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Article

A Framework to Assess Natural Chloride Background in Coastal Aquifers Affected by Seawater Intrusion in Eastern Spain

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Abstract: The protection of groundwater resources in coastal aquifers is an increasingly important issue worldwide. To establish threshold values and remediation objectives, it is essential to know the natural background concentrations of relevant ions in groundwater. The rationale is to define the Natural Background Level (NBL) of chemical species determined by atmospheric and lithological forces. In many coastal aquifers, this evaluation worsens since atmospheric and lithological salinity combines with many other anthropogenic sources of salinity, including exogenous salinity induced by seawater intrusion (SWI). This paper presents a combination of six well-known statistical techniques and a new methodology (i.e., SITE index) in eight GWBs affected by SWI in Eastern Spain. The chloride ion was the selected conservative chemical specie to assess the qualitative status due to the variable SWI affection. The Natural Chloride Background (NCB) obtained from these methodologies at the GWB scale was compared with regional NCB data calculated with the Atmospheric Chloride Mass Balance (CMB) method in Continental Spain. The CMB method provides atmospherically derived NCB data that are not influenced by SWI or anthropogenic activities or lithological forces. This external evaluation can be considered the atmospheric fraction of NCB, which serves as a regional criterion to validate the more detailed statistical methodologies applied at the GWB scale. As a result, a conceptualization of NCB is obtained by means of a range of values between 115 mg L^{-1} and 261 mg L^{-1} in the studied coastal GWBs affected by SWI in Eastern Spain.

Keywords: groundwater; chloride; natural background; coastal aquifers; seawater intrusion; eastern Spain

1. Introduction

Groundwater protection constitutes the cornerstone of the principles of the sustainable management of groundwater resources. In the "United Nations World Water Development Report 2022: Groundwater: Making the Invisible Visible", it is emphasized that coastal aquifers provide a critical source of fresh water for people in many regions around the world [1]. Moreover, the report states that the impact of sea level rise is relatively of no great concern compared to the intensive exploitation of groundwater wells for supply and agriculture in those areas.

In this situation, the identification of natural background concentrations of chemical species in groundwater is the first step to assess the anthropogenic impacts and establish threshold values and cleanup goals [2].

In Europe, the European Water (EWD) and Groundwater (EGWD) Framework Directives [3,4] establish the regulation for prevention and control of pollution, and this includes provisions for assessing the natural occurrence threshold of chemical species in groundwater. For almost two decades, even before the publication of the EWD [3], an attempt has been made by the EU state members to establish criteria aimed at defining the natural quality of groundwater and the development of an evaluation methodology at the European scale that is aligned with the requirements of the EWD. In addition to these requirements, the state members are encouraged to implement the third River Basin Management Plan (RBMP) at each River Basin District; the RBMP is valid for the period 2022–2027. The evaluation of the qualitative status of a GWB is based on the estimation of the Natural Background Level (NBL) of different chemical species. This estimate is the starting point to calculate threshold values leading to the official declaration of the qualitative status of a given GWB.

This official declaration upon assessment is especially difficult in densely populated coastal GWBs subjected to high groundwater-pumping rates causing SWI. In these areas, atmospheric and lithological salinity combines with many other anthropogenic sources of salinity, as well as the first-order exogenous salinity induced by SWI. Different investigations have combined diverse methods to deduce the NBL. Some of them [5,6] analyzed the spatial distribution of hydrochemical facies to define the qualitative status of GWBs during different periods, while others [7] combined geophysical and geochemical methods in areas with limited data availability. In addition, several statistical techniques have also been used to assess NBL in coastal aquifers affected by SWI; the choosing and subsequent confidence of the results depended on the spatial and temporal distribution of the available data. The simplest techniques were basic statistics such as range, average, minimum, and maximum values. Other more complex ones enabled researchers to remove the anthropogenic influence identified as positive outliers into normal or lognormal distributions.

While numerical modeling of groundwater flow and solute transport requires an extensive effort [8,9], most of the statistical models applied to date are based on the elimination of outliers that cause the distribution to deviate from the one that would occur without direct human influence (usually the normal distribution).

The procedure to calculate the NBL varies in each EU member state. For instance, the EU-funded BRIDGE Project [10] proposed the evaluation of groundwater samples free from anthropogenic influences to obtain the natural qualitative status of GWBs. This procedure has the disadvantage that samples from deep aquifers may overestimate the influence of lithological sources and underestimate atmospheric forces and anthropogenic influences. Likewise, the hydrochemical modeling of water–rock interaction requires a great effort in regard to time and resources and is applicable in areas with huge data. In the case of Spain, the Ministry for the Ecological Transition and Demographic Challenge has proposed the BRIDGE's methodology despite the fact that the original conceptualization of samples can be misleading. When the BRIDGE methodology is applied to Mediterranean coastal aquifers affected by SWI, most (if not all) values exceed the requirements to be included in the calculations. This is the reason why preselection methods such as the BRIDGE methodology must be avoidable, and alternative methods are recommended instead. For instance, the EU-funded BaSeLine Project [11,12] focused on the different timescales that influence the NBL of chemical species in groundwater, as well as the speed at which these processes occur, among other topics. One of the aims of the EU-funded HOVER Project was to improve the rationale and limitations of NBLs and associated threshold values [13].

With regard to the concept of NBL, the absence of anthropogenic influences is far from occurring in most of the Mediterranean coastal aquifers, including the Spanish ones. The comparison of groundwater samples associated with a variable water–rock interaction (induced by different groundwater turnover times) also deteriorates the NBL definition and confidence of typical statistical techniques [14]. In a typical scenario of groundwater imbalance induced by high exploitation rates leading to SWI [15], it is not always possible to sample groundwater associated with pristine conditions, and indirect (often statistical) techniques that are especially addressed to evaluate the SWI affection are required. Another additional problem is the selection of a suitable chemical specie or tracer to identify SWI, whose level must usually be on the threshold of exceeding (or has already exceeded) the quality standards that the EWD [2] establishes. Chloride ion is an ideal conservative chemical specie that is widely used to assess groundwater quality degradation due to SWI [16,17]. For this reason, chloride was the chemical specie considered. Hereafter, NBL refers exclusively to the Natural Chloride Background (NCB).

