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Martí Ruffi-Salís^{1,2} , Jordi Garcia-Orellana^{1,3} , Gerard Cantero⁴, Jordi Castillo⁵, Almudena Hierro⁶, Joan Rieradevall^{1,2} and Joan Bach⁷

¹ Institut de Ciència i Tecnologia Ambientals (ICTA), Z Building, Universitat Autònoma de Barcelona (UAB), Campus UAB, 08193 Bellaterra, Barcelona, Spain

² Department of Chemical, Biological and Environmental Engineering, Q Building, Universitat Autònoma de Barcelona (UAB), Campus UAB, 08193 Bellaterra, Barcelona, Spain

³ Department of Physics, Facultat de Ciències, Universitat Autònoma de Barcelona (UAB), Campus UAB, 08193 Bellaterra, Barcelona, Spain

⁴ Fundació Mar, Llibertat Street 1, 17200 Palafrugell, Girona, Spain

⁵ Monteditorial, Carme Street 37, 08500 Vic, Barcelona, Spain

⁶ Institut de Tècniques Energètiques, Universitat Politècnica de Catalunya, 08028 Barcelona, Spain

⁷ External Geodynamics and Hydrogeology Group, Universitat Autònoma de Barcelona (UAB), Campus UAB, 08193 Bellaterra, Barcelona, Spain

E-mail: Jordi.Garcia@uab.cat

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Supplementary material for this article is available [online](#)

Abstract

The 20th century has been characterized by an exponential population growth, with a high density in coastal zones. The aim of this work is to study the impact of land use changes on the hydrological cycle, and possible consequences in marine environment. The study has focused on the relationship between coastal urbanisation and submarine groundwater discharge (SGD) that may cause an alteration in the biogeochemical cycles of marine ecosystems. An analysis of land use changes, historical salinities and nutrients data in the Maresme (Barcelona) coastal zone have revealed that the percentage of urban and forest zone land use has increased since 1990, corresponding to a decrease of nutrients in coastal waters through SGD. These impacts may cause changes in coastal biogeochemical cycles, like the decrease in chlorophyll concentrations. Results denote the correlation between land use, coastal aquifer dynamics and the effects of SGD in coastal environments.

1. Introduction

Urbanization of coastal areas worldwide has rapidly accelerated during the second part of the 20th and the first decade of the 21st centuries, and is one of the main causes of environmental changes (Grimm *et al* 2008). San Francisco, New York, Shanghai, Cape Town, Rio de Janeiro and Barcelona are remarkable examples of these rapidly growing coastal mega-cities. This urbanization increase has led to a cascade of environmental changes, as for example, by impacting groundwater resources (Foster 2001, Michael *et al* 2005). Groundwater resources, in terms of aquifer recharge and water geochemical properties, are also affected by rural flight or spread of forest areas.

The relationship between land uses changes, characterised by an urban area expansion, and the quality of groundwater resources has been extensively studied (McFarlane 1984, Tellam 1995, Cronin *et al* 2003, Taniguchi *et al* 2007). The majority of the studies have reported worsening groundwater quality in response to increased urbanization. There are several anthropogenic impacts that may alter the characteristic complexity of coastal groundwater systems, such as the construction of new neighbourhoods, facilities (e.g. harbours, airports), expansion of roads, railroads, malls, etc. One of the most important characteristics of this urban expansion with respect to coastal aquifer dynamics is the large-scale change in surface permeability, driven by an increase in paved-surfaces. Decreased infiltration can affect both water quality and quantity of aquifers systems,

assessed through specific parameters: piezometric levels, conductivity or nutrients concentrations (e.g. nitrates or silicates).

