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Seawater $^{87}Sr/^{86}Sr$ ratios along continental margins: Patterns and processes in open and restricted shelf domains

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ABSTRACT

To better constrain the Sr isotope budget in marginal domains without any fluvial inputs, we analyzed the chemical composition and $^{87}Sr/^{86}Sr$ ratio of waters and shells from four locations: two coastal lagoons, one hemipelagic platform and one open marine shelf. Our results highlight homogeneous $^{87}Sr/^{86}Sr$ ratios typical of oligotrophic oceanic waters (OOW) (i.e., 0.709172 ± 0.000023) in the Pacific Tatakoto atoll and along a Mediterranean shore to offshore transect (~25 km off Banyuls-sur-Mer, BSM). This attests that oceanic inputs from oligotrophic areas remain the main Sr source in open shelf areas compared with submarine groundwater discharges (SGD) or particulate dissolution influences. In BSM, only foreshore data are more radiogenic, possibly due to rainwater mixing, local groundwater springs or more efficient particle dissolution in the intertidal zone. In restricted areas, we report variable $^{87}Sr/^{86}Sr$ ratios between the Salses-Leucate (France) and Oualidia (Morocco) lagoons. The first one has homogeneous $^{87}Sr/^{86}Sr$ ratio typical of OOW except close to SGD. In Oualidia, $^{87}Sr/^{86}Sr$ ratios decrease by 1.2×10^{-3} from OOW values close to the Atlantic inlet to progressively less radiogenic ones upstream within the interior of the lagoon. These differences depend on several factors including the leaky, restricted or choked morphology of lagoons modulating the oceanic Sr inputs, but also SGD fluxes whose $^{87}Sr/^{86}Sr$ ratios and Sr concentrations are highly variable according to the nature of rocks leached in karstic aquifer. In Oualidia, the low $^{87}Sr/^{86}Sr$ ratios correspond to high Sr concentrations (up to 150 $\mu mol \cdot l^{-1}$) issued from the dissolution of Mesozoic evaporites, leading to SGD fluxes accounting for 60% of the local Sr budget. Through data compilation, we show that similar $^{87}Sr/^{86}Sr$ gradients and processes prevail at the whole Mediterranean scale. Finally, we postulate that high coastal water retention times can also account for anomalous coastal $^{87}Sr/^{86}Sr$ ratios and that the combination of water mass restriction, SGD, bioadsorption and early diagenetic processes could decrease seawater Sr concentrations in some marginal areas.

1. Introduction

Although the strontium isotope compositions ($^{87}Sr/^{86}Sr$) of river waters and submarine hydrothermal sources strongly vary across the globe (with respective averages of 0.7119 and 0.7037; Palmer and Edmond, 1989, 1992; Bach and Humphris, 1999; Davis et al., 2003; Pearce et al., 2015; Chavagnac et al., 2018), the $^{87}Sr/^{86}Sr$ of oligotrophic oceanic waters (OOW) remains homogeneous worldwide at 0.709172 ± 0.000023 (see El Meknassi et al., 2018 for a recent synthesis). This paradigm is due to the long residence time of Sr in the ocean (i.e., ~2.5 Myr; Hodell et al., 1990) compared to the global ocean mixing duration ranging from 1000 to 2000 years (DeVries and Primeau, 2011). However, the OOW $^{87}Sr/^{86}Sr$ ratios fluctuated markedly through the Phanerozoic, as reported from Sr isotope data of fossil archives (Peterman et al., 1970; Burke et al., 1982; Veizer, 1989; Zaky et al., 2018). These secular variations are ascribed either to global geodynamic events modulating the respective contribution of terrestrial (radiogenic) and hydrothermal (unradiogenic) Sr sources to oceans or

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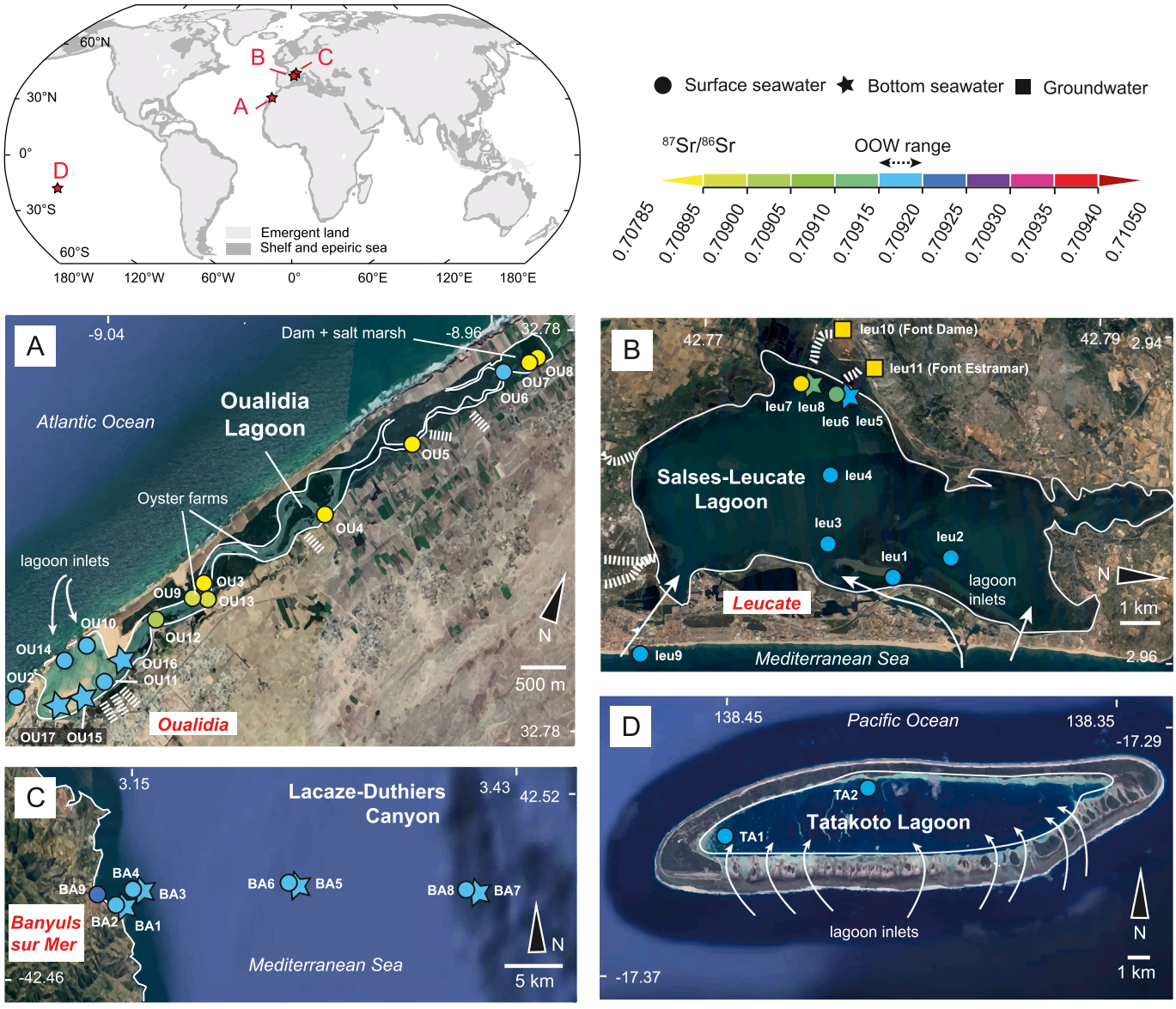

Fig. 1. Location and $^{87}Sr/^{86}Sr$ ratios of seawater and groundwater samples collected in the four studied sites: Oualidia lagoon (A), Salses-Leucate lagoon (B), Banyuls-sur-Mer (BSM) transect (C), Tatakoto atoll (D). Dashed lines indicate groundwater discharges.

to paleogeographic and climatic changes modifying the continental weathering of rocks of different age and lithology (Reeder et al., 1972; Brass, 1976; Goldstein and Jacobsen, 1987; Bataille et al., 2017; Peucker-Ehrenbrink and Fiske, 2019). By assuming a homogeneous OOW $^{87}Sr/^{86}Sr$ ratio at a global scale, these long-term $^{87}Sr/^{86}Sr$ variations are thus widely used for dating sedimentary series through chemostratigraphic calibration of new fossil data (McArthur, 1994; McArthur et al., 2012) or testing paleogeographic and climatic hypotheses (Goddéris et al., 2017).