This paper presents a combination of six well-known statistical techniques and a new methodology (SITE index) in eight coastal GWBs affected by SWI in Eastern Spain. The NCB obtained from these methodologies at the GWB scale is also compared with regional NCB data calculated with the Atmospheric Chloride Mass Balance (CMB) method in Continental Spain that provides atmospherically derived NCB data not influenced by SWI or anthropogenic activities and lithological forces. This external evaluation can be considered the atmospheric fraction of NCB, which serves as the regional criterion to validate the more detailed statistical methodologies applied at the GWB scale. Table 1 describes the acronyms used.

Table 1. List of acronyms used.

Acronym	Description
BaSeLiNe	Natural Baseline Quality in European Aquifers: a basis for aquifer management
BRIDGE	Background Criteria for the Identification of Groundwater Thresholds
CHJ	Júcar River Basin Authority
CMB	Atmospheric Chloride Mass Balance
EU	European Union
EGWD	European Groundwater Framework Directive
EWD	European Water Framework Directive
GWB	Groundwater Body
HOVER	Hydrological processes and geological settings over Europe controlling dissolved geogenic and anthropogenic elements in groundwater of relevance to human health and the status of dependent ecosystems
CN IGME–CSIC	Geological Survey of Spain
IQR	0.25–0.75 interquartile range
NAO	North Atlantic Oscillation
NCB	Natural Chloride Background
NBL	Natural Background Level
NCB	Natural Chloride Background

Table 1. *Cont.*

Acronym	Description
PC	Plana de Castellón GWB
PG	Plana de Gandía GWB
POT	Plana de Oropesa–Torreblanca GWB
PS	Plana de Sagunto GWB
PV	Plana de Vinaroz GWB
PVN	Plana de Valencia Norte GWB
PVS	Plana de Valencia Sur GWB
PX	Plana de Xeraco GWB
RBMPs	River Basin Management Plans
SWI	Seawater intrusion

2. Materials and Methods

2.1. Statistical Techniques

2.1.1. Overall Framework

Six statistical techniques that are well documented in the scientific literature were sequentially applied to the studied GWBs to deduce the NCB value unaffected by SWI, such as the calculation of averages, iterative 2-σ, iterative 2-σ after removing data series with increasing trend, distribution function, PT technique, and Walter approach.

2.1.2. Calculation of Averages

This technique calculates the annual median and 0.90 and 0.95 percentiles of the dataset. The technique is aimed at removing the influence of hyper-annual variations. The median of annual series is considered the representative NCB value.

2.1.3. Iterative 2-σ

This iterative technique calculates the median value and standard deviation (±2σ) of the normal distribution of the dataset. A range associated with each dataset is defined, from which outliers can be removed. The subsequent refined (corrected) normal distribution is built around the modal value. The Shapiro–Wilk test is applied to check the data-series normality.

2.1.4. Iterative 2-σ after Removing Data Series with Increasing Trend

This technique is a variant of the previous one that removes the data series with an increasing trend. The non-parametric Mann–Kendall statistical test [18] is used to deduce the existence of an increasing trend regarding the theoretical stationary invariant trend. This technique requires long-term monitoring records to identify trends.

2.1.5. Distribution Function

This technique calculates medians and removes those data located above those values. The median of the dataset is calculated, and the values located above are eliminated; with the remaining dataset, it adjusts towards values above the median and calculates the mean and the standard deviation [19]. The Lilliefors test [20] is used to evaluate the normality of the results. It is important that values in the minimum–median range be free of anthropogenic influences. This is a limiting factor in the same sense as it was with the BRIDGE methodology.

2.1.6. PT Technique

This technique calculates the interquartile 0.25–0.75, extracts the data within, and removes the data included in the quartiles 0–0.25 and 0.75–1 [21]. For this, the technique uses the quantile–quantile graph and/or a histogram to plot data and check the normal distribution. To check data normality, the Shapiro–Wilk test is applied once the result has been adjusted and the 0.95 percentile corresponding to the background level is calculated. This is the technique used by the Geological Survey of Portugal to define NBLs.

2.1.7. Walter Approach

This technique assesses the NCB by applying an iterative statistical process using probability plots [22]. It was developed by the Geological Surveys of the Federal States of Germany to implement the EWD. This method seeks to eliminate extreme, anomalous values affected by unknown processes that can modify NBLs, such as anthropogenic influences, mineralization, SWI, etc. This method considers an initial dataset of a mixed population of natural and affected data. This assumption is checked by performing the normal and lognormal probability plots' distribution graphics. The iterative process begins by removing samples above the 0.9 percentile until a normal distribution is reached. The mean, standard deviation, and percentiles of this normal distribution are calculated.

2.2. The SITE Index

SITE is a new index whose objective is to characterize the intrusion process in a specific groundwater body (GWB) through easily obtainable data. It is based on the analysis of the chloride ion concentration in different sections of an aquifer over a given study period. It produces numeric and alphanumeric results that allow for the characterization of the intrusion from spatial and temporal perspectives.

It is a parametric function coming from experimental relationships found in coastal aquifers in Eastern Spain to qualify the effect of SWI [23]. Its derivation comes from the following categorical variables:

$$\mathrm{SITE} = \frac{3S + 3\left(\frac{S}{4}\right)I + T + E}{30} \tag{1}$$

where S is the surface affected by SWI, I is the intensity, and T is the temporality of the process (permanent, seasonality, etc.); they use numeric codes between 0 and 4. Moreover, E is the evolution numerically codified between -2 and 2. The four parameters of SITE determine the qualitative incidence of the SWI through the chloride data measured at different points and times. In the numerator, 30 is the highest maximum possible. The utility of indices to summarize the dynamic of SWI [24–26] and assess potential management strategies [27] has been demonstrated in different applications.