Submarine Groundwater Discharge (SGD) is an important component of the hydrological cycle in coastal aquifers and is a source of nutrients and micronutrients to coastal areas (Rengarajan and Sarma 2015, Rodellas *et al* 2015, Trezzi *et al* 2016). SGD is generally composed of a mixture between freshwater and circulated seawater, the latter of which is either recirculated through intertidal and subtidal sediments or derived from seawater intrusion which has infiltrated into depleted coastal aquifers (Burnett *et al* 2003). Therefore, any alteration in coastal aquifers due to changes in land use could trigger an impact in SGD flows, and hence its impact on coastal biogeochemical cycles. For example, along the Kona coast of Hawaii, SGD-driven nitrate and silicate loads were found to vary along shoreline segments with respect to population density, the percentage of bare land and the presence or absence of a golf course. On Maui, SGD-driven nitrogen loads are greatest for sugarcane and pineapple farming land use areas (Bishop *et al* 2017). A Long Island, NY embayment experiences high SGD-driven nitrogen loadings along shoreline segments that are classified as high-development land use (Young *et al* 2015).

Considering these premises, the aim of this work is to study the interrelation between land use changes, the alteration of coastal aquifers dynamics and the consequent alteration of submarine groundwater discharge (SGD) flow and nutrients fluxes to coastal ocean.

2. Materials and methods

2.1. Study area

Maresme county has an area of 398 km² and is located in the western Mediterranean Sea (figure S1(A) is available online at stacks.iop.org/ERC/1/031005/mmedia), in the region of Catalonia (figure S1(B)). The county is situated along the sea, with 50 km of coastline and a width between 5 and 15 km. The climatology of this region is characterized by Western Mediterranean conditions, and the region experiences high temperatures in summer (mean ~23 °C) and mild temperatures in winter (mean ~10 °C). The annual precipitation of the area ranges from 500 to 800 mm and is mainly related to intense and short storms during the fall and spring season. The urban extension of Maresme county has significantly increased (Mateu 1985, Benaiges and Corbera 2007) during the second part of the 20th century, in part due to its proximity to the metropolis of Barcelona, with a population of more than 3,000,000 inhabitants. The present study is focused on the central area (CA) of the Maresme region, including the municipalities of Argentona, Cabrera de Mar and Mataró (county capital). This decision was based on the importance of the Argentona ephemeral stream (i.e. Riera d' Argentona) where surface water flows are restricted to the rainy seasons, particularly during major rainfall events (e.g., >50 mm in a few hours) (Cerdà-Domènech *et al* 2017). Relevant data from the northern area (NA) (with the municipalities of Arenys de Munt, Arenys de Mar and Sant Vicenç de Montalt) and southern area (SA) (with the municipalities of Alella, Teià and El Masnou) were included in this study, when necessary, to help with the discussion. The Maresme region is composed of a series of coastal aquifers, which can be assessed according to: (1) land use, (2) aquifer dynamics and (3) SGD and its influence on the coastal environment.

2.2. Land use temporal quantification

Land use was assessed by the digitalization of orthophotos from the Cartographic and Geological Institute of Catalonia, by using software Miramon 7.1. The reference years selected were 1956 (first cartographic data available), 1990, 2000 and 2010 (figures S2 to S5, respectively). Types of land use considered were Urban, Forest and Agriculture.

2.3. Hydrology data acquisition

A summary of data used in the present study is shown in table S1. Data sources are described in Content 1 of the Supporting Information. The location of the sampling points is shown in figures S6 to S9.

2.4. Hydrology data assumptions

There are several techniques and analytical methods that have been used as tracers of SGD (e.g. salinity, Rn, Ra-isotopes, heat, seepage meters, etc). Dissolved inorganic silicate (DSi) data was selected as the main approximation of SGD because unlike nitrate or phosphate, DSi is not a fundamental macronutrient in the Mediterranean Sea and consequentially is less biologically-sensitive in the coastal marine environment (Hwang *et al* 2005, Garcia-Solsona *et al* 2010, Null *et al* 2014). Chlorophyll data was selected as an approximation for phytoplankton biomass coastal productivity (Eisner *et al* 2016). Recharge estimation was obtained according to the values stated at table 1. Recharge model used is described in the Supplementary Information.