Despite the popularity of this paleoenvironmental proxy, the question of whether fossil carbonate shells may be used to reconstruct past OOW $^{87}Sr/^{86}Sr$ fluctuations is still a matter of debate as most of these organisms thrived in epeiric and shelf domains potentially disconnected from the global oceanic Sr reservoir. This is partly supported by the apparent heterogeneity of modern shell and (euhaline to brachyhaline) seawater $^{87}Sr/^{86}Sr$ ratios from worldwide marine shelves, with respective ranges of 0.707636–0.710483 and 0.704638–0.710648 (precision on measured $^{87}Sr/^{86}Sr$ ratio is better than 10^{-5}; El Meknassi et al., 2018). Significant offsets between well-dated fossil (or bulk carbonate) $^{87}Sr/^{86}Sr$ ratios and the Phanerozoic isotope curve are also regularly reported, even in sedimentary facies indicating outer shelf contexts and/or normal marine environments (Cochran et al., 2003; Nieto et al., 2008; Sessa et al., 2012; Wierzbowski et al., 2012; Eidvin et al., 2014; Schildgen et al., 2014; Briard et al., 2020). While potential analytical, stratigraphical and diagenetic biases may be discarded (Martin and Scher, 2004: Marcano et al., 2015; Bellefroid et al., 2018; Zaky et al., 2018), these mismatches can indicate that, sporadically, seawater $^{87}Sr/^{86}Sr$ ratios of epeiric and shelf domains are not necessarily representative of the global OOW value. In most case, these variations result from dissolved Sr supplies delivered by river waters, with contrasted influences on seawater $^{87}Sr/^{86}Sr$ ratios according to their Sr concentrations and isotopic ratios linked to the age and lithology of local bedrocks (Palmer and Edmond, 1989, 1992; Bryant et al., 1995; Peucker-Ehrenbrink et al., 2010). This terrestrial Sr contribution is common in restricted environments like epeiric seas, estuaries, fjords, lagoons, and bays where oceanic influences are more restricted and where $^{87}Sr/^{86}Sr$ ratios covary landward with salinity (Andersson et al., 1992; Ingram and Sloan, 1992; Israelson and Buchardt, 1999; Basu et al., 2001; Major et al., 2006; Jones et al., 2014; Beck et al., 2013; Wang and You, 2013; Chakrabarti et al., 2018; Shao

et al., 2018). However, this river water influence can remain limited in euhaline marginal contexts as, on average, measurable effects on the seawater $^{87}Sr/^{86}Sr$ ratio are not expected to occur if freshwater inputs do not drop salinity below a threshold value of 12 (Bryant et al., 1995). Only 5% of world river systems have water $^{87}Sr/^{86}Sr$ ratios sufficiently different from the OOW value to significantly modify the local seawater $^{87}Sr/^{86}Sr$ ratio in euhaline marine environments.

Although poorly documented, seawater and shell $^{87}Sr/^{86}Sr$ ratios ranging from 0.70890 to 0.70921 have been yet reported in deep outer shelf domains and lagoons not directly subject to major river supply (Müller et al., 1990a, 1990b; Peckmann et al., 2001; Major et al., 2006; Huang et al., 2011; El Meknassi et al., 2018). This suggests that submarine groundwater discharges (SGD) or strontium released from particulate dissolution have a much stronger impact on the coastal Sr budget than currently expected (Beck et al., 2013; Jones et al., 2014; Trezzi et al., 2017). A direct consequence is that, depending on oceanic mixing and water retention times on shelves (i.e., ranging from a couple of days to decades or centuries; Liu et al., 2019), the Sr concentrations and $^{87}Sr/^{86}Sr$ of marginal areas, could not always be representative of the global oceanic reservoir. In other words, seawater from open shelves and epeiric domains could record Sr isotope anomalies just through cumulative effects of sporadic terrestrial Sr inputs in coastal sub-reservoirs not sufficiently mixed with the global oceanic reservoir.

Here, we test how submarine groundwater discharges (SGD) and water mass restriction can influence the Sr isotope composition of shelf water devoid of river influences by measuring and comparing the $^{87}Sr/^{86}Sr$ ratios of waters and shells from four coastal domains with different levels of connection to the open ocean. These include an open shelf Mediterranean transect, two coastal lagoons influenced by groundwater discharges in the Atlantic and Mediterranean contexts, and a Pacific hemipelagic platform. Through data compilation, our aim is also to better document the worldwide variability of seawater $^{87}Sr/^{86}Sr$ ratios in the coastal ocean and to better constrain the environmental processes acting on seawater $^{87}Sr/^{86}Sr$ patterns in these transition zones.

2. Geological setting

The seawater $^{87}Sr/^{86}Sr$ ratios of three restricted lagoons exhibiting different geomorphological, hydrogeological, hydrodynamical and climatic conditions is investigated in this study (Fig. 1): 1) The Tatakoto atoll in the eastern Tuamotu Archipelago (French Polynesia) in the southern part of the Pacific Ocean; 2) the Salses-Leucate lagoon (France) in the northwestern Mediterranean domain; and 3) the Oualidia lagoon (Morocco) in the North Atlantic domain. These three sites were chosen because they are under the influence of different littoral or reefal barriers, their water resources are issued from various karstic reservoirs, and they have different waves and tide forcing overprinted by different climatic conditions. To get a comparison with an open shelf domain, sampling was also carried out in the Mediterranean Sea along a 25 km-long transect from the rocky coastline of Banyuls-sur-Mer to the submarine Lacaze-Duthiers Canyon (western part of Gulf of Lion).

2.1. Oualidia lagoon (Morocco)

Formed around 8000–6500 BP (Ballouche and Carruesco, 1986), the Oualidia coastal lagoon is located on the western coast of Morocco (Abda Doukkala region) (Fig. 1A) and belongs to a coastal endorheic basin called the Coastal Sahel of Oualidia. The lagoon is 7 km long, 400 m wide, and 4 m water deep during the high tide season (Kaid Rassou et al., 2005). It is organized around a main channel of 180 m wide parallel to the coastline and separated from the Atlantic Ocean by a Plio-Quaternary shoal (Ballouche and Carruesco, 1986). The lagoon is connected to the ocean by two inlets to the south (130 and 70 m wide) (El Khalidi et al., 2011), but recent anthropic developments for salt exploitation and eutrophication prevention along the northern part allowed punctual oceanic inflows (Hilmi et al., 2009). Its hydrodynamic balance is influenced by: 1) Atlantic marine inflows linked to important semi-diurnal tidal dynamics (i.e., average amplitude of 0.97 m and main currents > 1 $m.s^{-1}$; Hilmi et al., 2005, 2009), 2) submarine groundwater supplies estimated from 0.2 to 1.2 $m^3.s^{-1}$ (Fakir et al., 2019), and 3) evaporation rates of 1.3 to 1.5 $m.yr^{-1}$ characteristic of semi-arid conditions in this region (with precipitation rates of 30 $mm.yr^{-1}$) (Hilmi et al., 2009). Along seasonal climatic and hydrodynamic conditions, the recurrent groundwater discharge to the lagoon is highlighted by a net salinity gradient ranging from 4 to 36 along a north to south transect (Hilmi et al., 2017). These groundwater supplies occur all along the lagoon (Fakir et al., 2019) and are predominantly linked to the extended karstic aquifer of the Coastal Sahel of Oualidia (Fakir et al., 2002; Kaid Rassou et al., 2005; Bouchaou et al., 2017). Its upper part (i.e., the most exploited for freshwater consumption) is composed of a 50 m thick Plio-Quaternary limestone-sandstone overlying a less permeable level of upper Hauterivian red clays. Below, the 30 m thick Dridrate limestone formation represents the lower aquifer which is less exploited but still provides the most abundant freshwater resources (Fakir et al., 2002; Kaid Rassou et al., 2005). Finally, the upper Valanginian marls constitute the substratum of the Dridrate aquifer and cover the Berriasian limestones, the upper Jurassic limestones rich in gypsum levels, and Triassic evaporites (Fakir et al., 2002; Fadili et al., 2015). According to Fakir et al. (2002), the dissolution of evaporites would be responsible for high Sr concentrations in local groundwater, reaching 2 to 2.5 times the OOW value.

2.2. Salses-Leucate lagoon (South of France)

The Salses-Leucate lagoon formed during the Flandrian (i.e., 15 kyr BP; Arnaud and Raimbault, 1969; Clanzig, 1987). It corresponds to a shallow coastal basin separated from the Mediterranean Sea by a sandy barrier interrupted by three narrow and dispatched marine inlets (two of them are artificial ones) (Ladagnous and Le Bec, 1997; Fig. 1B). The lagoon is 14 km long and 5 km wide, with an average water depth of 1.7 m reaching 3.7 m in its deepest areas (Stieglitz et al., 2013). The region of Salses-Leucate is characterized by a dry season typical of the Mediterranean climate, with annual rainfalls of 588 $mm.yr^{-1}$ and annual evaporation rates of 1.5 $m.yr^{-1}$ (Ladagnous and Le Bec, 1997). Basically, wind-driven currents control the hydrodynamic balance of the lagoon by provoking lagoon outflows and Mediterranean seawater inflows, both of them overprinted by moderate tidal influences (i.e., tidal range of 0.40 m; Ladagnous and Le Bec, 1997). The lagoon hydrology is affected by freshwater inputs from two main karstic systems in its southwestern parts (i.e., the Font Dame and Font Estramar discharges) that are responsible for 98% of freshwater inputs into the lagoon, with fluxes of 8400 and 12,500 $m^3.h^{-1}$ respectively (Ladagnous and Le Bec, 1997; Stieglitz et al., 2013). Altogether, these parameters produce spatial and seasonal salinity variations in the lagoon ranging from 20 to 30 (Bejannin et al., 2017). Finally, it is worth noting that the Font Estramar and Font Dame karstic resurgences belong to the well-studied Corbières karstic domain, which is composed of Jurassic and Cretaceous dolomites, limestones and marls covering Triassic evaporites. It constitutes one of the biggest karstic network from Europe (Ladouche and Dörfliger, 2004; Aunay et al., 2003).