2.3. Atmospheric Chloride Mass Balance Method

The Atmospheric Chloride Mass Balance (CMB) is one of the most widely used methods to estimate net aquifer recharge from precipitation in different orographic, climatic, and geological contexts [28–30]. The CMB is a global method based on the principle of mass conservation of a conservative tracer—in this case, the chloride ion—atmospherically contributing to the land surface. The CMB method was recently applied to estimate the distributed spatial mean net aquifer recharge and its natural uncertainty (standard deviation) over Continental Spain. For a confident application, the long-term steady condition of the CMB variables, namely atmospheric chloride bulk deposition, chloride export flux by surface runoff, and recharge water chloride content, was verified [31–33]. The long-term recharge water chloride content data series used as input data corresponds to NCB exclusively attributed to the atmospheric forces [33–35]. The NCB data evaluation examined the influence of hydraulic properties (mostly permeability and storability) of different aquifer lithologies, as well as the potential contribution of non-atmospheric sources of chloride. Ordinary kriging was used to regionalize the CMB variables at the same 4976 nodes of a 10 km × 10 km grid. In each grid node, a mean NCB value and two main types of uncertainty, the natural variability of the variable and the error from its mapping, were calculated [30,32].

The evaluation covered a 10-year period, which represented the critical balance period for the CMB variables to reach comparable steady means and standard deviations. This 10-year period matches the decadal global climatic cycles acting on the Iberian Peninsula, with irregular ~5-year positive and negative phases that follow the North Atlantic Oscillation trend [36,37]. Considering that (1) at least a 10-year balance period is required for reliably steady NCB evaluations in Continental Spain and (2) the NCB datasets preferably spanned the period 1994–2007, the control period (1996–2005), which spans a full 10-year-long NAO climatic cycle, was chosen as the representative period for NCB data in this work. Other authors have also implemented these CMB datasets for local evaluations of net aquifer recharge and NCB in different climatic and geological settings, such as in carbonate GWBs in Southern Spain [38,39]; in varied geological contexts on the northern coast of Spain [40]; in high-mountain, weathered-bedrock basins in Southern Spain; and in the headwater of carbonate-bedrock basins in Southern Spain [41]. It has also been used in the assessment of the national or EU scale of variables that depend on aquifer recharge [42–44].

3. Study Area

The coastal fringe of Eastern Continental Spain [45] includes a succession of coastal plains from Castellón (North) to Alicante (South) provinces that are catalogued as detrital sedimentary GWBs (Figure 1). These coastal plains share common features, such as a flat geomorphology, maximum elevation in the 100–300 m above sea level (a.s.l.) range, and geological and hydrogeological characteristics. Similar geology determines multilayer aquifers formed by high-permeability upper formations with transmissivity in the 500–1200 $m^2\ d^{-1}$ range, intermediate low-permeability formations confining the underlying ones, and bedrocks formed by a variety of lithologies depending on the GWB. Based on the available data and knowledge gained during last decades about their hydrogeological functioning, the selected coastal GWBs were Plana de Vinaroz (PV), Plana de Oropesa–Torreblanca (POT), Plana de Castellón (PC), Plana de Sagunto (PS), Plana de Valencia Norte (PVN), Plana de Valencia Sur (PVS), Plana de Xeraco (PX), and Plana de Gandía (PG) (Figure 1). Hereafter, these acronyms are exclusively used to refer to each GWB. Table 2 describes the main geographical and hydrogeological features of these GWBs.

Table 2. Characteristics of the studied GWBs. Name and location of each GWB as in Figure 1.

GWB	Geological Formation	Area, km^2	Transmissivity, m^2/day	Storage Coeff.
PV	Gravel, sandstone, conglomerates, clays, gravel, and sand levels	117	4000	0.001
POT		88.64	30–1000	0.2–0.12
PC	Conglomerates, gravels, and sands of the Plio-Quaternary	464	6000	0.05–0.15
PS		127		0.1–0.12
PVN	Quaternary sand and gravel in a silty–clayey formation	1300		0.1–0.01
PVS				
PG	Levels of gravel and sand embedded in a silty–clayey matrix	250	300–1500	0.07
PX		59.77	500–3000	0.05–0.15

Large-scale use of water for agriculture, rapid urbanization, population growth since 1970, and seasonal tourism during summer have notably increased groundwater pumping [46]. These anthropogenic activities have led to an increase in the average chloride (Cl) concentration since the end of the 1970s in most of the GWBs. Other problems, such as saltwater upconing and reversal of groundwater flow direction, have caused groundwater quality degradation in some sectors of these GWBs. During the current decade, seasonal cycles of groundwater quality oscillations have taken place. However, piezometric evolution in some GWBs showed low variations during the last years, thus corroborating a low SWI affection.

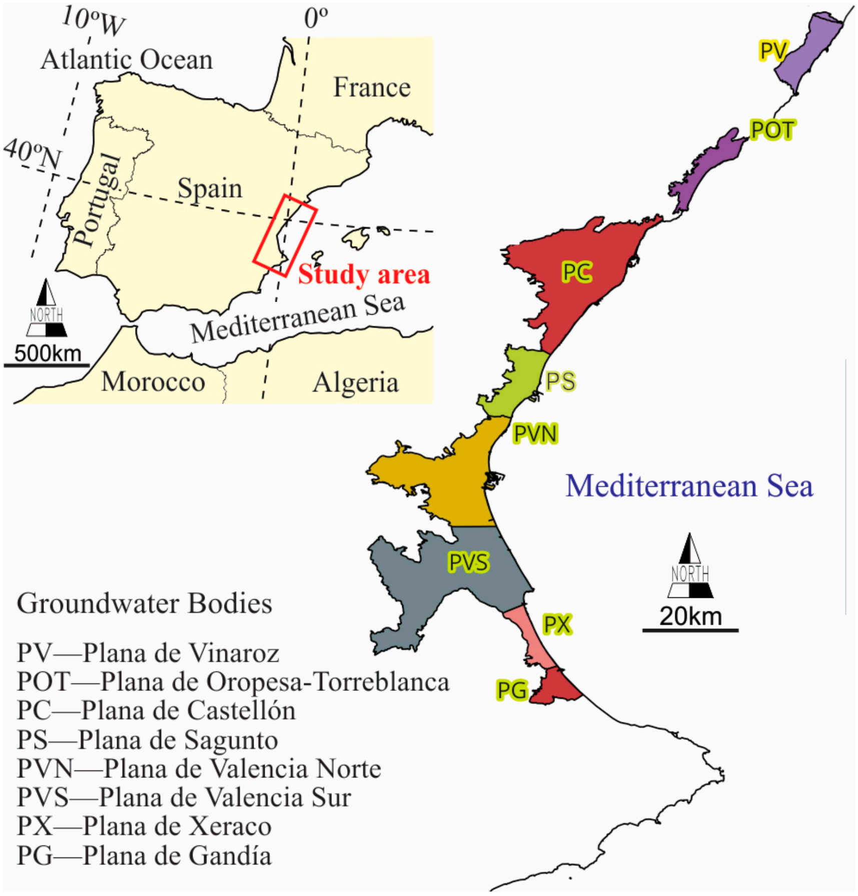

Figure 1. Location of the study area in Eastern Continental Spain.

