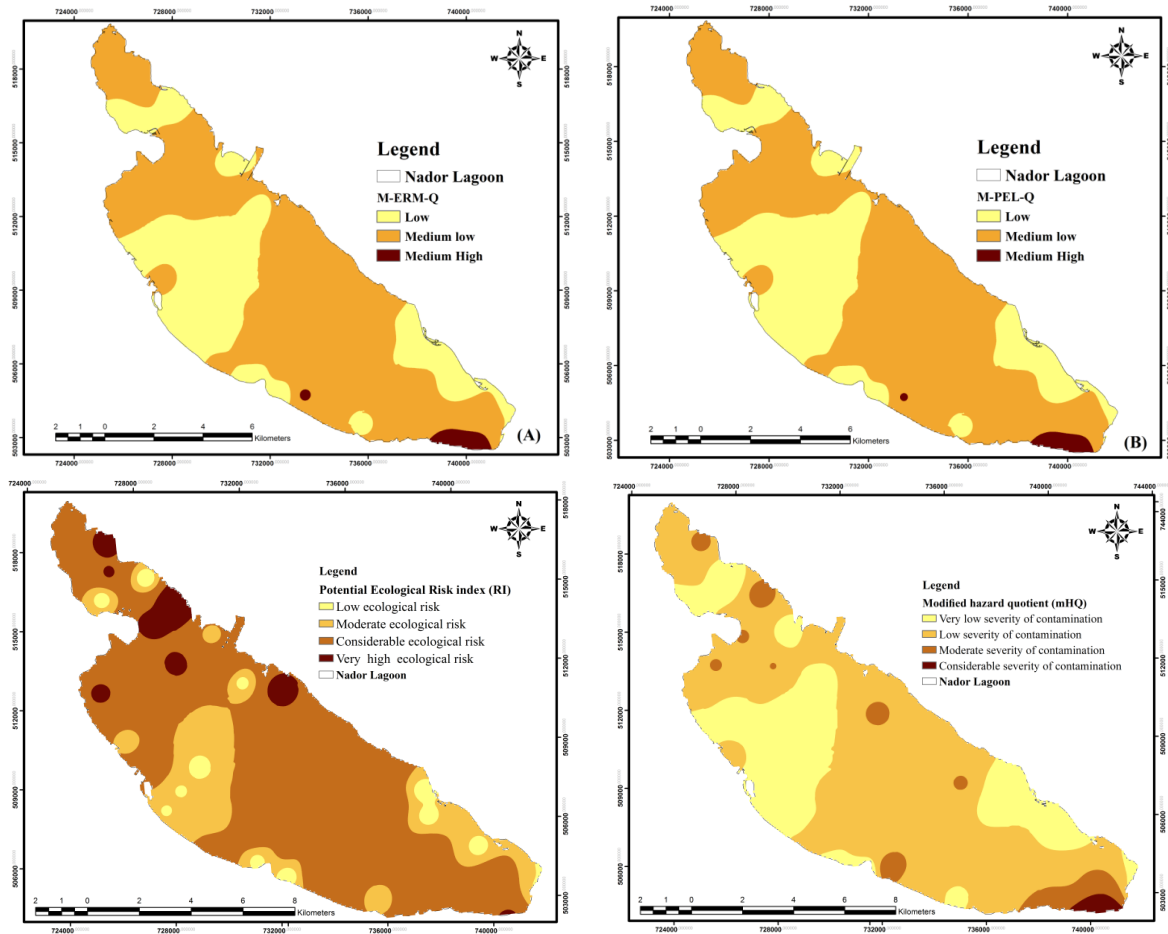


Figure 14. Spatial distributions of (A) mean ERM quotient (m-ERM-Q) and (B) mean PEL quotient (m-PEL-Q), PERI, and modified hazard quotient (mHQ).