2.3. Banyuls-sur-Mer transect (South of France)

The Banyuls-sur-Mer (BSM) coast is located on the most eastern part of the Pyrenean reliefs (i.e., Massif des Albères) where the substratum lithology is dominated by metamorphic and crystalline continental rocks of Cambrian age (Fig. 1C; Got and Stanley, 1974). Influenced by a Mediterranean climate (with annual rainfalls of 575 $mm.yr^{-1}$), this open rocky coast displays no major freshwater input apart from the Baillaury river (mean flow rate of 0.2 $m^3.s^{-1}$) that can show very important freshwater discharges in rainy season (up to 165 $m^3.s^{-1}$ during

floods). BSM is characterized by a low tidal range with an average of 0.40 m. From the BSM beach, a 25 km-long W-E transect was carried out at sea up to the Lacaze-Duthiers (LD) Canyon in order to collect surface and bottom seawater (i.e., up to 450 m water depth in the canyon). The LD canyon is one of the numerous canyons of the Gulf of Lion, with a strong bottom current and high-suspended sediment supplies from seasonal gravity flow events (Heussner et al., 2006). The salinity and temperature of seawater is regularly measured by the Oceanological Observatory of Banyuls-sur-Mer in the framework of the SOMLIT program, both at SOLA (in the Banyuls Bay) and MOLA (offshore above the canyon) buoys. The surface seawater salinity varies between 34 and 38 whereas bottom seawater varies between 37 and 39. The salinity at 450 m depth fluctuates slightly around 38 (Durrieu de Madron et al., 2013).

2.4. The Tatakoto lagoon (French Polynesia, Pacific Ocean)

The Tatakoto lagoon is a semi-enclosed hemipelagic carbonate platform located in the eastern Tuamotu Archipelago in the Pacific Ocean (Fig. 1D). This archipelago spreads over 1600 km and is characterized by 77 atolls whose reef-barrier caps volcanic basement dated between 44 and 37.5 Myrs (Rougerie, 1995; Pirazzoli, 1998). The climate is tropical and dominated by a seasonal rainfall regime of 50 to 100 mm monthly from June to October and 100 to 200 mm monthly from November to May (Andréfouët et al., 2001). The Tatakoto atoll is about 12 km long and 3 km large, and the lagoon covers an area of ~12 km^2 (Gilbert et al., 2006) with a maximal water depth measured at 18 m (C. Brahmi, personal communication). The atoll rim is closed in the northern part and semi-opened in the southern part. Water exchanges between the lagoon and the ocean occur through several shallow inlets located on the southern side and seawater mixing depends on tide, wave and wind regimes (Andréfouët et al., 2012; Van Wynsberge et al., 2017). The seawater exchanges between the atoll inner parts and ocean are weak owing to the low tidal range (with a maximum amplitude of 0.2 to 0.35 m). In the Tuamotu islands, water percolation through the reef porosity (i.e., 30 to 60%) is linked to erosion-dissolution and karstification processes (Rougerie, 1995). Note that intermediate oceanic seawaters flow through the porosity network of the barrier by *endo*-upwelling convection, leading to the formation of interstitial waters representing ~50% of the barrier volume (Rougerie, 1995). Thus, the renewal of the atoll seawater volume requires several weeks up to several months depending on the morphology, the porosity of the barrier and the seasonal hydroclimatic conditions (Rougerie, 1995; Tartinville et al., 1997). Combined with evaporation rates of 0.5 to 1 $m.yr^{-1}$, all these processes impose lagoonal salinities fluctuating between 30 and 44 (Rougerie et al., 1984).

3. Material and methods

3.1. Water and mollusc sampling

In order to check the spatial variability of seawater $^{87}Sr/^{86}Sr$ ratios in coastal domains, 38 water samples and 18 carbonate shells of living molluscs (i.e., bivalves and gastropods) were collected at the four studied sites (Fig. 1; Tables 1 and 2). When and where possible, we collected both the surface and bottom seawater together with molluscs for a given position.

In the field, the water samples were collected in 1 l LDPE bottles previously cleaned with ultrapure 1N HCl, filtered in situ using 60 ml sterile plastic syringes with 0.2 μm Millipore filters (in-situ ultrafiltration was not possible in the field due to logistic constraints). All water samples are void of any microparticles usually found as suspended particle matter in the natural environment. In-situ filtration during water collection can significantly reduce microparticle-water interaction and therefore improve the characterization of the dissolved chemical pool (Cotte et al., 2015). They were stored at 4 °C in a cold room prior to any sample preparation in a clean laboratory for chemical analyses. In the Oualidia and Salses-Leucate coastal lagoons, the water samples were collected (in April and June 2018, respectively) from 0 to 2 m water depth by diving from a chartered semi-rigid boat at various locations from landward parts affected by groundwater discharges to marine inlets and beaches facing the open marine realm (Fig. 1A, B). Bivalve shells from the Oualidia lagoon include *Crassostrea gigas*, *Venerupis decussata*, and *Solen marginatus*, whereas those from Salses-Leucate correspond to *Mytilus galloprovincialis*, *Crassostrea* sp., and *Glycymeris glycymeris*. Note that it was possible to collect directly the groundwaters from the Font-Dame and Font-Estramar resurgences at Salses-Leucate lagoon. However, the groundwater discharges of the Oualidia lagoon are mostly submerged, sparse and difficult to locate without in-situ chemical and salinity monitoring (Fakir et al., 2019).

In the Tatakoto lagoon, two water samples and four *Tridacna maxima* shells have been all collected at 2 m depth from two locations: one close to the southern oceanic inlets and the other one in the northern quiescent part of the inner lagoon (Fig. 1D).

Finally, a proximal-distal transect between the BSM beach and LD Canyon was carried out in May 2018 using the oceanographic research ship Néréis II (Station research vessel of Banyuls-sur-Mer, TGIR French Oceanographic Fleet; Fig. 1C), with a regular sampling of both surface and bottom seawater (down to 450 m below sea level; with ex-situ water filtration) as well as shells (encompassing unidentified bivalves fragments, *Mytilus galloprovincialis* and one gastropod *Turritella communis*) using a sediment grab.

3.2. Chemical composition of waters and shells

All the analytical steps have been performed at the Geosciences Environnement Toulouse laboratory (France). First, the collected water samples were prepared in a clean laboratory for chemical analyses including major (Mg, Ca, Na, K) and trace (Sr) element concentrations as well as the anion (Cl, SO_4, Br) concentrations. Anions were determined by anionic chromatography calibrated with an IAPSO seawater standard solution that was diluted at different proportion with MQ-H_2O to cover the entire range of anion concentrations. The IAPSO solution is a seawater standard solution provided by OSIL (UK) and certified for its salinity. The major and trace elements (Ca, Mg, Na, K, Sr) concentrations were measured using an Inductively Coupled Plasma optical emission spectrophotometer (ICP-OES), Horiba Jobin Yvon Ultima 2. We used the IAPSO standard solution to calibrate the instruments (Besson et al., 2014).

The shell samples were abraded, cleaned with Suprapur® acetic acid, and rinsed in Milli-Q water to remove external impurities and periostracum. Smaller bivalves and gastropods samples were entirely crushed with an agate mortar to get a powder, whereas only the inner parts of the umbo (recording the entire life span of the organism) were microdrilled for larger bivalve shells. For major element concentrations of molluscs, we dissolved 10 mg of sample using double-distilled 1 M HNO_3 on a hot plate at 70 °C. After evaporation of the solution to dryness, it was dissolved in 2N HNO_3 and diluted 100 times with MQ-H_2O. The solution was then analyzed for Mg, Ca, and Sr concentrations using an ICP-OES. The dissolved elements are 1000 times more concentrated than their limit of detection (> 10 ppb). The ICP-OES instrument was calibrated using a matrix-matched synthetic standard. One of the calibration solutions was measured every 5 samples to check the instrumental drift during the course of the analyses. The measurements achieved a precision of 2.3% for Ca, 2.8% for Mg and 4.3% for Sr, in line with previous analytical precision on Ca-rich sample (Chavagnac et al., 2005).

3.3. Sr isotopic compositions

To determine the $^{87}Sr/^{86}Sr$ ratios, 1 ml of filtered water was transferred to a Savillex beaker prior to evaporation on a hot plate at 70 °C and

Table 1
$^{87}Sr/^{86}Sr$ and elemental concentrations of surface and bottom seawater from the studied sites.