4. Results

4.1. Piezometric and Chloride Distribution

The piezometry was evaluated since its depletion in the coastal fringe of the studied GWBs may warn about a possible SWI. The Cl data used came from the Geological Survey of Spain (CN IGME–CSIC) and Júcar River Basin Authority (CHJ) [47] public databases. The CN IGME–CSIC database was in operation from 1972 to 2001, while the CHJ one includes data to date. The Cl concentration fluctuates from 1 mg L^{-1} to more than 9000 mg L^{-1} at PV GWB (Table 3). The existence of outliers (probably associated with different sources of salinity different from the atmospheric and SWI ones) makes the average Cl values significantly higher than the median values in all GWBs.

Table 3. Summary of chloride concentration statistics in mg L^{-1}. IQR is the p75–p25 range.

GWB	Size, km^2	Range	Mean (M)	Median (m)	m/M	St. Dev.	p25	p75	IQR
PC	4313	1–9375	411	170	0.41	527	92	568	476
PG	84	35–213	113	114	1.01	36	85	142	57
POT	1138	27–6250	902	852	0.94	612	469	1193	724
PS	3287	7.3–7818	523	312	0.60	604	142	696	554
PV	3501	4–5183	420	213	0.51	527	78	525	447
PVN	590	35–497	187	170	0.91	88	121	241	120
PVS	294	14–727	175	142	0.81	112	114	194	80
PX	1086	14–6930	357	199	0.56	456	128	412	284

Therefore, medians and 0.25–0.75 interquartile ranges (IQRs) are more suitable statistics to describe the typical behavior and variability of the Cl data series, while the mean-to-median ratio (m/M) identifies skewed and biased effects; m/M = 1 identifies a normal data distribution. Table 2 shows a range between 0.4 and 1, with means typically higher than medians. The medians range from 114 mg L^{-1} at PG GWB to 852 mg L^{-1} at POT GWB. PG GWB dataset shows the smallest IQR values; however, the number of samples in this GWB is quite limited. In general, there is information enough to perform the analysis. Cl concentrations up to 1250 mg/L were found in the POT GWB (Figure 2).

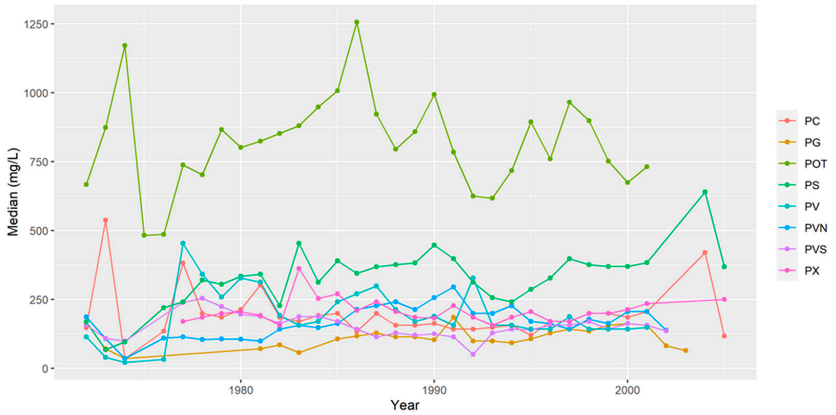

Figure 2. Temporal evolution of the median chloride concentration (mg L^{-1}) in the studied GWBs: PC—Plana de Castellón; PG—Plana de Gandía; POT—Plana de Oropesa–Torreblanca; PS—Plana de Sagunto; PV—Plana de Vinaroz; PVN—Plana de Valencia Norte; PVS—Plana de Valencia Sur; and PX—Plana de Xeraco.

A basic statistical analysis of the sample distribution by means of boxplots of Cl concentrations is shown in Figures 3 and 4. In Figure 3, the box width is proportional to the number of samples, while the notch indicates the median value. In Figure 4, the distribution frequency of the median Cl concentration in the sampling points for the selected pilots is also plotted. This information about the distribution of samples enables us to identify and discuss the reasons why the NCBs obtained with the different statistical techniques vary.

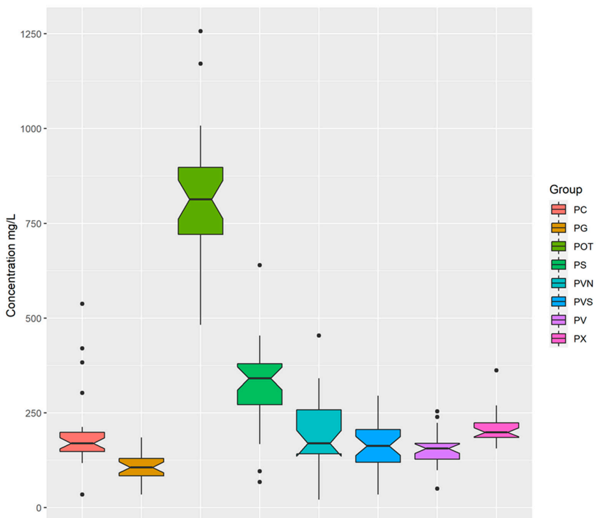

Figure 3. Box-whisker plots of the Cl concentration in mg L^{-1} at the selected GWBs: PC—Plana de Castellón; PG—Plana de Gandía; POT—Plana de Oropesa–Torreblanca; PS—Plana de Sagunto; PV—Plana de Vinaroz; PVN—Plana de Valencia Norte; PVS—Plana de Valencia Sur; and PX—Plana de Xeraco.