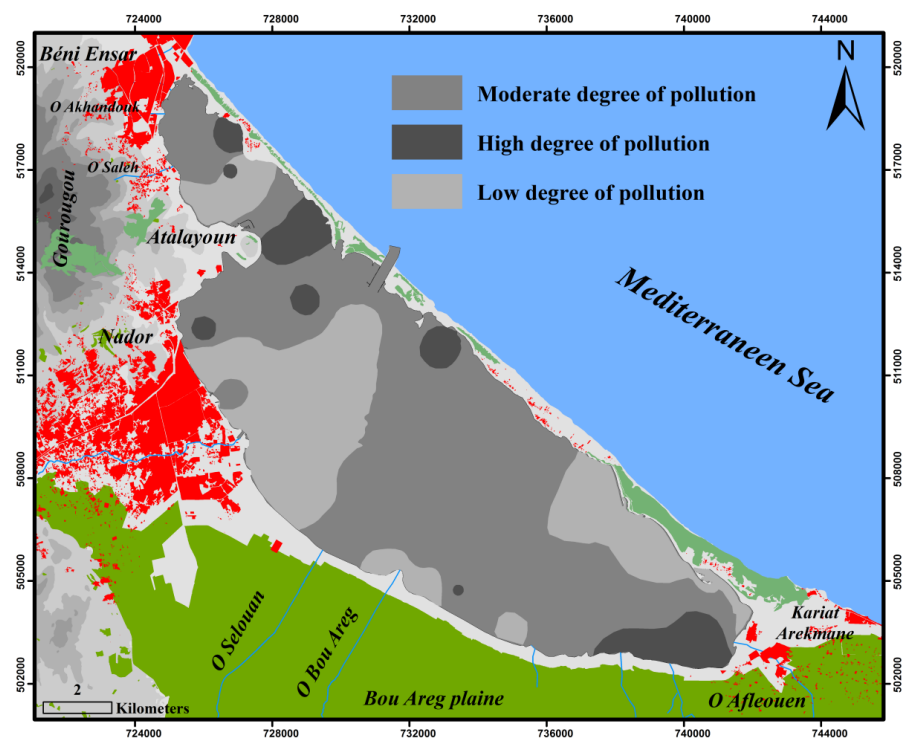


Figure 15. HMs pollution levels in Nador lagoon sediments.

### 5.13. Potential Assessment of Human Health Risk Quantification of Non-Carcinogenic Effects

The Hazard Index (HI) for non-carcinogenic risks in adults exhibited a descending order as follows: Cr ( $2.68 \times 10^{-2}$ ) > Pb ( $1.21 \times 10^{-2}$ ) > Cu ( $1.27 \times 10^{-3}$ ) > Co ( $1.08 \times 10^{-3}$ ) > Zn ( $3.61 \times 10^{-4}$ ) > Ni ( $4.65 \times 10^{-5}$ ). Whereas the non-carcinogenic risks for children, the HI for HMs decreases in the same order of HI for adults Cr > Pb > Cu > Co > Zn and Ni with  $6.24 \times 10^{-2} > 2.82 \times 10^{-2} > 2.95 \times 10^{-3} > 2.51 \times 10^{-3} > 8.39 \times 10^{-4} > 1.08 \times 10^{-4}$ , respectively.

Non-carcinogenic risks, as indicated by the Hazard Index (HI) for both adults and children, consistently registered values below one for each metal. This confirms the absence of potential non-carcinogenic risks arising from ingestion or dermal contact within the lagoon ecosystem (Table 5).

**Table 5.** Hazard quotient and cumulative hazard index (HI, HQ) for non-carcinogenic risk calculations for adults and children.