Studied site	Sample	Water type (environment)	Water depth (mbsl)	Salinity	Ca mmol·l^{-1}	Mg mmol·l^{-1}	Na mmol·l^{-1}	K mmol·l^{-1}	SO_4 mmol·l^{-1}	Cl mmol·l^{-1}	Li μmol·l^{-1}	Br μmol·l^{-1}	Sr μmol·l^{-1}	$^{87}Sr/^{86}Sr$	2 std dev
Oualidia lagoon, Morocco	OU2	Atlantic seawater (beach)	0	37.2	11.47	59.23	567	9.97	30.51	581	48.72	865	73.0	0.709158	0.000004
	OU3	Lagoon water	0	28.7	12.08	47.85	453	7.83	26.33	448	36.26	667	74.4	0.708772	0.000004
	OU4	Lagoon water	0	26.0	11.99	42.47	399	6.96	25.01	406	31.53	597	74.6	0.708682	0.000004
	OU5	Lagoon water	0	18.5	11.84	30.88	276	4.85	20.92	289	23.17	420	72.2	0.708389	0.000004
	OU6	Lagoon water (close to the dam)	0	9.9	12.53	18.48	138	2.60	17.19	155	15.85	223	78.4	0.709156	0.000004
	OU7	Lagoon water (salt marsh)	0	40.6	17.09	71.32	653	11.55	38.13	634	54.53	1012	96.4	0.708738	0.000004
	OU8	Lagoon water (salt marsh)	0	48.2	20.49	92.47	753	15.36	48.51	753	80.57	1302	116.8	0.708736	0.000004
	OU9	Lagoon water	0	33.1	11.83	53.14	535	9.37	28.53	517	42.62	768	73.0	0.708989	0.000004
	OU10	Lagoon water	0	37.1	11.16	57.34	586	10.08	30.49	580	47.44	862	71.7	0.709168	0.000004
	OU11	Lagoon water	0	36.6	11.38	58.32	580	10.09	30.10	571	45.66	862	69.5	0.709165	0.000004
	OU12	Lagoon water	0	34.1	11.60	53.28	542	9.34	29.12	532	43.26	786	73.0	0.709022	0.000004
	OU13	Lagoon water	0	32.9	11.61	52.32	524	9.04	28.53	514	41.77	762	73.0	0.708967	0.000004
	OU14	Lagoon water	0	37.0	11.54	58.44	591	10.41	30.38	578	48.10	856	72.1	0.709162	0.000004
	OU15	Lagoon water	3	37.1	11.34	57.23	598	10.20	30.47	579	49.57	855	71.8	0.709164	0.000004
	OU16	Lagoon water	3	36.8	11.51	57.00	601	10.22	30.26	575	46.99	846	71.7	0.709152	0.000004
	OU17	Lagoon water	3	36.9	11.76	58.65	600	10.26	30.35	576	47.06	833	73.1	0.709159	0.000004
Salses-Leucate lagoon, France	LEU9	Mediterranean seawater (beach)	0	37.6	11.95	61.04	598	10.71	30.82	586	46.71	871	69.7	0.709170	0.000004
	LEU1	Lagoon water (mussel farm)	0.5	36.3	11.31	57.40	577	10.24	30.00	567	45.72	848	71.0	0.709164	0.000004
	LEU2	Lagoon water	0	36.1	11.50	57.25	579	10.39	29.71	563	46.54	849	69.7	0.709170	0.000004
	LEU3	Lagoon water	0	37.4	11.69	59.52	593	10.61	30.75	584	47.81	880	71.9	0.709161	0.000004
	LEU4	Lagoon water	0	34.0	11.82	56.83	576	9.95	28.17	531	42.88	786	69.8	0.709155	0.000004
	LEU5	Lagoon water	1.5	32.8	11.61	54.76	553	9.65	27.25	512	39.48	762	69.8	0.709169	0.000004
	LEU6	Lagoon water	0	32.5	11.85	54.75	556	9.67	26.97	507	41.36	746	66.7	0.709132	0.000004
	LEU7	Lagoon water	0	11.8	6.82	20.23	186	3.25	10.93	185	11.42	235	34.2	0.708903	0.000004
	LEU8	Lagoon water	2.5	27.1	10.33	44.94	454	7.73	22.79	422	30.55	618	62.3	0.709109	0.000004
	LEU11	Groundwater (Font Estramar discharge)	0	4.3	3.49	6.83	39	1.07	4.52	67	4.78	86	19.3	0.708401	0.000004
	LEU10	Groundwater (Font Dame discharge)	0	2.7	3.71	4.37	9	0.62	3.78	42	2.77	51	17.2	0.708618	0.000004
Banyuls-sur-Mer transect, France	BA1	Mediterranean seawater (open shelf)	22	38.1	12.15	60.73	622	10.57	31.29	595	45.09	894	73.1	0.709174	0.000004
	BA2	Mediterranean seawater (open shelf)	0	37.8	11.76	58.50	602	10.25	30.94	590	48.45	884	72.7	0.709167	0.000004
	BA3	Mediterranean seawater (open shelf)	40	38.4	11.60	59.69	588	10.46	31.49	600	47.76	913	74.5	0.709167	0.000005
	BA4	Mediterranean seawater (open shelf)	0	38.0	11.24	59.91	586	10.33	31.14	593	45.55	893	74.0	0.709173	0.000004
	BA5	Mediterranean seawater (open shelf)	140	38.6	11.43	59.52	579	10.40	31.66	603	50.66	900	73.9	0.709170	0.000004
	BA6	Mediterranean seawater (open shelf)	0	38.5	11.75	60.35	594	10.60	31.58	601	49.11	914	73.6	0.709176	0.000004
	BA7	Mediterranean seawater (open shelf)	510	38.9	12.17	63.26	615	11.19	31.91	608	49.62	904	72.4	0.709166	0.000004
	BA8	Mediterranean seawater (open shelf)	0	38.6	11.71	60.22	612	10.75	31.64	603	47.38	911	72.2	0.709174	0.000004
	BA9	Mediterranean seawater (beach)	0	26.4	8.27	42.21	432	7.40	21.87	412	30.34	605	59.3	0.709206	0.000004
Tatakoto lagoon, French Polynesia	T1	Lagoon water	2	37.6	11.73	61.81	599	10.84	30.75	587	50.84	889	72.4	0.709165	0.000004
	T2	Lagoon water	2	37.5	12.02	62.77	619	11.16	30.75	585	50.03	887	73.5	0.709171	0.000004
Global ocean	–	Oligotrophic Oceanic water (OOW)	–	34.9	10.28	54.10	468	10.21	28.30	545	24.6	873	88.0	0.709172	0.000023

the residue was dissolved in 0.5 ml of 2N HNO_3. For the shells, ~10 mg of powder was dissolved in a Savillex beaker with 2N HNO_3 on a hot plate at 70 °C and centrifuged to exclude any residual fraction. Strontium was isolated from resulting solutions using Eichrom Sr-Spec column, a conventional elution protocol (Pin et al., 2014). The isolated Sr fraction was dissolved with 2 µl of 0.5N H_3PO_4 solution of which 1 µl was loaded on a tungsten filament together with 1 µl of Tantalum activator. The Sr isotopic composition was measured using a Thermo Triton Plus thermal ionization mass spectrometer. The $^{87}Sr/^{86}Sr$ ratios were defined as the average of 150 measurements of ion intensities following the static multi-collection mode normalized to $^{86}Sr/^{88}Sr = 0.1194$. During the measurement period, the standard NBS 987 gave $^{87}Sr/^{86}Sr$ ratios of 0.710280 ± 0.000003 (2SD, standard deviations, $n = 5$). Over the 2018 year, the measured NBS 987 isotopic compositions provided an external reproducibility of 0.710280 ± 0.000012 (2SD; $n = 26$). Regarding international seawater standards (IAPSO and NASS-6), they were processed in the clean room laboratory in the same manner as for our samples for Sr chemical isolation. The external $^{87}Sr/^{86}Sr$ reproducibilities are 0.709179 ± 0.000007 (2SD; $n = 7$) for IAPSO and 0.709180 ± 0.000014 (2SD; n = 5) for NASS-6, which agree very well with their certified values of 0.709179 ± 0.000005 and 0.709173 ± 0.000018, respectively (Neymark et al., 2014).

For the bivalve shells, we analyzed at repeated times the Sr isotopic composition of the JCt-1 international standard (a giant clam *Tridacna gigas*) treated in the clean room as for our shell samples. The measurements provide an external reproducibility of 0.709176 ± 0.000015 (2SD; $n = 11$) for a certified value of 0.709150 ± 0.000050 (Ohno and Hirata, 2007). For comparison purposes between the $^{87}Sr/^{86}Sr$ ratios reported in this study and the literature data (Tables 1 and 2), the $^{87}Sr/^{86}Sr$ ratios were corrected by adjusting the measured NBS 987 standard reference value to a certified value of 0.710250.

3.4. Geochemical and Sr isotopic database

To better depict potential influences of groundwater in the Oualidia and Salses-Leucate lagoons, we compiled a dataset of published major and trace elements concentrations (Ca, Mg, Na, K, SO_4, Cl, Br and Sr) and $^{87}Sr/^{86}Sr$ ratios corresponding to groundwaters representative of Moroccan aquifers ($n = 112$) and the Corbières aquifer in France ($n = 141$) (see Supplementary data). In order to analyze the geochemical patterns at a worldwide scale, we also compiled a dataset gathering 522 $^{87}Sr/^{86}Sr$ ratios and 571 Sr concentrations representative of OOW and shelf waters (SW) (see Supplementary data). As for our samples, all $^{87}Sr/^{86}Sr$ ratios reported in the literature were corrected by adjusting their respective NBS 987 standard reference value to a certified value of 0.710250.

4. Results

4.1. $^{87}Sr/^{86}Sr$ of waters and shells

The $^{87}Sr/^{86}Sr$ ratios of lagoon waters and shells are reported in Tables 1 and 2. First, at the Tatakoto lagoon, waters and shells exhibit $^{87}Sr/^{86}Sr$ ratios at 0.709165–0.709171 and 0.709164–0.709182, respectively, which are both indistinguishable from the average OOW value of 0.709172 (Figs. 1D and 2A). Similarly, seawater collected at different water depths along the BSM transect presents a slight dispersion ranging from 0.709166 to 0.709176, but still around the OOW value (Fig. 1C). The only sample deviating from this world value is BA9 collected in the Banyuls-sur-Mer beach: it presents a low salinity of 20 and a $^{87}Sr/^{86}Sr$ ratio of 0.709206 slightly more radiogenic than the OOW range (Fig. 2A). Molluscs from the BSM transect present Sr isotope compositions between 0.709164 and 0.709175, all identical to those of surrounding water (Fig. 2A).