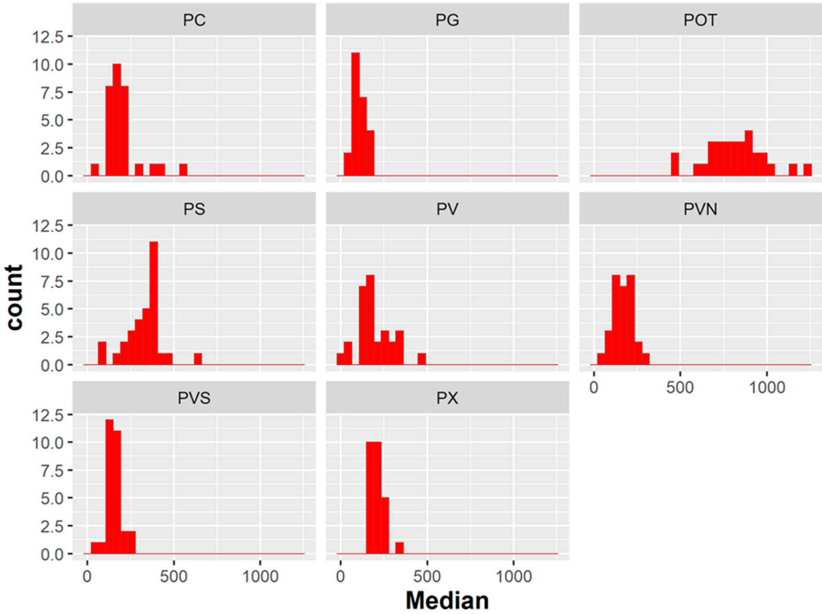

Figure 4. Frequency distribution of the annual median Cl concentration in mg L^{-1} in the selected GWBs: PC—Plana de Castellón; PG—Plana de Gandía; POT—Plana de Oropesa–Torreblanca; PS—Plana de Sagunto; PV—Plana de Vinaroz; PVN—Plana de Valencia Norte; PVS—Plana de Valencia Sur; and PX—Plana de Xeraco.

4.2. Statistical Techniques

For the Calculation of Medians technique, Figure 5 shows the evolution of NCB medians per data series (monitoring station) up to year 2006. A smoothing line in each data series is delineated. Table 4 includes the values obtained with each statistical technique.

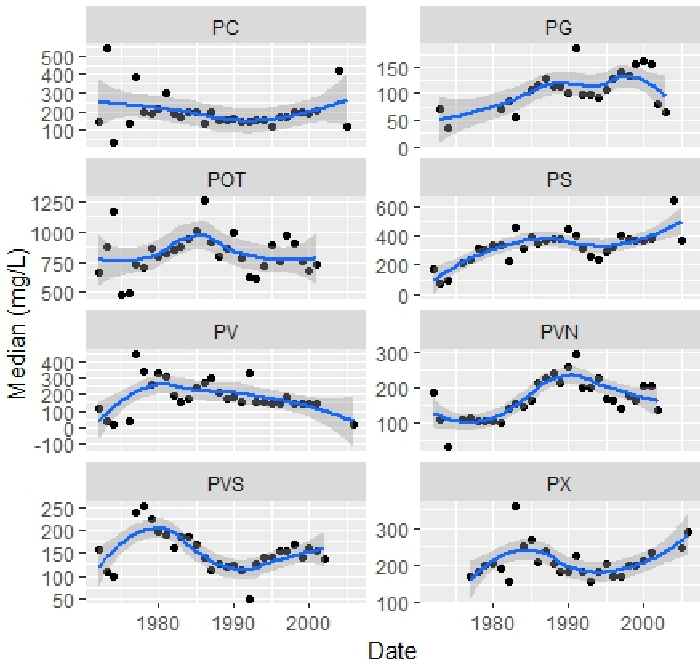

Figure 5. Evolution of median NCB values in mg L^{-1} vs. date in the selected eight GWBs: PC—Plana de Castellón; PG—Plana de Gandía; POT—Plana de Oropesa–Torreblanca; PS—Plana de Sagunto; PV—Plana de Vinaroz; PVN—Plana de Valencia Norte; PVS—Plana de Valencia Sur; and PX—Plana de Xeraco. Location of GWBs and monitoring stations are as shown in Figure 1.

Table 4. For the applied statistical techniques, reference NCB values (in mg L^{-1}) in the studied GWBs are PV—Plana de Vinaroz; POT—Plana de Oropesa–Torreblanca; PC—Plana de Castellón; PS—Plana de Sagunto; PVN—Plana de Valencia Norte; PVS—Plana de Valencia Sur; PX—Plana de Xeraco; and PG—Plana de Gandía.

Statistical Technique	PC	PG	POT	PS	PV	PVN	PVS	PX
Median	170	106	813	341	163	163	156	199
Iterative technique	92	114	767	142	71	156	128	135
Iterative technique with no trend	310	----	838	498	186	163	142	----
Distribution function	170	114	852	312	213	170	142	199
PT method	391	107	1099	480	306	234	180	344
Walter method	111	119	1399	99	92	258	195	242

For the Iterative 2-σ technique, after removing the data series with an increasing trend, in the PG GWB, the limiting factor was the absence of monitoring stations with observed increasing trends.

4.3. SITE Index

According to the definition provided in Section 2.2, we applied the methodology to the study area. The ranges of the SITE index obtained are included in Table 5, modeled after [23].

Table 5. Categories of seawater intrusion according to SITE index.

SITE Value	Intrusion Status
<0.10	Null
0.11–0.25	Low
0.26–0.50	Moderate
0.51–0.75	High
0.76–1.00	Extreme

While in Ballesteros et al. [23], the SITE index in PC, POT, PS, and PV GWBs was calculated, Milkov Ivanov [48] obtained values of 0.2, 0.1, and 0.1 in PG, PVN and PVS GWBs, respectively. This study completes a SITE evaluation of 0.32 in PX GWB (Figure 6). The overall SITE values evidence moderate SWI. The similar values of the T parameter (temporality or seasonality) provide evidence that they share similar hydrogeological characteristics. This highlights the idea that the NCB values should be similar for such aquifers. There exist sources of uncertainty, associated with several factors. For example, the accuracy of the isochloride map depends on the number of monitoring stations.