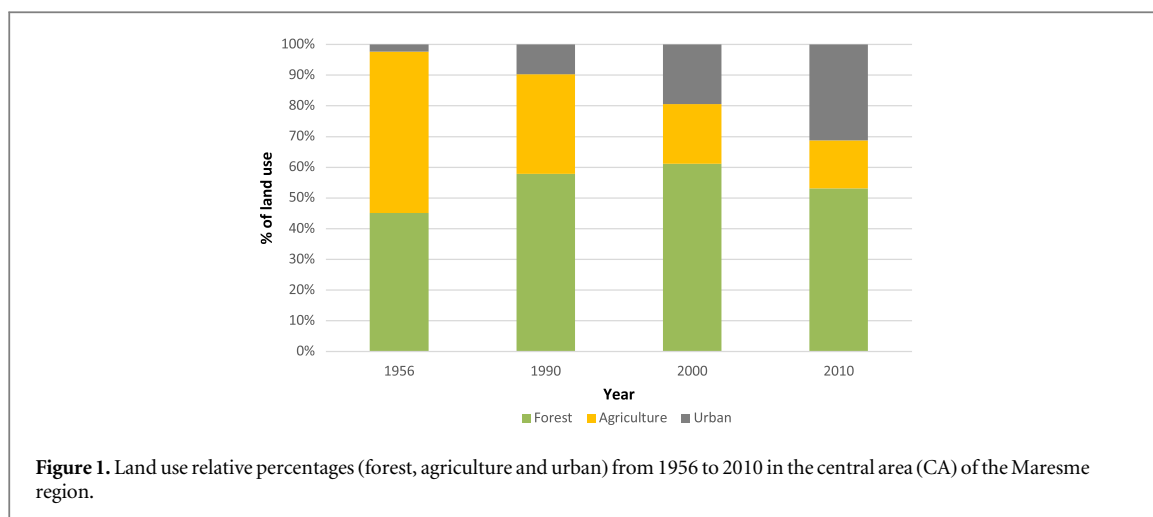


Table 1. Parameters used in the recharge model.

Parameter	Forest	Agriculture	Urban
Field capacity (volumetric contents)	0.25	0.25	—
Initial humidity (volumetric contents)	0.25	0.25	—
Root thickness (m)	0.5	0.15	—
Wilting point (volumetric contents)	0.1	0.1	—
Lamination value (mm)	100	100	0
Net reservoir (mm)	75	22.5	0
Initial reservoir (mm)	75	22.5	0

3. Results and discussion

3.1. Land use

Land use variation over the last 60 years is shown in figure 1. It can be observed that in 1956 there was a predominant use of land for agricultural purposes (71%) with respect to forests and urban areas (25% and 3%, respectively). In the succeeding years following 1956, the percentage of urban areas increased from 3% (1956) to 30% (2010). This urbanization process was greatest during the period 1990–2000 (from 12% to 29%). During this time, agricultural areas, historically known to be the most important in the Catalonia region and dedicated to growing potatoes (until 1968) and strawberries and ornamental plants (from 1965) (Badosa 2013), were abandoned and as a consequence, the percentage of forest mass increased. Regarding to the spatial distribution of land uses changes, urban areas tend to be located in the flattest zone of the region (closer to the coast), while forests are more likely to be located near the mountain range (*Serra de Marina*) (figures S2 to S5).

3.2. Aquifers

Piezometric levels of the aquifer did not follow a clear temporal trend (figure 2). It is difficult to establish a simple correlation between piezometric levels and a single factor, such as precipitation or land use. This complexity could be caused by the influence of other factors such as the supply of an external water network. Specifically, the local drinking water company (ATLL) has supplied an average of 21.8 hm³ of external water to the Maresme county yearly since 2005. As a consequence, groundwater pumping and withdrawal in the region is minor, thereby recharging the aquifer. In addition, another factor that could decrease water pumping is the drop of agricultural activities in the analysed time period, reducing the overall water needed for crop irrigation.