In the Salses-Leucate lagoon, the seawater $^{87}Sr/^{86}Sr$ ratios ranging from 0.709155 and 0.709170 are typical of the OOW variability range. Contrastingly, three water samples (LEU 6, 7, 8) from the western part of the lagoon display values ranging from 0.708903 to 0.709132, that is below the OOW variability range. However, these values are trending towards the signatures of the Font Dame and Font Estramar discharges (LEU 10, 11) at 0.708618 and 0.708401, respectively (Fig. 1B). Again, these relatively low isotopic ratios occur along a salinity drop from 37.4 to 11.8 (Fig. 2A). In this context, the least radiogenic Sr isotope composition is measured for the bottom water sample (LEU 8) compared to the surface one (LEU 7). Mollusc shells sampled in this lagoon present $^{87}Sr/^{86}Sr$ ratios varying between 0.709125 and 0.709177, being either similar to or less radiogenic than the OOW value (Table 2; Fig. 2A). Finally, it is worth mentioning that our isotopic results are within the variability of those obtained for local seawater and groundwater sampled in 1998, 2002 and 2012 by Ladouche et al. (2000), Ladouche and Dörfliger (2004) and Petelet-Giraud et al. (2016).

At the Oualidia lagoon, both seawater and mollusc shells display highly variable $^{87}Sr/^{86}Sr$ ratios ranging from 0.708389 to 0.709168 and 0.708939 to 0.709179, respectively (Tables 1 and 2; Figs. 1A and 2A). Downstream, close to Atlantic seawater incursions by the inlets, the $^{87}Sr/^{86}Sr$ ratios of waters stay within the OOW variability range, while they are less and less radiogenic upstream between the OU12 and OU8 locations (Table 1, Figs. 1 and 2). As in Salses-Leucate and Banyuls bay, this isotopic shift is accompanied by a salinity decrease from 37.2 to 9.9 (Fig. 2A). However, the water sample OU6 deviates from this general tendency: although close to the northern dam, it records a $^{87}Sr/^{86}Sr$ ratio typical of OOW values (Fig. 1A). Finally, we note that most bivalve shells record $^{87}Sr/^{86}Sr$ ratios typical of OOW values but different of surrounding water values (Fig. 2A, Table 2). Only the O4 sample (i.e., an endobenthic *Solen marginatus*) fits the general $^{87}Sr/^{86}Sr$ decrease of lagoonal waters.

4.2. Geochemical composition of waters

All concentrations of anions and major and trace elements measured on the collected waters are reported in Table 1. In order to determine the origin of the waters influencing the $^{87}Sr/^{86}Sr$ of local seawater, only the most discriminating elements such as Sr, SO_4, and Ca are used here, plotted as function of salinity, and compared to published values of regional groundwater aquifers (Fig. 2).

In the Pacific Tatakoto atoll, seawater has salinity and $^{87}Sr/^{86}Sr$ ratios typical of OOW while they display lower Sr contents (~73.5 µmol·l^{-1} against 88 µmol·l^{-1} for OOW; Millero et al., 2008). Slight enrichments in SO_4 (30.8 mmol·l^{-1}) and Ca (10.3 mmol·l^{-1}) are recorded but still within the OOW variability range. These geochemical characteristics also concern most water samples from the Mediterranean BSM transect, whatever the water depth, with respective Sr, SO_4 and Ca concentrations of 72.2–74.5 µmol·l^{-1}, 30.9–32 mmol·l^{-1}, and 11.2–12.2 mmol·l^{-1}. The only exception is the beach sample BA9 which low salinity value of 26.4 is mirrored by low element concentrations compared to those of OOW (Table 1 and Fig. 2).

Euhaline waters from the inner parts of the Oualidia and Salses-Leucate lagoons are also characterized by similar depletions in Sr and slight enrichments in Ca and SO_4 when compared to the areas described above. However, the element concentrations evolve differently in Salses Leucate and Oualidia while the salinity and $^{87}Sr/^{86}Sr$ are decreasing landward. In Salses-Leucate, the Sr, SO_4 and Ca contents are dropping down to 17.2 µmol·l^{-1}, 3.8 mmol·l^{-1} and 3.5 mmol·l^{-1}, respectively, close to the Font Dame and Font Estramar discharges. In agreement with previous analyses of Ladouche et al. (2000) and Ladouche and Dörfliger (2004), these low concentrations are similar to those prevailing at the scale of the regional aquifer and the covariations between salinity, $^{87}Sr/^{86}Sr$ ratio and these elemental concentrations are consistent with freshwater-seawater binary mixing. In Oualidia, the salinity and $^{87}Sr/^{86}Sr$ decrease while Sr and Ca concentrations remain relatively high and stable (i.e., ~73 µmol·l^{-1} and ~11.5 mmol·l^{-1}, respectively), except in the salt marshes where values are much higher (i.e., 117 µmol·l^{-1} and

Table 2
$^{87}Sr/^{86}Sr$ and elemental concentrations of studied shells.

Studied site	Shell sample	Corresponding water sample	Species	Ca ($mg{\cdot}g^{-1}$)	Mg ($\mu g{\cdot}g^{-1}$)	Sr ($\mu g{\cdot}g^{-1}$)	$^{87}Sr/^{86}Sr$	2 std dev
Oualidia lagoon, Morocco	O 1	OU 3	*Crassostrea gigas*	370	3888	1091	0.709174	0.000004
	O 2	OU 9	*Crassostrea gigas*	371	1361	575	0.709173	0.000004
	O 3	OU 12	*Venerupis decussata*	334	118	1689	0.709179	0.000004
	O 4	OU 9	*Solen marginatus*	361	268	2797	0.708939	0.000004
Salses-Leucate lagoon, France	S1	LEU 9	*Mytilus galloprovincialis*	354	483	746	0.709143	0.000004
	S2	LEU 2	*Glycymeris glycymeris*	*na*	*na*	*na*	0.709125	0.000004
	S3	LEU 3	*Crassostrea* sp.	*na*	*na*	*na*	0.709136	0.000004
	S4	LEU 4	*Mytilus galloprovincialis*	364	945	969	0.709174	0.000004
	S5	LEU 5	*Mytilus galloprovincialis*	378	1291	932	0.709158	0.000004
	S6	LEU 6	*Mytilus galloprovincialis*	349	135	1511	0.709177	0.000004
Banyuls-sur-Mer, France	Bs 1	B 1	Bivalve fragments	360	792	1513	0.709174	0.000004
	Bs 2	B 3	*Turritella communis*	362	413	1645	0.709173	0.000004
	Bs 3	B 5	Bivalve fragments	*na*	*na*	*na*	0.709165	0.000004
	Bs 4	B 7	Bivalve fragments	381	11,554	2214	0.709171	0.000004
Tatakoto lagoon, French Polynesia	Ts 1	T1	*Tridacna maxima*	*na*	*na*	*na*	0.709178	0.000004
	Ts 2	T1	*Tridacna maxima*	365	810	1587	0.709164	0.000004
	Ts 3	T2	*Tridacna maxima*	*na*	*na*	*na*	0.709182	0.000004
	Ts 4	T2	*Tridacna maxima*	357	205	1644	0.709172	0.000004

20.5 $mmol{\cdot}l^{-1}$, respectively). Contrastingly the SO_4 contents decrease with salinity but towards a brackish groundwater end-member with anomalously high SO_4 concentrations (~15 $mmol{\cdot}l^{-1}$). Oualidia geochemical patterns are compatible with the high Sr, SO_4, and Ca contents recorded in some Moroccan groundwater aquifers, and especially those related to the dissolution of Mesozoic carbonate and gypsum levels (see Supplementary data).

5. Discussion

Studies with high precision $^{87}Sr/^{86}Sr$ ratio have confirmed that modern oligotrophic oceanic waters (OOW) without terrestrial inputs have a quite homogeneous $^{87}Sr/^{86}Sr$ ratio of 0.709172 with a variability range of only ± 0.000023 at the global scale (Andersson et al., 1992; Winter et al., 1997; Huang and You, 2007; Huang et al., 2011; Mokadem et al., 2015; Pearce et al., 2015; Trezzi et al., 2017; El Meknassi et al., 2018). This homogeneity is yet regularly challenged for shelf domains as more radiogenic or unradiogenic terrestrial Sr supplies from rivers or SGD can introduce a dispersion to the measured $^{87}Sr/^{86}Sr$ of 6×10^{-3} or more (El Meknassi et al., 2018) (Fig. 3A). Nevertheless, this estimate mainly concerns estuaries, fjords, bays or epeiric seas supplied by major river systems for which data are relatively abundant (e.g., Ingram and Sloan, 1992; Jørgensen and Banoeng-Yakubo, 2001; Négrel et al., 2005; Jørgensen et al., 2008; Wang and You, 2013; Eissa et al., 2016; Casse et al., 2019). In this context, new Sr isotope data from open shelves and coastal lagoons become crucial to get a better view of processes at play in neritic domains, especially those locations deprived of river and delta influences. These areas represent 40–50% of the world coasts (Dürr et al., 2011).

