Seasonal Trace-Metal Contamination in the Coastal Waters of Al Hoceima Bay (Mediterranean, Morocco)

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Abstract. Mediterranean coastal waters are highly exposed to dissolved trace metals from urban, port and riverine inputs. This study analyses the seasonal variability of six dissolved metals (Pb, Cd, Ni, As, Cr, Zn) in Al Hoceima Bay (Morocco) based on four sampling campaigns (2022–2023). Data were processed in Python, and non-parametric tests (Kruskal–Wallis, Mann–Whitney) were used to assess seasonal differences. Concentrations were compared with USEPA chronic saltwater criteria and the European AA-EQS standards. Arsenic and nickel showed recurrent exceedances during summer and autumn, while only nickel exceeded the EU AA-EQS when evaluated on an annual basis. Lead and cadmium remained below the quantification limit. These results highlight seasonally-driven contamination and the need for enhanced monitoring during high-risk periods, together with complementary analyses in sediments and biota.

Keywords: trace metals, Mediterranean Sea, coastal pollution, environmental standards, seasonal variability.

1 Introduction

The coastal areas of the Mediterranean Sea are subject to increasing environmental pressures linked to coastal urbanization, the discharge of domestic and industrial wastewater, port activities, and agricultural runoff enriched with contaminants[1–3]. Among the most persistent pollutants in these environments, trace metals (TM) play a central role due to their toxicity, chemical stability, and ability to bioaccumulate in marine organisms[4,5]. Even at low concentrations, certain metals such as lead (Pb), cadmium (Cd), nickel (Ni), and arsenic (As) can have significant ecotoxicological effects, affecting both biodiversity and human health via the food chain[4,6–9]. The Mediterranean, considered a semi-enclosed sea with low water turnover, is particularly vulnerable to this type of contamination[10,11]. Several studies have already shown the existence of metal pollution gradients linked to urban areas, coastal estuaries, fishing ports, and untreated discharges[12,13]. However, the majority of available studies favor global approaches based on extensive analytical batteries or synthetic water quality indices, which makes specific and seasonal analysis of regulatory priority metals less visible[14,15].

In this context, this study proposes a targeted assessment of six priority metals—Pb, Cd, Ni, As, Cr, and Zn—selected on the basis of their toxicity, frequency of detection in coastal waters, and status in international water quality assessment systems. The concentrations of these metals were monitored over a one-year period, covering four sampling seasons (2022–2023) in a bay representative of the Moroccan Mediterranean coastline. The values obtained are analyzed along two complementary axes: (i) seasonal and annual variability of concentrations in coastal environments, (ii) verification of regulatory compliance with USEPA chronic criteria in saltwater [16] and European Union marine environmental quality standards (EQS)[17]. In addition to concentration monitoring, regulatory interpretation differs between frameworks: the USEPA establishes chronic criteria applicable at seasonal resolution, whereas the European Water Framework Directive (WFD) defines annual-average EQS (AA-EQS) as the basis for evaluating chemical status. This distinction is particularly relevant in dynamic coastal systems where seasonal peaks may not be reflected in annual means.

The study aims to identify metals that pose a potential risk of exceeding health and environmental thresholds, and to contribute to the implementation of priority monitoring and integrated management strategies for Mediterranean coastal areas.

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2 Materials and Methods

2.1. Study area

The Bay of Al Hoceima, located along the northern Mediterranean coast of Morocco, stretches from Ras Terf (Cape Quilates) in the east to Rass El Abed (Cape Maure) in the west [18,19].It encompasses key fishing grounds as well as coastal habitats of high ecological value. This area is representative of the hydrological conditions and environmental pressures characteristic of semi-enclosed Mediterranean coastal ecosystems[9].