Adults							
HHRA HMs	Mean	HQ Ing Min	Max	Mean	HQ Derma Min	Max	HI
Co	$2.15 \times 10^{-5}$	$1.42 \times 10^{-7}$	$4.40 \times 10^{-5}$	$8.2 \times 10^{-8}$	$5.4 \times 10^{-10}$	$1.6 \times 10^{-7}$	$1.08 \times 10^{-3}$
Cr	$8.01 \times 10^{-5}$	$8.57 \times 10^{-7}$	$1.06 \times 10^{-3}$	$3.0 \times 10^{-7}$	$3.2 \times 10^{-9}$	$4.0 \times 10^{-6}$	$2.68 \times 10^{-2}$
Cu	$4.69 \times 10^{-5}$	$8.57 \times 10^{-7}$	$1.30 \times 10^{-4}$	$1.7 \times 10^{-7}$	$3.2 \times 10^{-9}$	$4.9 \times 10^{-7}$	$1.27 \times 10^{-3}$
Ni	$3.95 \times 10^{-5}$	$8.57 \times 10^{-7}$	$3.56 \times 10^{-4}$	$1.5 \times 10^{-7}$	$3.2 \times 10^{-9}$	$1.3 \times 10^{-6}$	$4.65 \times 10^{-5}$
Pb	$4.22 \times 10^{-5}$	$8.57 \times 10^{-7}$	$1.04 \times 10^{-4}$	$1.6 \times 10^{-7}$	$3.2 \times 10^{-9}$	$3.9 \times 10^{-7}$	$1.21 \times 10^{-2}$
Zn	$1.08 \times 10^{-4}$	$4.2 \times 10^{-7}$	$4.66 \times 10^{-4}$	$4.1 \times 10^{-7}$	$1.6 \times 10^{-9}$	$1.7 \times 10^{-6}$	$3.61 \times 10^{-4}$
Children							
HHRA HMs	Mean	HQ Ing Min	Max	Man	HQ Derma Min	Max	HI
Co	$5.01 \times 10^{-5}$	$3.3 \times 10^{-7}$	$1.03 \times 10^{-4}$	$8.2 \times 10^{-8}$	$5.4 \times 10^{-10}$	$1.6 \times 10^{-7}$	$2.51 \times 10^{-3}$
Cr	$1.87 \times 10^{-4}$	$2 \times 10^{-6}$	$2.48 \times 10^{-3}$	$3.0 \times 10^{-7}$	$3.2 \times 10^{-9}$	$4.0 \times 10^{-6}$	$6.24 \times 10^{-2}$
Cu	$1.09 \times 10^{-4}$	$2 \times 10^{-6}$	$3.04 \times 10^{-4}$	$1.7 \times 10^{-7}$	$3.2 \times 10^{-9}$	$4.9 \times 10^{-7}$	$2.95 \times 10^{-3}$
Ni	$9.22 \times 10^{-5}$	$2 \times 10^{-6}$	$8.30 \times 10^{-4}$	$1.5 \times 10^{-7}$	$3.2 \times 10^{-9}$	$1.3 \times 10^{-6}$	$1.08 \times 10^{-4}$
Pb	$9.84 \times 10^{-5}$	$2 \times 10^{-6}$	$2.43 \times 10^{-4}$	$1.6 \times 10^{-7}$	$3.2 \times 10^{-9}$	$3.9 \times 10^{-7}$	$2.82 \times 10^{-2}$
Zn	$2.51 \times 10^{-4}$	$1 \times 10^{-6}$	$1.09 \times 10^{-3}$	$4.1 \times 10^{-7}$	$1.6 \times 10^{-9}$	$1.7 \times 10^{-6}$	$8.39 \times 10^{-4}$

In the toxicity assessment measure, noncancerous effect refers to an impact on the development, size, or overall functioning of the body or specific organs, such as the skin and central nervous system, without, however, causing the formation of cancer cells. For the Nador lagoon, the non-carcinogenic risk is qualified as acceptable and no action is required from the point of view of human health [52].

The cancer risk associated with HMs in surface sediment samples from the Nador lagoon is considered negligible, with lifetime cancer risk (LCR) values below the recommended limit for Co, Cr, Ni, and Zn for children and adults (according to [48]). This finding confirms the absence of any significant lifetime cancer risk for these elements (Table 5).

Based on lifetime cancer risk values, it appears that Cu and Pb are outside the recommended range ( $1.10 \times 10^{-6}$  to  $1.10 \times 10^{-4}$ ), indicating that long-term exposure through various pathways can increase cancer risk in the population surrounding the lagoon (Table 6).

**Table 6.** Lifetime cancer risk (LCR) values of carcinogenic human health risks through total exposure (ingestion and dermal contact) for adults and children in the study area.

LCR Adults						
HMs	Co	Cr	Cu	Ni	Pb	Zn
-	$2.15 \times 10^{-5}$	$4.02 \times 10^{-5}$	$1.58 \times 10^{-4}$	$3.33 \times 10^{-5}$	$3.60 \times 10^{-7}$	$1.29 \times 10^{-4}$
LCR × Children						
HMs	Co	Cr	Cu	Ni	Pb	Zn
-	$5.01 \times 10^{-5}$	$9.36 \times 10^{-5}$	$1.65 \times 10^{-4}$	$7.76 \times 10^{-5}$	$8.38 \times 10^{-7}$	$2.51 \times 10^{-4}$