5.1. Seawater $^{87}Sr/^{86}Sr$ ratios in oceanic and open shelf domains

With an average Sr concentration of 87.4 $\mu mol{\cdot}l^{-1}$ (de Villiers, 1999), oceanic inflows are the main dissolved Sr source to shelf domains and hemipelagic carbonate platforms. If these oceanic inflows remain constant through time and sufficiently mixed with coastal waters, locally homogeneous seawater and shell $^{87}Sr/^{86}Sr$ ratios close to OOW value of 0.709172 would be expected, even in semi-enclosed areas. Far from any terrestrial influence, this is clearly illustrated in the isolated Tatakoto atoll where Pacific seawater is the main dissolved Sr source, conveyed to the shelf by endo-upwelling circulation through the reef porosity followed by surface currents (Rougerie, 1995). Rainwater infiltrations responsible for fossil reef karstification and possibly volcanic basement weathering are important in the Tuamotu archipelago (Guy et al., 1992; Waljeski, 2003). However, unradiogenic Sr supplied from local SGD appears too limited to modify the OOW $^{87}Sr/^{86}Sr$ ratio of studied waters and giant clams in this Pacific atoll (Tables 1 and 2).

In the nearshore domain exemplified by the 25 km-long Mediterranean BSM transect, seawater and shells from surface down to ~450 mbsl display a homogeneous $^{87}Sr/^{86}Sr$ ratio with an average value at 0.709171 ± 0.000008 (2SD; n = 12) indistinguishable from the OOW value (El Meknassi et al., 2018). In the absence of noticeable dissolved Sr inputs from rivers or SGD, this implies that the Sr inputs resulting from boundary-exchange processes acting at the sediment-water interface are not sufficient to modify the seawater $^{87}Sr/^{86}Sr$ ratio in these well-mixed open shelf conditions. This contrasts with estuaries or deltas where dissolved Sr released from suspended particulate dissolution may alter the $^{87}Sr/^{86}Sr$ of seawater in a few months (Jones et al., 2014; Jeandel and Oelkers, 2015). Moreover, the shell data from the BSM transect indicate that the Sr isotope composition of local seawater did not significantly change over life spans of 2 to 3 years. However, these results do not preclude caution in paleoenvironmental interpretation of shell $^{87}Sr/^{86}Sr$ ratios from the BSM coast and more generally nearshore contexts. This is because SGD influence may strongly vary in time and space along continental margins (Burnett et al., 2006), and especially in the Gulf of Lion where SGD are very important (Rodellas et al., 2015).

In the foreshore zone, additional factors can impact the seawater $^{87}Sr/^{86}Sr$ ratios. This is illustrated by the BA9 water sample collected on the BSM sandy beach, characterized by a more radiogenic value and a much lower salinity (i.e., 26.4) than the other samples. We propose that this more radiogenic Sr value can be related to three factors typical of intertidal zones: 1) mixing with rainwater whose regional $^{87}Sr/^{86}Sr$ ratios range from 0.708993 to 0.709225 (Khaska et al., 2013), 2) sparse groundwater supplies whose Sr concentrations and radiogenic $^{87}Sr/^{86}Sr$ ratios mirror those from Quaternary, Pliocene, and Paleozoic aquifers (i.e., 0.708142 to 0.713782; Petelet et al., 1998; Petelet-Giraud et al., 2016), and 3) important dissolved Sr amounts released from suspended sandy particles continuously shaked in the surf zone (Kalnejais et al., 2010; Jeandel, 2016; Fabre et al., 2019) with particles $^{87}Sr/^{86}Sr$ ratios of 0.72232–0.72901 reflecting their metamorphic substratum (Brems et al., 2013). At broader scale, similar local processes can explain part of isotopic deviations reported in the rare water and shell data from open shelves (e.g., North Sea, Gulf of Guinea, South China Sea; Jørgensen and Banoeng-Yakubo, 2001; Jørgensen et al., 2008; Huang et al., 2011; El Meknassi et al., 2018) (Fig. 3). Moreover, marginal areas are transition zones where the oceanic mixing is not instantaneous and may take several years according to littoral currents, coastal

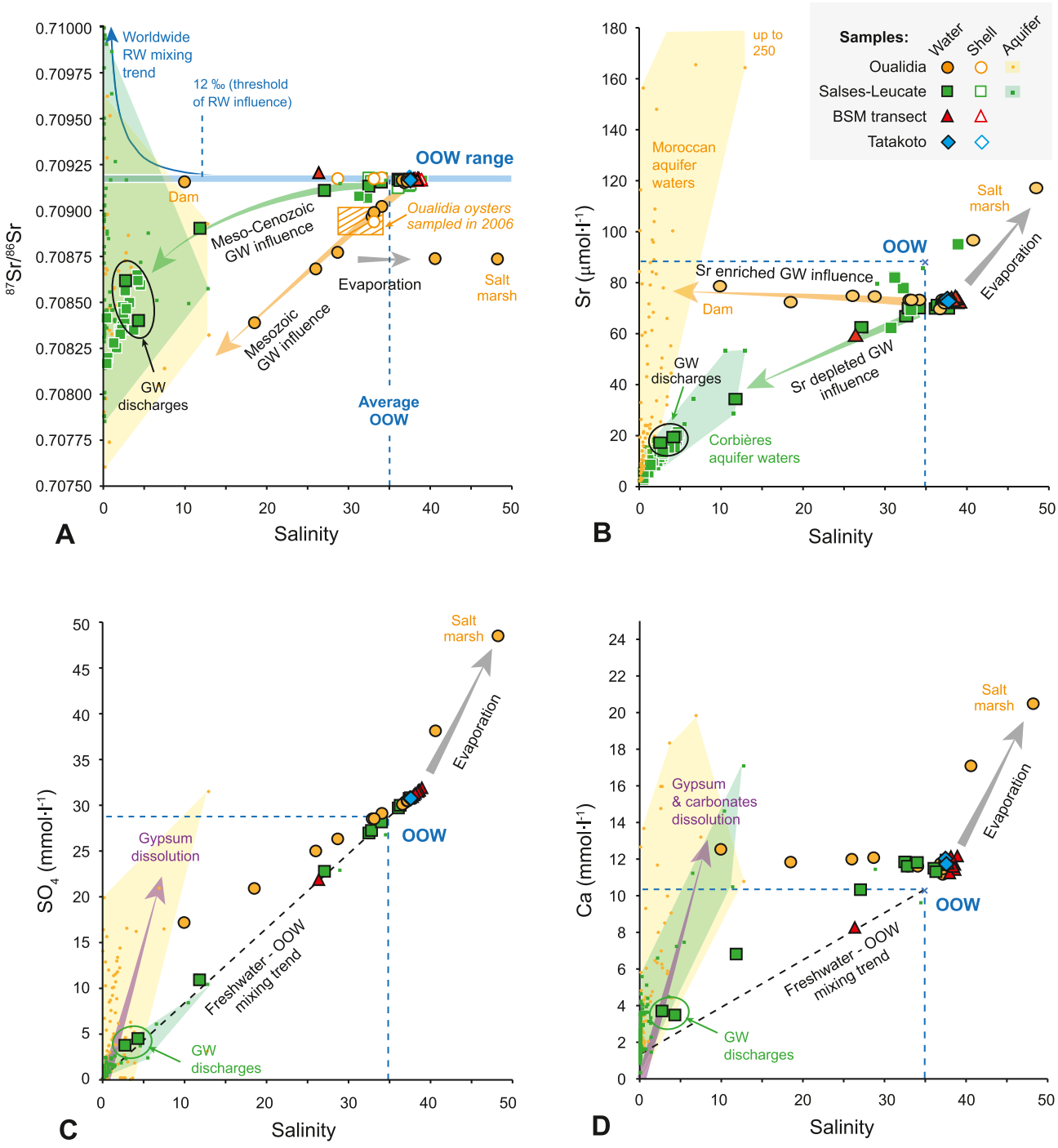

Fig. 2. $^{87}Sr/^{86}Sr$, Sr, Ca, and SO_4 concentrations of waters as a function of salinity. (A) For a same sampling position with a given salinity, the $^{87}Sr/^{86}Sr$ ratios of living mollusc shells are compared to $^{87}Sr/^{86}Sr$ ratios of ambient seawater as well as oysters sampled in 2006 by El Meknassi et al. (2018) (see Table 2 for samples correspondence). Additional literature data from Salses-Leucate (i.e., water sampled in 1998, 2002 and 2012) are represented by squares with white contours (Ladouche et al., 2000; Ladouche and Dörfliger, 2004; Petelet-Giraud et al., 2016). The isotopic range and the average elemental concentrations of OOW are from El Meknassi et al. (2018) and Millero et al., (2008), respectively. The salinity threshold defining significant influence of river water (RW) inputs on seawater $^{87}Sr/^{86}Sr$ ratios is from Bryant et al. (1995). Seawater and SGD values from the Oualidia and Salses-Leucates lagoons are compared to groundwater data available at the scale of Moroccan aquifers (Fakir et al., 2002; Kaid Rassou et al., 2005; Vinson et al., 2013; Fadili et al., 2015; Bouchaou et al., 2017) and the Corbières aquifer (southern France) (Petelet et al., 1998; Aquilina et al., 2002; Khaska et al., 2013; Petelet-Giraud et al., 2016). The analytical uncertainty bars are smaller than the width of symbols.

physiography, and shelf width (Liu et al., 2019) (Fig. 3B). Terrestrial dissolved Sr sources, even remote, could slightly impact local seawater $^{87}Sr/^{86}Sr$ ratios through cumulative effects. Thus, we advocate to reject any water and/or shell sample from beach or intertidal zones to infer modern or past OOW $^{87}Sr/^{86}Sr$ ratios.