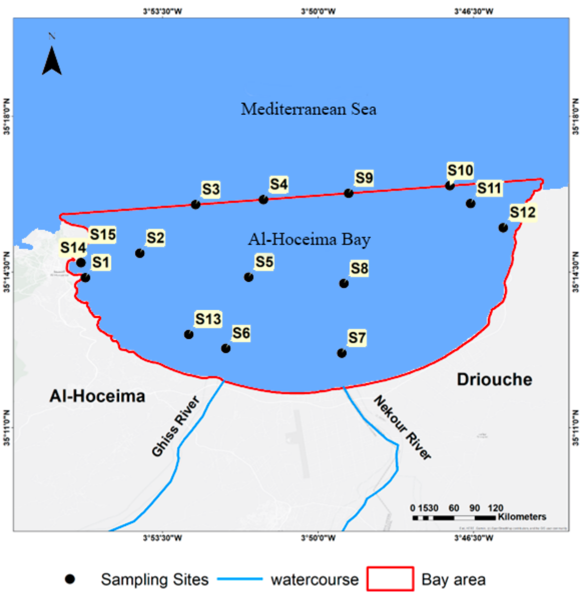

Fig.1. Geographical location of Al Hoceima Bay and sampling network (2022–2023).

2.2 Sampling and preservation

Four sampling campaigns were carried out during 2022–2023, corresponding to the fall, winter, spring, and summer seasons. Surface water samples –50 cm were collected at several stations using pre-acidified polyethylene bottles[20,21]. The samples were stored at 4°C, filtered through a 0.45 µm membrane, then acidified with suprapure HNO_3 (pH < 2) before analysis[22–24]. Fifteen sampling stations were strategically distributed across the bay to provide representative coverage of port areas, urban shorelines, and river-influenced zones (Fig.1).

2.3 Analysis of trace metals

Dissolved trace metal concentrations were quantified by ICP-MS (Inductively Coupled Plasma Mass Spectrometry) [25–27].The study focuses on six priority metals subject to international regulation: lead (Pb), cadmium (Cd), nickel (Ni), arsenic (As), chromium (Cr), and zinc (Zn).

2.4 Statistical treatment

Seasonal mean concentrations were calculated by averaging all sampling stations within each campaign (autumn, winter, spring, summer), and an annual mean was subsequently derived from the four seasonal means. All concentrations are expressed in $\mu g \cdot L^{-1}$. Measurements reported as "<LOQ" were converted to missing values and excluded from numerical calculations. Data handling and statistical processing were performed in Python (v. 3.10) using the Pandas and NumPy libraries[28,29].
Seasonal variability among the four campaigns was evaluated using a non-parametric Kruskal–Wallis test, followed by Mann–Whitney pairwise comparisons with Holm correction for multiple testing (SciPy, Statsmodels) [1,30]. For each metal, individual station values are represented in the figures as scatter points, while the seasonal mean is shown as a connected trend line. This graphical representation reflects the descriptive statistics reported in Table 1 (seasonal means) and Table 2 (annual means). Dissolved arsenic, nickel, chromium and zinc are illustrated in Figs. 2–5.

2.5 Regulatory benchmarks

The measured concentrations were evaluated against two regulatory frameworks: the USEPA Saltwater Chronic Criteria (CCC)[31,32], which define threshold values for dissolved metals in marine waters, and the European Environmental Quality Standards (AA-EQS marine), available for Ni, Pb and Cd. Compliance or exceedance of these limits was examined for each metal and for each season[32–35].

3 Results

3.1 Seasonal patterns of dissolved metals

Seasonal mean concentrations are reported in Table 1. Among quantified elements, arsenic (As) and nickel (Ni) exhibited the highest concentrations and marked seasonal contrasts. As ranged from 25.04 $\mu g \cdot L^{-1}$ in spring to 49.55 $\mu g \cdot L^{-1}$ in autumn, with 41.44 $\mu g \cdot L^{-1}$ in summer and 28.20 $\mu g \cdot L^{-1}$ in winter. Ni varied between 6.59 $\mu g \cdot L^{-1}$ in winter and 11.98 $\mu g \cdot L^{-1}$ in summer, with intermediate means of 10.86 $\mu g \cdot L^{-1}$ in autumn and 7.76 $\mu g \cdot L^{-1}$ in spring.