4.4. Comparison of Statistical Techniques and SITE Index

The proposed approach aims to combine information from different sources in such a way that decision-makers can obtain a unified view of the data within the planning process. Hence, the method allows more flexibility for the NCB calculation under the uncertainty inherent to the heterogeneous spatial and temporal distributions.

To synthesize the information from the different sources, we propose plotting the regression line on the pairs composed by the reference levels obtained with each method on their corresponding SITE index (Figure 7). As mentioned above, PG GWB is not included in the linear fit of the Iterative 2-σ with no trend, because data from all the monitoring stations show a monotonic trend.

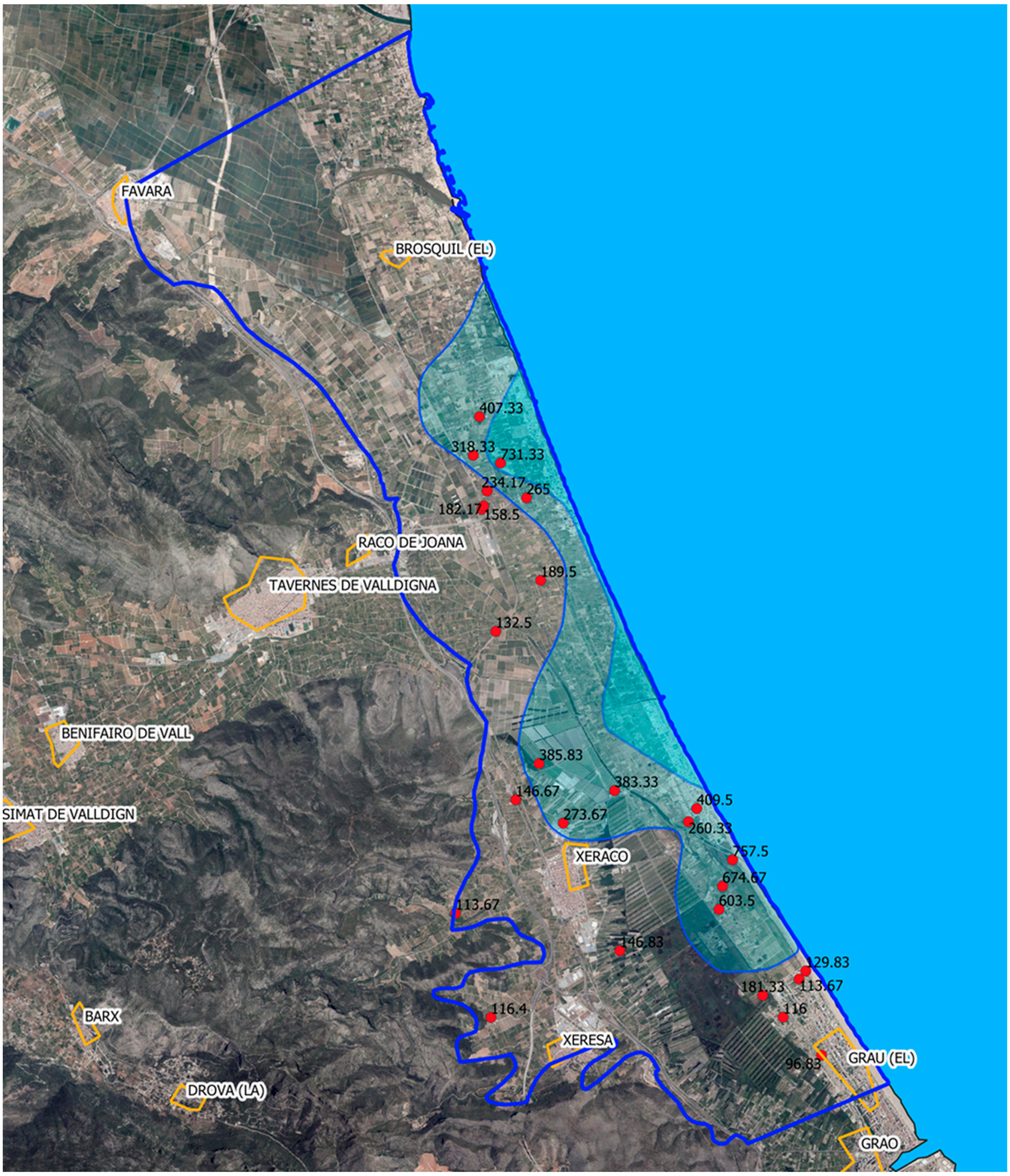

Figure 6. Example of the SITE index application to infer isochloride maps in the Plana de Xeraco GWB.

4.5. Regional NCB Data

As described above, the regional NCB data deduced from the application of the CMB method in Continental Spain identify the atmospheric fraction that contributed to the calculated NCB with the statistical techniques and SITE index. Therefore, the NCB data from the CMB method are the key to conceptualizing the findings of the statistical techniques and SITE index. The NCB data (Figure 8; Table 6) from the CMB method vary from 24.2 mg L^{-1} in PVS GWB to 48.6 mg L^{-1} in PG GWB.