In terms of conductivity, different values and trends are observed, depending on the distance of any individual well to coastline (figure 3). Conductivity values in piezometers closest to the coastline were higher (up to 2200 $\mu\text{S}\cdot\text{cm}^{-1}$ in some cases) than inland piezometers as a result of coastal aquifer-seawater interactions (figures 3 and S8). Nitrate concentrations generally decreased in the majority of wells analysed over time. However, a notable peak was observed during 2006–2008, corresponding to a drought in Catalonia during those three years (March and Saurí 2013) (figure 2).

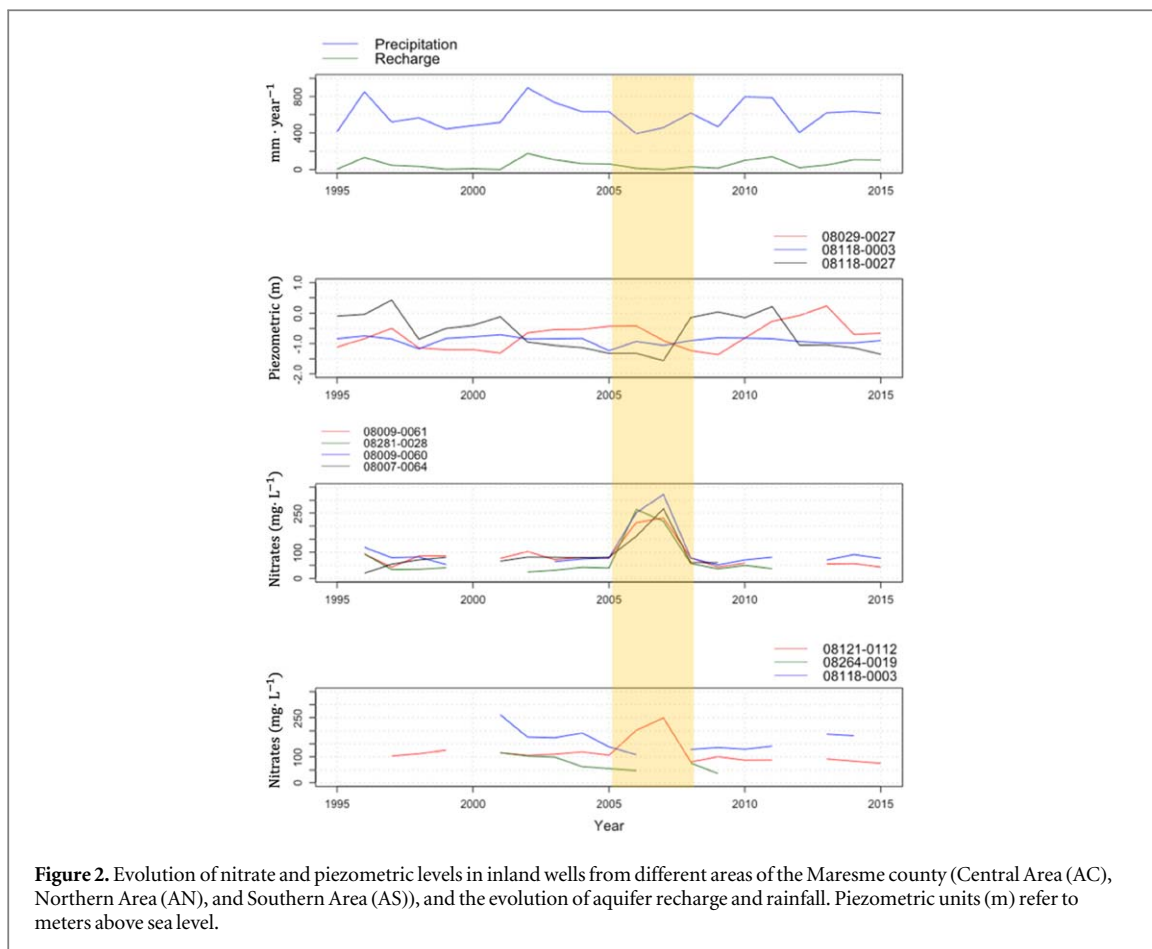


Figure 2. Evolution of nitrate and piezometric levels in inland wells from different areas of the Maresme county (Central Area (AC), Northern Area (AN), and Southern Area (AS)), and the evolution of aquifer recharge and rainfall. Piezometric units (m) refer to meters above sea level.