Table 1. Seasonal mean concentrations ($\mu g \cdot L^{-1}$)

Metal	Autumn	Winter		Spring	Summer
Pb	LOQ	LOQ		LOQ	LOQ
Cd	LOQ	LOQ		LOQ	LOQ
Ni	10.86	6.59		7.76	11.98
As	49.55	28.20		25.04	41.44
Cr	19.80	14.74		0.69	0.96
Zn	7.07	6.23		11.44	14.31

Chromium (Cr) and zinc (Zn) showed moderate to low concentrations overall, with maxima of 19.80 $\mu g \cdot L^{-1}$ for Cr in autumn and 14.31 $\mu g \cdot L^{-1}$ for Zn in Summer. Lead (Pb) and cadmium (Cd) remained below the quantification limit in all seasons and are therefore not

further detailed graphically. Station-level dispersion and seasonal trends are displayed in Figs. 2–5.
Statistical testing confirmed that seasonal differences were significant for all dissolved metals detected above the quantification limit (Ni, As, Cr and Zn; Kruskal–Wallis, $p < 0.001$). Post-hoc Mann–Whitney tests (Holm-adjusted) indicated that the strongest contrasts occurred between the warm seasons (summer–autumn) and the cooler seasons (winter–spring), while some pairs such as winter vs spring showed no significant difference. Lead and cadmium were not analysed statistically because all measurements were below the limit of quantification.