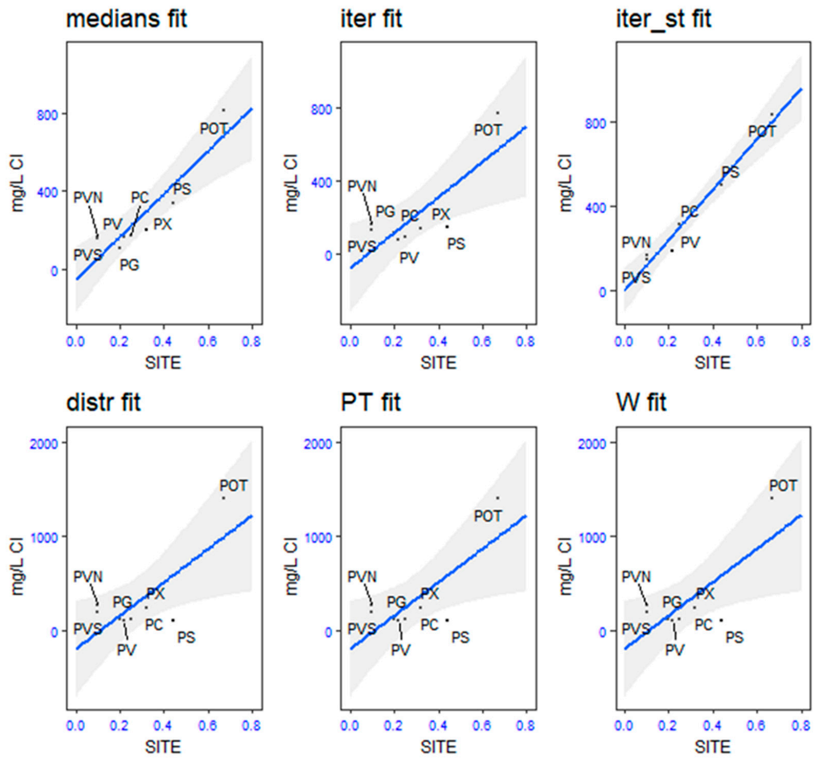

Figure 7. For the eight studied GWBs, comparison of average NCB values from the six statistical techniques and SITE index. The GWBs are labeled as follows: PC—Plana de Castellón; PG—Plana de Gandía; POT—Plana de Oropesa–Torreblanca; PS—Plana de Sagunto; PV—Plana de Vinaroz; PVN—Plana de Valencia Norte; PVS—Plana de Valencia Sur; and PX—Plana de Xeraco.

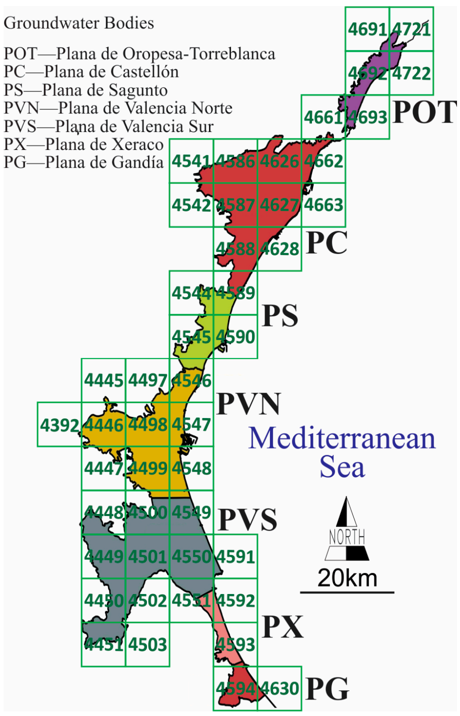

Figure 8. Location of the selected coastal GWBs in Eastern Spain and discretization of the 10 km × 10 km cells (with original labeling) for distributed NCB in the part of Continental Spain covered in the study area.

Table 6. Categories of seawater intrusion according to the SITE index.

	POT	PC	PS	PVN	PVS	PX	PG
[1] mNCB	41.4	35.4	40.4	36.3	24.2	36.2	48.6
σNCB	11.6	9.31	10.1	8.89	6.11	9.55	12.5
cvNCB	0.28	0.26	0.25	0.25	0.25	0.26	0.26

Notes: [1] mNCB and σNCB are mean value and standard deviation of NCB data in mg L^{-1}, and cvNCB is the dimensionless coefficient of variation of NCB. GWBs: PC—Plana de Castellón; POT—Plana de Oropesa–Torreblanca; PS—Plana de Sagunto; PG—Plana de Gandía; PVN—Plana de Valencia Norte; PVS—Plana de Valencia Sur; and PX—Plana de Xeraco.

5. Discussion

Figure 9 shows the results obtained by combining the six selected methods described above and the SITE index for each aquifer. Assuming that the Cl concentrations depend on the level of SWI affection in each aquifer, a linear regression can be parameterized, which relates the NCBs obtained through each method and the SWI index.