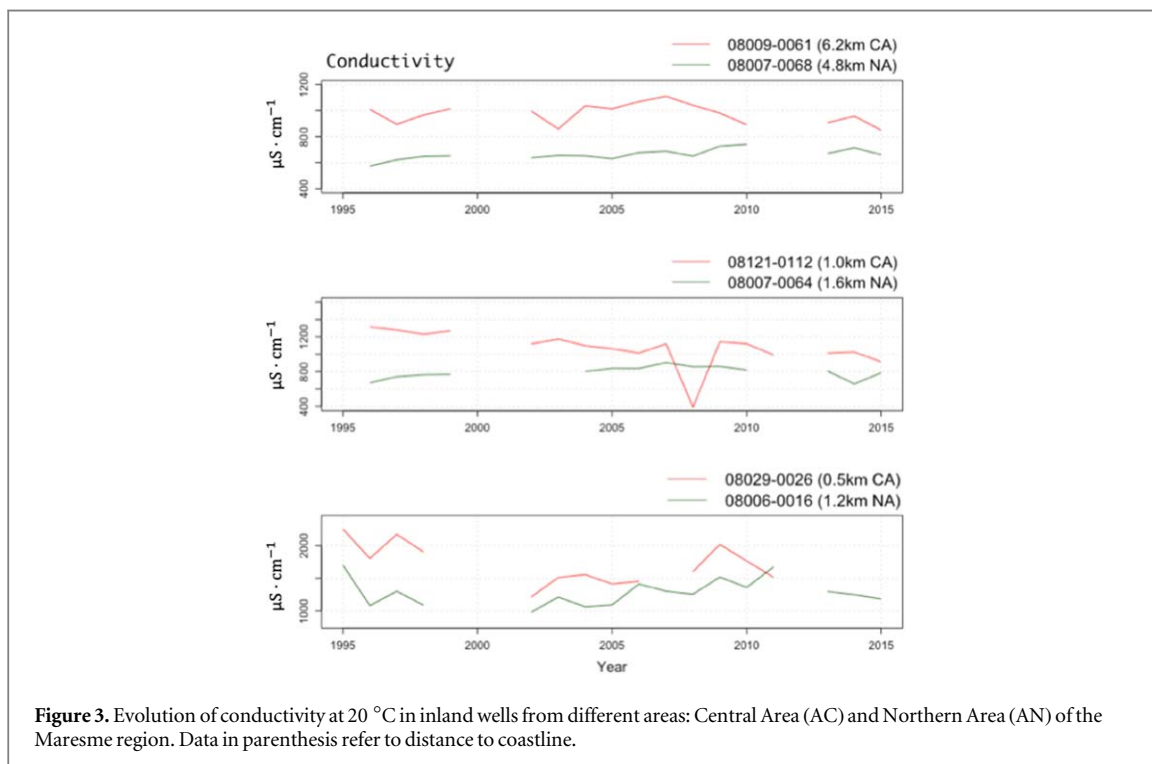


Figure 3. Evolution of conductivity at 20 °C in inland wells from different areas: Central Area (AC) and Northern Area (AN) of the Maresme region. Data in parenthesis refer to distance to coastline.

3.3. Seawater quality

In the central part of the studied area (CA), an increase of salinity in coastal waters can be observed, from ~34 to 38 psu from 1994 year to 2014 year (figure S10). Silicate concentrations decreased over the same time period (figure S11), from 0.31 mg·L⁻¹ (1995) to 0.08 mg·L⁻¹ (2012), while chlorophyll concentrations (figure S12)