5.2. $^{87}Sr/^{86}Sr$ patterns in restricted coastal domains

Supplementing sparse literature data (e.g., Müller et al., 1990b; Beck et al., 2013; Shao et al., 2018), the seawater and shell results from the Oualidia and Salses-Leucate lagoons show clear evidence that

restricted coastal domains, even without river influence, display very distinct $^{87}Sr/^{86}Sr$ ratios depending on the respective contribution of oceanic and SGD influxes (Fig. 1): 1) quite homogeneous OOW $^{87}Sr/^{86}Sr$ ratios except in close vicinity of SGD in Salses-Leucate; or 2) variable $^{87}Sr/^{86}Sr$ ratios ranging from OOW values at oceanic inlets to unradiogenic signals in the upstream parts of the Oualidia lagoon.

At broader scale, such Sr isotopic variabilities are also noticeable in epeiric seas. By compiling literature data and excluding the anomalous $^{87}Sr/^{86}Sr$ ratios (i.e., samples from beach or close to SGD) from Salses-Leucate, Banyuls-sur-Mer and Venice (Fig. 4A), we observe that seawater $^{87}Sr/^{86}Sr$ ratios are typical of OOW values in western Mediterranean Sea (i.e., Spain, France, Italy), whereas those from eastern domains like the Bannock Basin, Aegean Sea, Marmara Sea and Black Sea decrease down to 0.70912 (this study; Müller et al., 1990a; Clauer et al., 2000; Peckmann et al., 2001; Major et al., 2006; Beck et al., 2013; Petelet-Giraud et al., 2016; Trezzi et al., 2017; Teichert et al., 2018). Highlighted here for the first time, these regional $^{87}Sr/^{86}Sr$ variations are consistent with the isotopic variability of Mediterranean water masses illustrated by the Nd isotopes (Tachikawa et al., 2004). Whether local or supra-regional, spatial Sr isotopic variability is insightful to decipher the respective influence of SGD and oceanic inputs on the Sr isotope budget of semi-enclosed areas.

5.2.1. SGD influences

With relatively high Sr concentrations (i.e., 2.9 μmol·l^{-1}) compared to river waters (i.e., 0.5 to 1.2 μmol·l^{-1}), SGDs represent an unradiogenic Sr source that can account for 13–30% of the global Sr ocean budget (Basu et al., 2001; Krabbenhöft et al., 2010; Beck et al., 2013). Depending on their geochemical and hydrodynamical characteristics related to the age and nature of weathered rocks but also oceanic intrusions, meteoric recharges, and water residence time in the aquifers (e.g., Négrel et al., 2003; Fadili et al., 2015, 2016; Santoni et al., 2016), their impact on the elemental and isotopic Sr budgets are, however, highly variable in coastal domains (e.g., Beck et al., 2013; Trezzi et al., 2017; Shao et al., 2018). This is illustrated by comparing the Salses-Leucate and Oualidia lagoons.

In the Salses-Leucate lagoon, the low salinities and the low Sr, SO_4 and Ca concentrations of Font Estramar and Font Dame discharges show that these groundwaters are typical of meteoric freshwaters percolating through the local karstic aquifer (Fig. 2B, C, D). To better constrain the origin of the collected seawaters in the lagoon, we applied the isotopic mixing equation (Albarède, 1996) to geochemical data of regional end members presented in Table 3:

$$^{87}Sr/^{86}Sr_{(mix)} = \left(\frac{^{87}Sr/^{86}Sr_{(SW)} \times [Sr]_{(SW)} \times \%_{(SW)}}{[Sr]_{(mix)}}\right) + \left(\frac{^{87}Sr/^{86}Sr_{(GW)} \times [Sr]_{(GW)} \times \%_{(GW)}}{[Sr]_{(mix)}}\right),$$

where the mixed water $^{87}Sr/^{86}Sr_{(mix)}$ ratio varies between the local seawater (SW) and groundwater (GW) end members of given Sr concentrations and $^{87}Sr/^{86}Sr$ ratios, using mixed water Sr concentrations $[Sr]_{(mix)}$ calculated from cumulative 10% increments of GW to SW. The low $^{87}Sr/^{86}Sr$ ratios of Font Estramar and Font Dame discharges correspond to the lowest isotopic values of regional groundwaters and indicate a preferential dissolution of Jurassic and Cretaceous carbonates characterized by low $^{87}Sr/^{86}Sr$ ratios compared to the Pliocene and Quaternary silicates ones (Fig. 4A; Petelet et al., 1998; Ladouche et al., 2000; Aquilina et al., 2002; Ladouche and Dörfliger, 2004; Khaska et al., 2013; Petelet-Giraud et al., 2016). Given the low Sr concentrations of Jurassic and Cretaceous GW endmembers (Table 3; Fig. 2B), our results suggest that the SGD contribution to the lagoon Sr budget is broadly less than 15% despite local SGD fluxes exceeding 5.55 $m^3.s^{-1}$ (Fig. 4B; Ladagnous and Le Bec, 1997; Stieglitz et al., 2013).

In Oualidia, the dissolved Sr supply of SGD with much lower estimated fluxes of 0.2–1.6 $m^3.s^{-1}$ (Hilmi et al., 2005; Fakir et al., 2019) markedly modifies the seawater $^{87}Sr/^{86}Sr$ ratios within the upstream parts of the lagoon (Fig. 1A). These induced modifications can be related to considerable unradiogenic Sr inputs while maintaining constant dissolved Sr concentrations (Fig. 2A, B). Indeed, groundwaters from the Coastal Sahel of Oualidia exhibit variable but generally high Sr and Ca concentrations reaching 2 to 2.5 times the OOW value as well as high SO_4 amounts (Fakir et al., 2002; Hilmi et al., 2005; Fig. 2B, C, D). Even if oceanic water intrusions are not excluded in the coastal aquifer, specific Ca and SO_4 excesses indicate that the higher Sr contents would result from a preferential dissolution of Sr rich gypsum and celestite ($SrSO_4$) present in the Jurassic (and Cretaceous) limestones (Fakir et al., 2002; Hilmi et al., 2005; Kaid Rassou et al., 2005; Bouchaou et al., 2017). This hypothesis is in agreement with the unradiogenic $^{87}Sr/^{86}Sr$ ratio of lagoon waters (i.e., down to 0.708389) whose values are close to Jurassic aquifers of Morocco (Bouchaou et al., 2017; Vinson et al., 2013) (Fig. 4A). We estimate that groundwater fluxes dissolving Mesozoic sulphates spatially contribute from ~10 to 60% to the lagoon Sr budget, the higher Sr concentrations being related to high evaporation rates in the salt marshes (Fig. 4A). This Sr supply markedly varied in a few years, as shown by the isotopic scatter between local seawater and shell data at a same location or shells sampled in 2006 (El Meknassi et al., 2018) and 2018 (this study) (Fig. 2A). In contrast, the low $^{87}Sr/^{86}Sr$ ratio recorded by one endobenthic *Solen marginatus* suggests that groundwater influence remained more constant in the interstitial waters at 80 cm sediment depth (i.e., the maximal burial depth of this species) than in the lagoon waters.

With Sr concentrations up to 150 μmol·l^{-1} leading to local contributions of 60% to the Sr budget of lagoons, our results call to reconsider the importance of SGD in link to the dissolution of evaporitic deposits. In association with geothermal brines, their impact on the global ocean Sr budget still remains difficult to appraise but could be regionally important given the proximity of Messinian or Triassic evaporites such as those of Mediterranean margins (e.g., Stein et al., 2000). Considering their elevated Sr concentrations, it is also important to note that, contrarily to river-dominated contexts requiring considerable freshwater inputs to alter the coastal seawater $^{87}Sr/^{86}Sr$ ratio, SGD may quickly change this isotopic ratio in euhaline conditions. Indeed, most major world river systems produce measurable effects on seawater $^{87}Sr/^{86}Sr$ ratios when salinity drops below an average threshold of 12 (Bryant et al., 1995). Only ~15, 10 and 5% of the rivers have significant effects at salinities above 20, 25 and 30, respectively. In contrast, significant $^{87}Sr/^{86}Sr$ shifts occur at salinities close to 32–35 in the studied lagoons (Fig. 2A). From a paleoenvironmental point of view, these results call to be very careful in any paleosalinity deduction from coastal fossil $^{87}Sr/^{86}Sr$ data, especially since saline SGD are difficult to detect from biosedimentary facies or sismic data compared with freshwater SGD (Lecher and Mackey, 2018; Goff, 2019).