## 6. Discussion

Heavy metal pollutants affect the coastal environment through industrial, agricultural, and sewage effluents from coastal cities and resorts [53]. They are also products of rock weathering that are transported to water bodies by rainfall and wind. Many factors influence the toxicity and availability of metals in coastal sediments, such as pH, dissolved oxygen, the concentration of metal ions, organic and inorganic carbon content, and the oxidation-reduction potential. In this study, toxicity and availability are assessed by assessing sediment quality through a holistic approach to pollution indicators, one of the main adopted strategies for understanding the ecological state of a certain environment. In fact, as reported by [54], the assessment of anthropogenic metal contributions in sediments can be assessed through various indices such as the contamination factor (Cf), the degree of contamination (Cd), the modified degree of contamination (mCd), and the potential contamination index (Cp). These indices offer a relative ranking of sampling sites. In this study, we applied a considerable number (16 indices were selected and evaluated) of the main indices for sediment pollution and quality. This allowed us, first of all, to give an overall picture of the environmental state of the Nador lagoon and, second, to compare these indices and their significance. In summary, the assessment of pollution indices is an excellent approach to assessing sediment quality. The results highlight specific contaminations in the Nador lagoon and their spatial distribution, recognizing the presence of metals also identified in previous research.

The overall ‘big picture of contamination and pollution’ reported in this study is the first attempt to display the ecological quality and human health risk of the entire Nador lagoon by using a very large number of different assessment methods. Overall, the maps (Figures 10 and 15) reported a vast area that ranged from moderate to potentially high pollution from north to south. These different contaminants have potentially originated from several pollution sources.

Our results indicate that the main contaminations derived from Sr, sulfurs, and Pb (following Cf, mCd, PI, and Pc), and they were principally located in the northern area and near the old pass channel (Figure 9). This result is similar to that reported by [11] even if they also indicated a high concentration of Zn and Cu.

The Cp values also indicated high concentrations of Cr, Ce, Ni, and Zn (the latest three with concentrations similar to Pb). These concentrations were relatively high (from 3.13 to 8.28; Figure 8) and could be derived from anthropogenic activities considering previous studies such as [11,55].

The spatial distribution of HMs based on Igeo Nemerow—MPI and Pc can show us some different trends. While Igeo partially confirmed the pollution in the northern section of the lagoon, it also highlighted the severe pollution in the middle part of the basin. Otherwise, MPI and Pc confirmed the pollution in the southern section (Pc also allowed us to appreciate severe but smaller contaminations near the Oued Bousardoun mouth—in the middle of the basin—and the northern part; Figure 9). In this regard, we recall that Igeo showed different HM contaminations compared to other indices. This may be because Igeo adopted background values that are not useful for detecting certain HMs (e.g., Sr, S), while it is more efficient in detecting Nd, Th, Pb, and Sc.

The highest amount of Cu, Zn, and Cd was found between the public treatment station of Nador and Tirekaa town, and of Cr, close to the effluent of the WTP (wastewater treatment plant) of Nador and Beni Ensar. The presence of elevated levels of Zn and Cd, in particular, suggested that the potential contamination could have originated from industrial, agricultural, or sewage effluents discharged into the lagoon. High concentrations of Zn and Cd can pose risks to aquatic organisms due to their toxicity and potential bioaccumulation in the food chain [56].

A higher concentration of Pb was detected near a Nador WTP effluent and an old fish-farming area. Values obtained for the Geo-accumulation index indicate the presence of an average polluted spot by Pb and Cu near a WTP of Nador, Beni Ensar, and the old fish-farming area. Pb is a toxic metal that can have severe impacts on aquatic ecosystems,

including affecting fish health and reproductive success. Identifying and mitigating sources of Pb contamination in these areas is crucial to protect the ecological integrity of the lagoon and the health of the population [57].

Sediments close to Kariat Arekmane exhibited elevated levels of Ni, Cr, and Cu. Similarly to Zn and Cd, these metals could stem from various anthropogenic activities in the vicinity. Ni, Cr, and Cu are known to have toxic effects on aquatic life and can disrupt the functioning of the ecosystem. The presence of these metals highlights the importance of identifying and addressing pollution sources in Kariat Arekmane to prevent further ecological consequences [8].

Cr concentrations were notably detected near the effluent of the Nador water treatment plant (WTP) and Beni Ensar. The presence of Cr in these areas could be attributed to industrial discharges or other human activities. Cr is a dangerous heavy metal, and its accumulation in sediments can adversely affect benthic organisms and species that inhabit sediments. Close monitoring and appropriate treatment of wastewater from the Nador WTP and other potential sources are necessary to minimize Cr contamination [58].