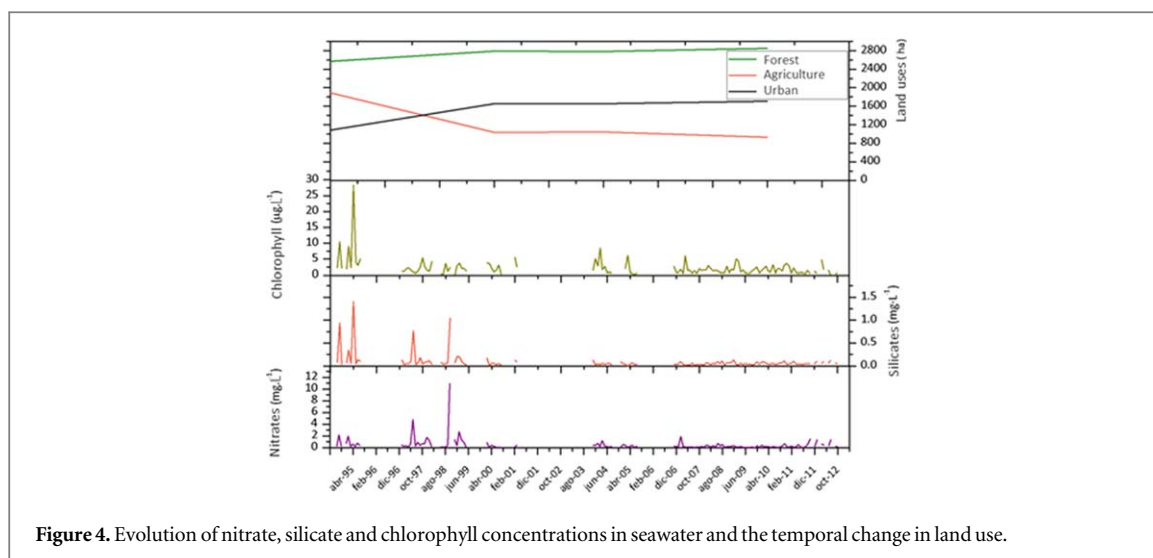


Figure 4. Evolution of nitrate, silicate and chlorophyll concentrations in seawater and the temporal change in land use.

decreased from $3.86 \mu\text{g}\cdot\text{L}^{-1}$ (1994) to $1.33 \mu\text{g}\cdot\text{L}^{-1}$ (2012). Both trends are observed throughout the study period in the seawater adjacent to the Argenton ephemeral stream (CA). In other locations of the Maresme region, silicate concentrations and salinity were nearly constant over the studied period (figures S11 and S12). These parameters denote that there were not visible changes in the SGD flow in these zones during the period under study.

3.4. Interrelation between land use, aquifers and submarine groundwater discharge

Threshold precipitation (necessary precipitation for runoff) decreased with increasing urban land use ($378 \text{ ha}\cdot\text{y}^{-1}$ from 1990 to 2000), leading to a ground waterproofing process. This urbanization process triggered an increase of urban water (i.e. rainwater fallen on urban land) during the 1956–2010 period, from 5 to 49 hm^3 . Concurrently, agricultural land also decreased ($-523 \text{ ha}\cdot\text{y}^{-1}$ from 1990 to 2000) (figure 1).

Additional factors influenced the decrease in recharge, including the annual variation in rainfall and water uptake and evapotranspiration of the forest mass due to its increase in surface area (50 to 70 hm^3 in the 1956–2010 period). The drop of agriculture activities (from 3.5% to 1.6% in GDP contribution of the primary sector in the 1991–2006 period (Tirado 2007)), related to the reduction of groundwater pumping from aquifers, could also influence groundwater recharge due to a pressure decrease from groundwater extraction.

Nitrates concentrations decreased in most of the wells closest to the shoreline (figure 2), (e.g. well 08009–0060 from $118.8 \text{ mg}\cdot\text{L}^{-1}$ in 1996 to $77.1 \text{ mg}\cdot\text{L}^{-1}$ in 2015) where agricultural and urban land uses have historically been located (figures S2 and S3). Despite the influence of the 2006–2008 drought on both nitrates and conductivity, the latest was also found to decrease overtime. The decrease in nitrate concentration and conductivity could be linked to the decrease in agricultural land use ($20,750$ to $6,895 \text{ ha}$ in the 1956–2010 period), related to the extensive use of fertilizers and water demand for agricultural purposes, respectively. There was a strong relationship between the decrease in precipitation and piezometric levels, and the increase in nitrate concentrations, during the general drought of 2006–2008 (yellow shade in figure 2).