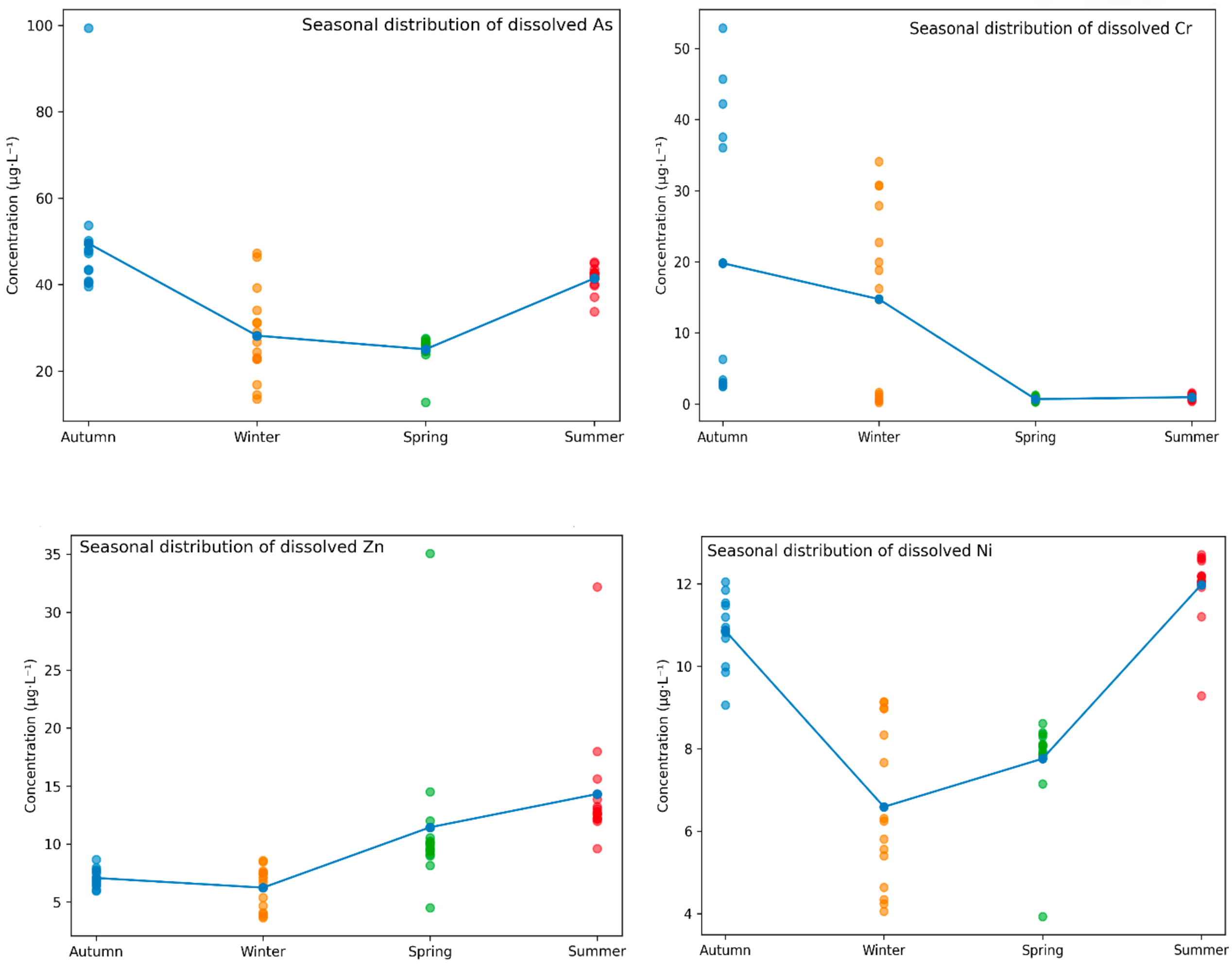

Fig. 2. Seasonal distribution of dissolved trace metals (As, Ni, Cr, Zn) in Al Hoceima Bay.

3.2 Annual dissolved metal concentrations Annual mean

concentrations (average of the four seasonal means) confirm the predominance of As and Ni in the dissolved phase of the bay (Table 2). As reached 36.06 µg·L^{-1} and Ni 9.30 µg·L^{-1}, while Cr and Zn were 9.05 µg·L^{-1} et 9.76 µg·L^{-1}, respectively. Pb and Cd remained below the quantification limit throughout the year.

Table 2. Annual mean concentrations (µg·L^{-1})

Metal	Pb	Cd	Ni	As	Cr	Zn
Annual	—	—	9.30	36.06	9.05	9.76

3.3 Regulatory compliance

The assessment of regulatory compliance was revised to align with the European Environmental Quality Standards (EU AA-EQS), which are defined as annual average concentrations under Directive 2013/39/EU[17]. Therefore, only the annual means (Table 2) were used for comparison with EU standards, while seasonal values were compared exclusively with the USEPA chronic saltwater criteria.

Pb and Cd remained below the analytical limit of quantification throughout the year and could not be evaluated against USEPA or EU benchmarks. For nickel (Ni), the annual mean concentration (9.30 µg·L^{-1}) slightly exceeded both the USEPA chronic criterion (8.2 µg·L⁻¹) and the EU AA-EQS value for coastal and transitional waters (8.6 µg·L⁻¹). Arsenic (As), which is not regulated under the EU EQS list, exceeded the USEPA criterion (36 µg·L⁻¹) based on both seasonal maxima and the annual average (36.06 µg·L⁻¹). Chromium (Cr) and zinc (Zn) remained well below their respective USEPA limits across all seasons and in annual means. Accordingly, only nickel shows non-compliance with the EU AA-EQS, while arsenic and nickel both exceed USEPA criteria. No exceedances were observed for Cr or Zn. These results confirm that the regulatory concern is driven by metals with the lowest permissible thresholds (As and Ni), whereas other elements remain within acceptable ranges.

Table 3. Regulatory thresholds and compliance assessment (corrected according to EU AA-EQS requirements)

Metal	Regulatory Threshold	Framework	Basis of assessment	Measured (Annual or Seasonal)	Compliance
Pb	8.1 µg·L⁻¹	USEPA CCC	—	<LOQ	Not evaluated
Pb (EU)	1.2 µg·L⁻¹ (AA-EQS, annual)	EU WFD	Annual	<LOQ	Compliant
Cd	8.8 µg·L⁻¹	USEPA CCC	—	<LOQ	Not evaluated
Cd (EU)	0.2–0.45 µg·L⁻¹* (AA-EQS)	EU WFD	Annual	<LOQ	Compliant
Ni	8.2 µg·L⁻¹	USEPA CCC	Seasonal & annual	Annual = 9.30	Exceeded
Ni (EU)	8.6 µg·L⁻¹ (AA-EQS, annual)	EU WFD	Annual	9.30	Exceeded
As	36 µg·L⁻¹	USEPA CCC	Seasonal & annual	Annual = 36.06	Exceeded
Cr	50 µg·L⁻¹	USEPA CCC	Seasonal	Annual = 9.05	Compliant
Zn	90 µg·L⁻¹	USEPA CCC	Seasonal	Annual = 9.76	Compliant