The dispersion of HMs in the Nador lagoon implies multiple sources of pollution and potential ecological risks. Restoration actions have been carried out around this lagoon in the last decade to protect its ecological value and develop tourist activity, but without real improvement of the ecological state of the lagoon [12]. Therefore, identified contamination hotspots require thorough investigation and effective management strategies to mitigate the impacts on aquatic organisms, biodiversity, and overall ecosystem health. Regular monitoring and continued efforts to reduce pollutant input are essential for long-term restoration and protection of ecological integrity [59]. These additional considerations provide further information on the distribution and concentrations of HMs within the Nador lagoon. The specific areas mentioned highlight the hotspots of pollution for different metals, highlighting the need for targeted monitoring and management strategies to mitigate the ecological impacts associated with heavy metal pollution in these specific locations [60].

Concerning non-carcinogenic risks, population exposure levels to HMs are considered acceptable and no immediate action is required for non-carcinogenic health effects. However, there is a potential risk of cancer associated with exposure to Cu and Pb, which may warrant further monitoring and assessment, or consideration of preventive measures to mitigate the risk.

Contamination and pollution indices play vital roles in environmental management, as they can help identify the presence and concentrations, guide targeted management efforts, and provide benchmarks for sediment quality assessment, signaling potential harm to aquatic ecosystems. These indices quantify contamination levels precisely, allowing the prioritization of areas that need immediate attention, and the categorization of pollution levels, forming the basis for prioritizing management actions [61]. For risk assessment, contamination indices managed to incorporate risk assessments, crucial for understanding the urgency and nature of required mitigation strategies. Pollution indices explicitly assess ecological risks associated with pollutant concentrations, guiding the development of risk-based management strategies. Contamination and pollution indices are also crucial in mitigation strategies, as they pinpoint specific contaminants, including source control, remediation, and targeted interventions, and guide the formulation of mitigation measures by highlighting pollutants exceeding acceptable levels, involving strategies like sediment remediation, habitat restoration, or regulatory measures. Both sets of indices provide a foundation for ongoing monitoring and adaptive environmental management, contributing to a dynamic understanding of changing environmental conditions [62].

## 7. Conclusions

This study investigates the impact of heavy metal pollutants on the coastal environment, primarily influenced by industrial, agricultural, and sewage effluents from coastal areas. Pollutants also result from rock weathering and are transported to water bodies through rain and wind. The study adopts a holistic approach to pollution indicators,

employing indices such as the contamination factor (Cf), degree of contamination (Cd), modified degree of contamination (mCd), and potential contamination index (Cp) to assess sediment quality.

Seventeen pollution indices are applied to assess sediment pollution and quality in the Nador lagoon, offering a comprehensive understanding of its environmental state. The results reveal specific contaminations and their spatial distribution, identifying metals previously documented in other research. The study pioneers a comprehensive assessment of the entire Nador lagoon, using various assessment methods to present the 'big picture of contamination and pollution'. In conclusion, the HMs present in the Nador lagoon could have several potential ecological impacts. These impacts can vary depending on the specific metals involved, their concentrations, and the sensitivity of the local ecosystem.

Spatial distribution analysis indicates significant contamination, particularly in the northern area and near the old pass channel. Certain metals, such as Sr, Pb, and sulfur, emerge as primary contaminants, consistent with previous findings. The study highlights potential anthropogenic sources for Cr, Ce, Ni, and Zn concentrations, highlighting the importance of considering previous research in interpreting the results.

The research detects elevated levels of Cu, Zn, Cd, and Pb, suggesting potential origins from industrial, agricultural, or sewage effluents. Specific areas, such as those near the Nador public treatment station and Tirekaa town, exhibit high concentrations of Cu, Zn, and Cd, posing risks to aquatic organisms. Pb concentrations near effluent from a wastewater treatment plant and an old fish-farming area indicate potential ecological risks and emphasize the need for source identification and mitigation.

Sediments near Kariat Arekmane reveal elevated levels of Ni, Cr, and Cu, which are known for their toxic effects on aquatic life. Monitoring and addressing pollution sources in this area are crucial to preventing further ecological consequences. The study highlights contamination hotspots that require thorough investigation and effective management strategies to mitigate ecological impacts.

Despite restoration actions in the last decade, the ecological state of the lagoon has not improved significantly. Identified contamination hotspots require targeted monitoring and management strategies to protect aquatic organisms, biodiversity, and overall ecosystem health.

Regarding non-carcinogenic risks, population exposure levels to heavy metals are considered acceptable, except for Cu and Pb, which pose potential cancer risks. This suggests a need for further monitoring and preventive measures.