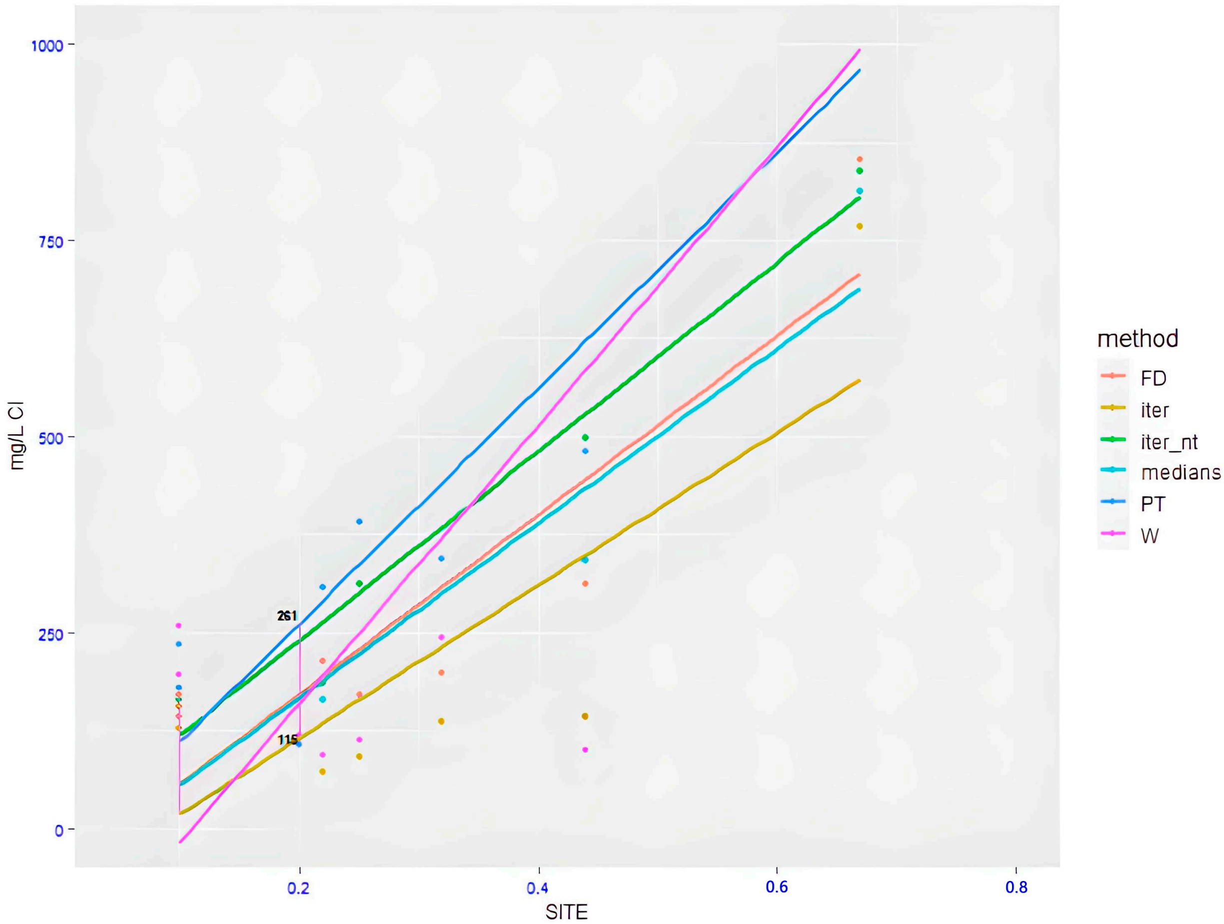

Figure 9. Interception of regression lines with SITE index 0.1 and 0.2. Abbreviations: FD—distribution function; iter—iterative technique; iter_nt—iterative technique with no trend; medians—medians; PT—PT method; and W—Walter method.

In most of the studied GWBs, the NCBs obtained by applying the 2-σ technique are significantly lower than the ones obtained with the other approaches (Table 4). The exceptions are PG GWB, where the medians show similar values than the rest of the methods, and PS GWB, where only the results provided by the Walter approach are lower than the 2-σ method technique. In this case, this may be related to the fact that the data exhibit strong non-normality (p-value of 2×10^{-16}), so the majority of the upper range of values is removed, resulting in an increase in the slope of the linear regression model.

The Walter and PT techniques are not sensitive to the applied salinity constraint in most of the cases. In general, they provide higher values than the other techniques, while the iterative 2-σ is the technique that produces the lowest results.

The calculated NCB values vary between 115 mg L^{-1} and 261 mg L^{-1} (Table 7). The results correspond to the point of intersection between the regression line and the value 0.2 on the ordinate axis. The reason for selecting this value (Table 5) is based on an extensive knowledge of the area and the expert judgement of local hydrogeologists. Indeed, for values of SITE in the neighborhood of 0.1 or less, SWI can be considered negligible, while for values in the neighborhood of 0.2, it is is low. Certainly, GWBs with values of SITE less than 0.2 match areas where groundwater overexploitation has less importance. Some clear examples are the PVN and PVS GWBs, where piezometric levels remained constant during the evaluated period.

Table 7. NCB values (mg L^{-1}) obtained in the studied GWBs by applying the different statistical techniques.

Intersection of Regression Line for Each Method and SITE Index		
SITE value	0.1	0.2
Median	56	167
2-σ	18	115
2-σ no trend	119	240
Distribution Function	57	172
PT	111	261
Walter	−18	159

Both the NCB data from the statistical techniques and SITE index were also compared with regional NCB data deduced from the CMB method applied in Continental Spain. From Table 6, it follows that Cl concentrations in the selected GWBs (Figure 8) are between 24.2 mg L^{-1} and 48.6 mg L^{-1}, attributable to the atmospheric salinity that reaches the water table. Consequently, based on the NCB values obtained from the CMB method, it makes sense to select 0.2 (Figure 9) as the default value of the SITE index to consider the increase in concentration of the NCB due to the interactions described.

6. Conclusions

Characterizing groundwater flow in coastal aquifers with complex hydrogeological boundary conditions is a problematic issue, as the magnitude of SWI at a specific monitoring station depends on several factors, such as the well depth, distance to coastline, and hydrogeological properties of the aquifer. Moreover, the observation points of the monitoring networks show high spatial and temporal heterogeneities, thus contributing to unconfident classifications. Despite this, the estimation of NCBs must be addressed to determine threshold values and quality standards.

Several approaches have been addressed to estimate NCBs, such as numerical modeling and different statistical methods. A majority of these models applied to date are based on the elimination of outliers that make the distribution deviate from the one that would occur without direct human influence (usually the normal distribution). However, all of them require a large amount of information, which is not always available. In addition, when the degree of intrusion is high, all monitoring stations can be affected, so there are no affordable criteria to eliminate a part of the values. In view of the above, it follows that it is difficult to tackle the problem of NCBs' definition from the perspective of traditional approaches applied to date.

This paper introduces a methodological framework to deduce NCB values in coastal GWBs affected by SWI which includes different statistical techniques, an indicator of SWI affection called SITE index, and a previous regional evaluation exclusively attributed to atmospheric Cl.

This paper also discusses the applicability of the applied methodology in eight porous coastal GWBs in Eastern Spain. These GWBs share similar hydrogeological features, making them suitable for reliable comparisons, but they have different spatial and temporal CL data coverage and degree of SWI affection induced by different local groundwater demands, thus providing different NCB values.

This methodology allows us to demonstrate the wide uncertainty and discrepancies in the results that different techniques lead to due to numerous sources of uncertainty, simplifying assumptions made in each of them, and the availability and reliability of existing data. Therefore, this paper highlights the risks for an adequate assessment of uncertainty and obtaining reliable results if only one technique is used.

In this way, the procedure enables us to calculate NCBs for GWB sharing different SWI effects, including those completely degraded. The results show how the applied methodology is confident enough to characterize NCBs in coastal GWBs and possesses the advantage of having a significantly lower computational cost than conventional groundwater numerical models using variable-density modeling tools. In particular, in the Eastern Spanish coastal fringe, we recommend selecting a threshold of 0.2 for the SITE index, which gives a concentration range between 115 mg L^{-1} and 261 mg L^{-1} for NCBs. In general, the approach could also be generalized to other coastal regions by means of a comprehensive analysis of the SWI indices and other sources of Cl, as well as the basic aquifer characteristics of the area. To achieve this, the hydrogeological expertise is mandatory in order to interpret the hydrogeological meaning of the results achieved from available Cl datasets from different monitoring networks operated over different time and spatial resolutions.

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