Moreover, the influence of the sea is clearly observed on the groundwater conductivity (figure 4). Higher average conductivity values (up to $2000 \mu\text{S}\cdot\text{cm}^{-1}$) and wider fluctuations denote a higher sensitivity of wells closest to the sea. In specific times, there were piezometric levels (in sample points close to the coastline) that were below sea level (figure 2); such difference could drive saltwater intrusion into the coastal aquifer, as indicated by higher conductivity values in nearby wells.

Focusing on seawater, figure 4 shows weak correlations between nutrient concentrations (nitrate and silicate) and biotic activity (chlorophyll concentration), such as silicates ($1.4 \text{ mg}\cdot\text{L}^{-1}$) and chlorophyll ($28.3 \mu\text{g}\cdot\text{L}^{-1}$) in 1995. However, other factors like ocean currents (Raimbault *et al* 1993) or nutrient uptake by biological activity may affect these correlations, as in 1998 (peaks of nitrate and silicate).

The largest variations in both nutrients and chlorophyll ($3.5 \mu\text{g}\cdot\text{L}^{-1}$ on average) were located within the period 1990–2000, where land use showed the largest variation. Land use variation got stabilised in 2000, corresponding to fairly constant nutrient (e.g. silicates $0.10 \text{ mg}\cdot\text{L}^{-1}$) and chlorophyll ($1.5 \mu\text{g}\cdot\text{L}^{-1}$) concentrations. This suggests that there was a relationship between land use changes and the alteration of the biochemical cycles in the coastal sea. These observations are similar to those showed by other studies where Ra-isotopes were used as a SGD tracer (Tang *et al* 2015).

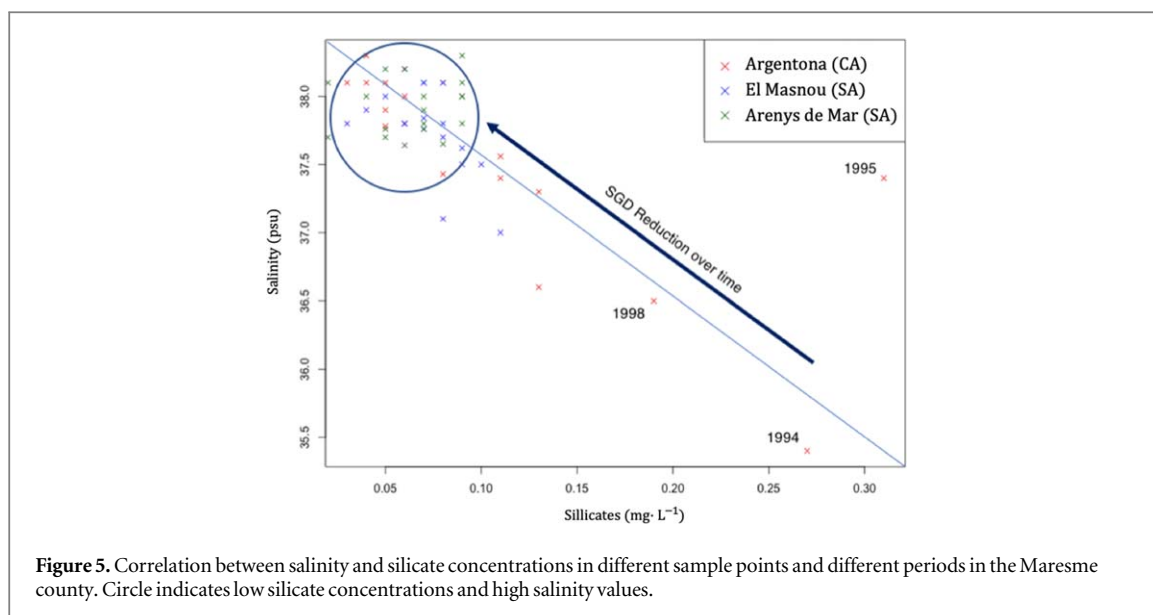


Figure 5. Correlation between salinity and silicate concentrations in different sample points and different periods in the Maresme county. Circle indicates low silicate concentrations and high salinity values.