4 Discussion

The seasonal pattern observed in dissolved metal concentrations reflects the combined influence of hydrodynamics, temperature, and coastal inputs. The highest levels of As, Ni, Cr and Zn were recorded during summer and autumn, periods typically characterized by reduced water renewal, thermal stratification and intensified port and urban activity, all of which favour the build-up of dissolved contaminants. Conversely, winter showed the lowest concentrations, driven by strong hydrodynamic mixing, rainfall-induced dilution and shorter water residence time. These patterns were statistically confirmed, with significant differences among seasons for all quantified metals (Kruskal–Wallis, $p < 0.001$), particularly between the warm (summer–autumn) and cold (winter–spring) periods. Pb and Cd remained below the limit of quantification throughout the year, likely reflecting their stronger association with particulate matter and sediments rather than the dissolved phase.

Although Cr and Zn exhibited seasonal fluctuations similar to those of As and Ni, their concentrations consistently remained well below the corresponding USEPA saltwater chronic criteria. This contrast does not imply a weaker environmental response but results from differences in toxicological thresholds and geochemical behaviour. As and Ni have substantially lower permissible concentrations (36 µg·L⁻¹ and 8.2 µg·L⁻¹, respectively), reflecting their higher chronic toxicity and bioaccumulation potential, whereas Zn and Cr are regulated at much higher limits (90 and 50 µg·L⁻¹). Consequently, identical environmental processes can elevate multiple metals simultaneously, yet only those with lower regulatory thresholds (As, Ni) reach concentrations that exceed guideline values.

From a regulatory perspective, the corrected compliance assessment highlights an important distinction between seasonal and annual interpretations. USEPA chronic criteria, which can be applied at seasonal resolution, indicate exceedances for both As and Ni during summer and autumn. In contrast, the European AA-EQS—legally defined as annual averages under the Water Framework Directive—identify non-compliance only for Ni when considering annual mean values, while As is not regulated within the EU EQS list. This divergence shows that annual averages may mask short-term concentration peaks that are ecologically relevant, especially in semi-enclosed Mediterranean bays where hydrodynamics can produce transient yet intense exposure events.

The fact that exceedances are seasonal rather than permanent has direct implications for monitoring and risk assessment. Reliance solely on annual means may underestimate exposure risk and lead to a classification of chemical status that does not reflect episodic contamination events. This underscores the need for monitoring strategies that incorporate temporal dynamics, in line with the requirements and guiding principles of both the Water Framework Directive (WFD) and the Marine Strategy Framework Directive (MSFD) [36,37]. In particular, the MSFD emphasises the evaluation of long-term contaminant trends in water, sediment and biota, as well as the avoidance of significant increases over time.

Future work should include the analysis of particulate and sediment fractions, trace metal speciation, and higher-frequency time series to better identify sources, assess bioavailability and refine ecological risk evaluations. Integrating these complementary approaches would help move beyond descriptive assessments toward a more comprehensive understanding of metal dynamics in Mediterranean coastal ecosystems.

5 Conclusion

This study demonstrated a clear seasonal variability in dissolved trace metals in the coastal waters of Al Hoceima Bay, with the highest concentrations occurring during summer and autumn. While several metals showed seasonal increases, only arsenic and nickel exceeded international regulatory thresholds. The comparison with the USEPA chronic saltwater criteria revealed seasonal exceedances for both metals, whereas the evaluation using the EU AA-EQS—legally defined as annual averages—indicated non-compliance only for nickel. This distinction underscores the limitations of relying solely on annual means, which may mask short-term peaks that are ecologically significant in semi-enclosed Mediterranean environments.

The results highlight the importance of integrating temporal variability into monitoring and risk-assessment frameworks, in line with the principles of both the Water Framework Directive (WFD) and the Marine Strategy Framework Directive (MSFD). Enhanced monitoring approaches are needed to better capture exposure dynamics, particularly during warm periods when hydrodynamic conditions favour contaminant accumulation. Future investigations should incorporate particulate and sediment fractions, metal speciation and higher-frequency sampling to more accurately identify sources, assess bioavailability and evaluate potential impacts on marine organisms and ecosystem health.

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