Contamination and pollution indices play a vital role in environmental management, helping identify the presence, concentrations, and ecological risks of contaminants. These indices guide targeted management efforts, prioritize areas that need attention, and inform risk-based management strategies. Pollution indices contribute to ongoing monitoring and adaptive environmental management, forming the foundation for mitigation measures and a dynamic understanding of changing environmental conditions. Moreover, we must consider that HMs are often persistent in the environment and can remain in sediments for extended periods. This persistence means that even if the sources of pollution are reduced or eliminated, the effects of HM contamination can persist for a long time and continue to impact the ecosystem. Therefore, it is essential to consider these potential ecological impacts when evaluating heavy metal pollution in the Nador lagoon. Management and mitigation strategies should aim to reduce pollutant input, monitor water and sediment quality, and implement measures to protect and restore the affected ecosystem. Therefore, the results can be applied to mitigate the pollution load of the HMs, augment water management efficiency, and assist in the protection of water resources, also by the implementation of coastal wetland restoration projects, the practice of routine ecological monitoring, and the promulgation of local wetland conservation statutes and specific regulations.

Ultimately, the study sheds light on the extent and sources of heavy metal contamination and pollution in the Nador lagoon, highlighting the urgency for proactive coastal management measures. The findings underscore the importance of integrating pollution control

strategies, such as improved wastewater treatment and industrial regulations, into coastal management plans to mitigate the adverse impacts of heavy metals on ecosystem health and human well-being. By incorporating our research into coastal management frameworks, policymakers and stakeholders can work collaboratively to safeguard the ecological integrity and resilience of coastal lagoon ecosystems for current and future generations.

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## Appendix A

Table A1. Geochemical data [7].