During the studied period of time, there was an inverse correlation between silicate concentration and salinity in the Argentona ephemeral stream (Mataró) before 2000, for example in 1994 ($0.27 \text{ mg}\cdot\text{L}^{-1}$; 35.4 psu) and in 1998 ($0.19 \text{ mg}\cdot\text{L}^{-1}$; 36.5 psu) (figure 5). This inverse correlation shows the existence of important SGD flows (considering silicate as SGD tracer). Furthermore, as seen in other sampling sites such as Arenys de Mar or El Masnou, also in the Maresme county, the correlation between silicate concentration and salinity was not as significant as in the Argentona ephemeral stream, suggesting that SGD was less important in these areas. In addition, the silicate-salinity correlation in the Argentona ephemeral stream faded with time, reaching a salinity of ~ 38 psu with negligible silicate by year 2000. Therefore, this drastic fading of the relationship between salinity and silicates, the stabilization of salinity after 2000 and the no observation of any significant peak of silicates, nitrates and chlorophyll would indicate a decrease in the constant influence of the SGD in the area rather than a decrease in the influence of the ephemeral stream.

Therefore, land impermeability due to the construction of new infrastructures such as malls, motorways or airports and changes in land use from agriculture to urban or forest areas around large coastal cities, such as Barcelona, may alter land-ocean interactions and reduce, for example, allochthonous nutrient fluxes to coastal water triggered by SGD. These changes and the pollution of water and sediments due to anthropogenic activities may considerably alter coastal ecosystems, reducing primary production and biodiversity, especially in semi-arid regions as the oligotrophic Mediterranean Sea, where runoff is limited.

4. Conclusions

There was a clear relationship between urbanization over areas occupied by unconfined coastal aquifers and SGD in Maresme (Barcelona). The exponential population growth of the last century has created a high population-density in coastal areas. Urbanization has therefore triggered changes in the hydrological cycle by altering groundwater recharge through the paving of large surface coastal areas, the abandonment of agricultural areas and an increase of forest cover. In the present example, it was not possible to establish quantitative relationships in terms of historical piezometric levels, salinity and nutrient concentrations in groundwater (DIN) and coastal seawater (DSi) with respect to land use changes; however, we have established a qualitative pattern. There was no clear trend in piezometric levels and land use, according to the location of the piezometers; this may be partially driven by the abandonment of agricultural areas, improvements in agricultural irrigation or external water supply from a drinking water company. Moreover, the increase in forest cover could have caused a decrease in the water table in some specific areas. Nitrate concentrations decreased over 2000–2010 time interval, probably due to the abandonment of agricultural areas in the flatter part of the region and improvements of agricultural techniques. These combined alterations to the hydrological cycle and groundwater quality altered submarine groundwater discharge (SGD) and its associated nutrient flux to the coastal ocean. Analysis of historical salinity and nutrient concentrations in coastal waters showed a steady reduction in the central area (CA) of the study area. Land use driven changes to groundwater quantity and quality may have caused changes in some biogeochemical cycles of the marine environment through SGD, as qualitatively observed by decreasing chlorophyll concentrations in coastal surface waters (from $7.7 \mu\text{g}\cdot\text{L}^{-1}$ in

1995 to $1.3 \mu\text{g}\cdot\text{L}^{-1}$ in 2012). Further research is needed to extrapolate the results found in the present study to worldwide locations where the process of urbanization affects coastal zones. This aspect is of great importance in terms of groundwater resources management and for biotic activity in coastal seawater.

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All data used in this study is available upon request to the corresponding author.

ORCID iDs

Martí Rufi-Salís  <https://orcid.org/0000-0003-3696-1033>

Jordi Garcia-Orellana  <https://orcid.org/0000-0002-0543-2641>

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