Stations	ID	Ba	Ce	Co	Cr	Cu	Ga	Hf	La	Nb	Nd	Ni	Pb	Rb	S	Sc	Sr	Th	V	Y	Zn	Zr
N1	1	591.5	0.6	20.3	51.3	42.5	14.2	4.1	71.2	16.8	29.7	22.3	34.3	103.5	10,499.6	17.7	589.6	18.3	128.9	9.9	105.3	137.5
N2	2	522.5	0.6	17.6	41.1	46.1	12.5	3.2	65.7	15.0	30.0	18.9	55.9	90.1	9681.5	15.8	812.3	15.0	113.8	7.8	125.6	124.9
N3	3	246.9	74.0	6.9	74.1	43.0	5.4	0.9	11.8	3.9	8.4	7.3	38.5	43.2	19,177.5	7.2	424.3	10.3	86.4	1.9	89.8	50.7
N4	4	330.7	0.0	19.9	44.8	73.6	11.0	2.2	65.9	14.4	27.5	24.1	43.9	68.8	13,030.7	14.8	854.0	11.6	122.9	7.2	134.4	106.2
N5	6	443.4	0.0	22.4	59.8	66.4	16.4	3.8	74.0	22.3	33.6	34.0	55.9	104.9	14,246.8	18.5	573.8	17.4	134.1	8.8	127.0	137.1
N6	7	85.6	43.4	3.4	1.6	4.1	0.0	0.0	0.0	0.6	2.2	0.0	9.7	10.8	2589.4	1.0	606.7	2.9	16.0	0.2	1.4	25.1
N7	8	98.1	0.0	4.9	3.4	3.6	0.0	0.0	36.9	2.7	9.4	0.0	0.0	8.5	1985.8	0.0	617.9	2.4	20.0	0.0	1.5	33.1
N8	9	533.7	0.0	30.8	47.8	91.2	9.6	3.0	72.5	10.9	25.6	12.1	72.8	60.0	7638.7	13.0	633.8	9.7	164.9	4.1	325.9	118.7
N9	11	316.5	9.0	19.4	56.8	60.1	14.7	1.8	38.5	15.3	26.9	29.9	62.1	82.4	5687.6	12.5	765.6	9.7	117.7	10.4	125.9	109.9
N10	13	505.6	50.2	30.7	152.1	27.4	15.8	4.9	51.5	34.5	33.8	21.0	24.2	106.3	3495.6	16.6	570.6	19.6	348.3	10.5	66.5	225.5
N11	20	312.8	15.8	22.9	50.1	56.7	13.7	1.3	32.1	14.5	26.0	28.9	57.1	74.6	5744.6	14.4	734.0	7.8	109.4	10.7	112.5	110.5
N12	21	144.5	0.0	8.0	21.6	15.8	3.1	0.1	11.0	4.7	14.5	8.0	23.0	23.3	3332.3	7.5	523.1	2.9	48.7	2.4	35.9	57.4
N13	25	318.0	31.8	19.7	59.7	54.2	15.5	1.9	36.3	12.0	27.0	27.4	59.5	83.4	7793.7	14.6	636.3	9.2	127.1	9.7	135.0	91.4
N14	34	182.4	0.0	10.6	26.0	22.5	6.8	0.1	24.6	8.6	14.8	12.5	35.4	37.4	3221.1	6.1	662.6	3.9	65.9	6.6	53.9	65.5
N15	35	138.6	0.0	10.3	18.9	15.5	4.3	0.0	17.1	4.9	12.9	7.7	25.8	25.4	2583.1	3.5	589.8	2.9	52.2	2.8	37.8	46.7
N16	37	270.4	6.1	20.3	58.7	47.0	15.5	1.4	33.3	10.2	26.7	31.3	49.3	72.8	4906.9	14.4	823.1	8.4	129.4	9.5	113.5	91.7
N17	45	81.1	48.8	0.1	4.6	0.6	4.7	0.5	2.5	0.4	3.7	33.7	3.1	2.4	2337.6	1.0	0.4	2.5	20.6	4.8	17.8	3.5
N18	50	122.0	0.0	8.1	16.1	7.5	2.6	0.0	13.0	4.3	11.0	2.4	20.5	14.0	1688.6	5.5	1002.7	1.6	42.3	2.8	19.8	60.5
N19	59	823.3	60.1	27.8	37.2	60.5	14.6	3.8	39.5	21.3	34.6	15.9	64.9	145.8	5476.8	9.5	480.9	17.1	139.3	12.3	168.1	222.2
N20	64	316.2	0.0	23.2	58.9	57.4	14.4	2.4	67.6	12.6	31.2	31.3	41.7	85.2	6413.7	18.1	918.2	13.4	135.7	9.7	128.1	110.5
N21	83	546.4	81.3	0.0	23.4	26.4	0.0	0.0	17.5	0.4	15.5	0.0	2.7	0.0	2185.3	4.1	0.0	5.5	77.9	0.0	0.0	0.0
N22	92	146.7	0.0	10.2	16.2	11.7	2.3	0.0	47.4	5.0	11.6	5.5	10.6	16.9	1590.1	4.3	1082.9	3.4	56.4	1.9	29.6	61.1
N23	94	120.3	0.0	9.4	16.0	13.0	0.5	0.0	47.7	4.8	10.6	9.2	10.3	16.9	2056.7	0.0	1420.4	3.4	53.3	2.1	27.6	49.4
N24	96	273.2	4.1	19.4	60.9	38.3	15.0	2.0	27.9	11.3	23.2	51.7	52.5	75.6	2930.0	16.1	646.5	8.8	128.6	11.0	103.3	100.6
N25	99	266.6	8.7	19.4	56.6	43.5	14.2	1.5	21.5	10.6	22.4	29.0	47.3	66.6	4188.3	15.7	1075.0	8.3	123.3	8.6	104.5	91.3
N26	109	313.1	13.5	20.0	49.2	40.3	12.9	1.2	31.3	11.1	22.9	23.3	43.4	63.0	3983.6	13.6	1066.9	8.2	109.5	8.3	93.7	106.9
N27	110	314.7	0.0	18.3	49.7	40.2	11.3	1.7	63.3	10.2	26.1	25.0	28.0	62.4	5056.3	10.7	1009.4	11.0	111.1	7.5	96.0	109.3
N28	111	352.4	0.0	15.5	24.0	13.9	5.3	0.6	56.2	10.8	19.8	9.4	12.0	41.2	1412.7	6.3	1106.7	8.5	71.2	7.4	46.4	188.5
N29	120	246.8	0.0	18.5	46.1	36.4	11.8	1.1	23.0	10.9	23.8	23.7	46.6	59.2	4521.5	13.7	991.1	6.3	105.1	8.4	83.4	94.4
N30	129	296.1	0.0	15.6	53.6	28.8	11.5	2.2	60.1	11.8	24.5	57.3	21.1	65.5	1920.6	11.0	593.6	11.4	100.9	11.4	76.5	168.2
N31	143	278.9	16.1	22.0	64.8	41.8	14.8	2.0	27.1	12.3	25.5	35.5	51.5	81.0	2786.6	14.4	714.4	8.5	124.9	11.7	108.7	107.5
N32	145	292.1	0.0	21.4	67.7	39.9	14.3	2.5	68.1	10.8	26.3	39.7	30.6	76.9	2497.4	14.8	517.9	12.8	125.9	8.8	107.8	95.3
N33	147	284.8	0.0	16.6	60.1	30.7	13.0	1.8	63.8	11.0	32.4	33.1	21.4	68.1	1736.7	12.8	379.3	12.2	114.1	10.8	89.9	123.8
N34	151	222.6	0.0	9.7	19.3	9.7	3.4	2.1	53.8	7.1	16.4	10.0	2.4	26.3	337.5	2.8	311.5	6.5	43.7	4.2	28.7	148.0
N35	157	195.4	47.5	0.0	19.1	6.8	0.0	0.0	1.5	0.3	5.9	0.0	2.5	0.0	534.5	4.2	0.0	3.6	34.6	0.0	0.0	0.0
N36	164	270.0	0.0	18.3	59.3	31.7	11.9	2.0	62.8	12.1	27.4	124.0	18.4	65.4	2935.6	13.3	297.6	12.3	107.1	11.1	81.3	150.2
N37	173	343.5	0.0	23.1	43.3	42.2	6.9	1.2	64.8	9.0	23.3	23.7	18.4	47.2	6402.8	11.5	898.7	10.1	95.9	6.3	80.1	130.7

Table A1. Cont.

Stations	ID	Ba	Ce	Co	Cr	Cu	Ga	Hf	La	Nb	Nd	Ni	Pb	Rb	S	Sc	Sr	Th	V	Y	Zn	Zr
N38	180	260.6	9.4	19.4	58.4	36.8	14.4	1.4	30.6	11.2	24.7	29.1	41.4	67.5	3540.2	12.1	873.7	8.6	121.6	8.9	92.8	90.7
N39	186	255.9	0.0	20.6	33.1	22.4	6.1	1.2	56.8	10.3	18.6	23.1	14.3	39.9	3704.8	7.7	847.8	8.1	77.9	8.0	52.4	145.5
N40	192	265.2	18.2	19.5	68.0	40.4	16.6	1.5	30.5	11.4	27.2	41.5	51.5	76.6	3782.2	17.1	628.4	8.0	126.5	10.5	103.9	95.8
N41	221	306.5	0.0	19.6	66.2	39.2	14.2	2.8	63.4	13.5	25.3	39.8	32.7	84.1	3884.3	16.5	467.0	13.1	125.3	11.8	104.3	122.8
N42	227	285.5	0.0	21.6	71.2	38.7	14.6	1.9	68.3	12.4	31.4	39.0	33.7	80.6	2518.2	17.2	535.4	13.4	130.0	10.3	101.9	103.6
N43	230	237.0	0.0	21.7	49.1	45.4	9.2	0.8	60.4	11.6	21.4	48.3	25.5	57.6	6544.7	11.9	1202.6	9.7	112.9	8.0	84.0	103.6
N44	231	110.7	0.0	10.8	10.9	11.1	0.0	0.0	44.6	3.7	12.0	5.6	3.7	14.0	3123.9	0.0	1644.4	3.1	41.2	0.7	17.7	46.8
N45	242	57.2	40.3	0.0	0.0	3.8	0.0	0.2	1.6	0.4	0.8	0.0	2.9	0.0	2108.8	0.0	0.0	1.6	15.1	0.0	0.0	0.0
N46	244	79.1	0.0	4.5	4.4	2.8	0.5	0.0	9.4	2.8	8.9	0.0	14.6	7.6	1746.1	3.1	806.1	1.2	22.0	0.5	1.3	52.1
N47	246	257.1	4.3	19.3	59.3	36.1	14.5	1.4	22.7	12.6	26.8	34.5	45.2	70.4	3314.3	14.8	1035.9	7.1	122.6	11.7	91.4	107.6
N48	248	462.5	369.8	0.0	745.3	61.2	6.1	0.1	7.6	0.2	2.2	249.0	2.2	3.8	17,111.2	7.3	0.3	31.5	322.6	10.3	34.7	0.5
N49	249	66.6	0.0	6.2	8.1	1.8	0.0	0.0	4.5	2.1	3.4	0.0	7.8	4.7	770.8	0.0	698.4	1.2	19.0	0.0	0.3	50.3
N50	250	68.7	60.2	0.0	13.9	10.0	0.0	0.3	5.5	0.4	1.5	0.0	3.1	2.6	6477.0	0.3	0.0	3.3	30.0	0.0	0.0	0.0

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