5.3. Influence of water mass restriction

From small lagoons to larger epeiric areas like Mediterranean and Baltic seas, oceanic mixing is essential to buffer regional SGD and river effects and maintain homogeneous seawater $^{87}Sr/^{86}Sr$ patterns typical of OOW. However, water mass restriction imposed by geographic barriers and water column stratification may be highly variable from one domain to another one. In lagoons, the local OOW renewal is more or less perennial and important depending on their leaky, choked or restricted geomorphology (Kjerfve and Magill, 1989; Umgiesser et al., 2014). Basically, restricted and leaky lagoons are parallel to the coast and present, respectively, few to numerous inlets that allow an efficient oceanic water turnover (e.g. every one to a few days) driven by tidal dynamics and/or wind forcing (Kjerfve and Magill, 1989; Umgiesser et al., 2014). These characteristics contribute therefore to homogenize seawater $^{87}Sr/^{86}Sr$ ratio close to the OOW one like in the Salses-Leucate or Venice lagoons (Beck et al., 2013) (Fig. 1). To the extreme, choked lagoons are characterized by only one or two oceanic inlets with a dominant wind forcing implying several days to several months for a total water renewal in the lagoon (Kjerfve and Magill, 1989; Umgiesser

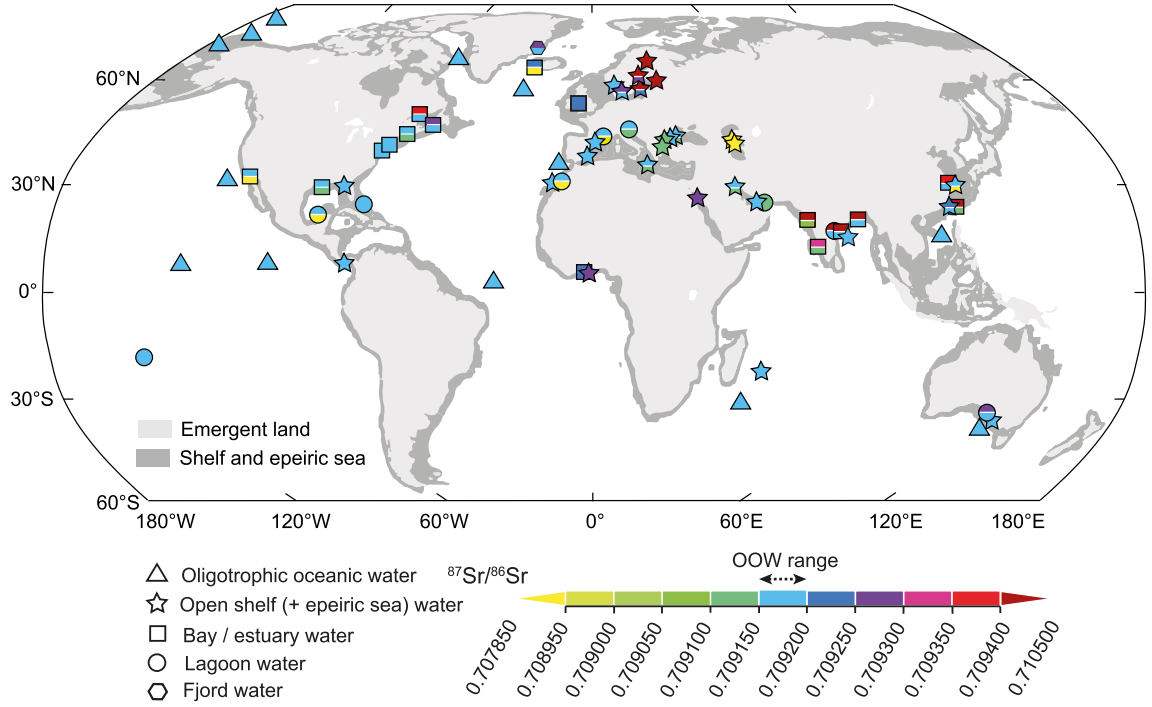

Fig. 3. (A) Worldwide distribution of published and new $^{87}Sr/^{86}Sr$ ratios for euhaline to brackish shelf waters and oligotrophic oceanic waters (OOW). If locally variable, the maximal and minimal $^{87}Sr/^{86}Sr$ ratios are represented by two colours. The OOW $^{87}Sr/^{86}Sr$ variability range is from El Meknassi et al. (2018). Literature data from Müller et al. (1990a, 1990b), Andersson et al. (1992), Ingram and Sloan (1992), Winter et al. (1997), Barker et al. (1998), Israelson and Buchardt (1999), Clauer et al. (2000), Ladouche et al. (2000), Jørgensen and Banoeng-Yakubo (2001), Peckmann et al. (2001), Wang et al. (2001), Négrel et al. (2005), Major et al. (2006), Huang and You (2007), Sharma et al. (2007), Xu and Marcantonio (2007), Jørgensen et al. (2008), Martin and Moore (2008), Huang et al. (2011), Patra et al. (2012), Rahaman and Singh (2012), Beck et al. (2013), Uddin et al. (2013), Wang and You (2013), Jones et al. (2014), Mokadem et al. (2015), Pearce et al. (2015), Eissa et al. (2016), Petelet-Giraud et al. (2016), Trezzi et al. (2017), Shao et al. (2018), Teichert et al. (2018), Casse et al. (2019), Danish et al. (2020).

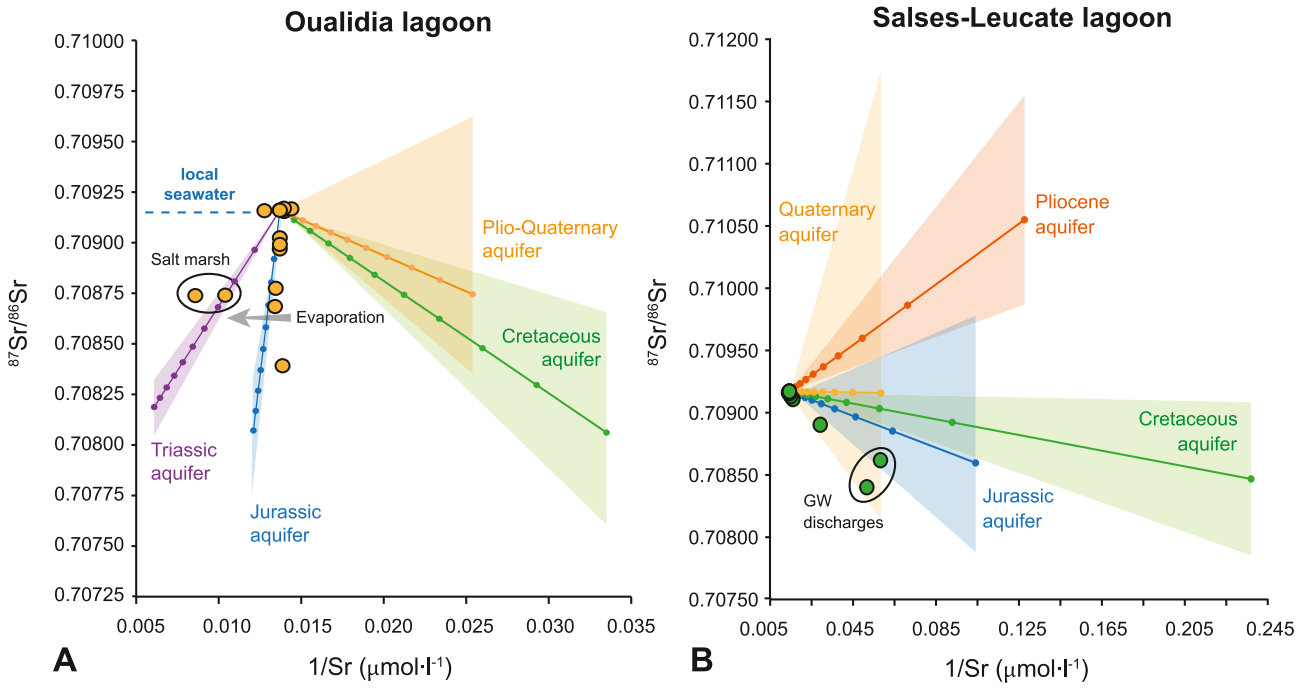

Fig. 4. $^{87}Sr/^{86}Sr$ vs. 1/Sr diagram for the Salses-Leucate (A) and Oualidia (B) lagoon waters. The data are compared to mixing trends between local seawater and different groundwaters (GW) from Moroccan (A) and Corbières aquifers (B). End members data are presented in Table 3. The mixing curves show the $^{87}Sr/^{86}Sr$ ratios for each 10% increment. The analytical uncertainty bars are smaller than the width of symbols.

Table 3
Average, minimal and maximal $^{87}Sr/^{86}Sr$ and Sr concentrations of seawater (SW) and groundwater (GW) end members used for mixing model of Fig. 4. GW data refer to waters percolating through different aquifers of the Corbières region (South France) and Morocco. As GW $^{87}Sr/^{86}Sr$ and Sr concentration data are missing or sparse, respectively, for the Coastal Sahel of Oualidia, we used GW data from Moroccan aquifers with host rocks of the same age and lithology (see Supplementary data).

Studied sites	End-members	Water type	Average Sr ($\mu mol \cdot l^{-1}$)	Minimal Sr ($\mu mol \cdot l^{-1}$)	Maximal Sr ($\mu mol \cdot l^{-1}$)	Average $^{87}Sr/^{86}Sr$	Minimal $^{87}Sr/^{86}Sr$	Maximal $^{87}Sr/^{86}Sr$	References
Morocco	Plio-Quaternary aquifer	GW	39.4	1.4	148.0	0.70874	0.70835	0.70962	Fakir et al., 2002; Kaid Rassou et al., 2005; Vinson et al., 2013
	Cretaceous aquifer	GW	29.8	3.0	116.4	0.70760	0.70760	0.70866	Fakir et al., 2002; Fadili et al., 2015; Bouchaou et al., 2017
	Jurassic aquifer	GW	82.7	36.3	249.9	0.70807	0.70774	0.70833	Kaid Rassou et al., 2005; Bouchaou et al., 2017
	Triassic aquifer	GW	164.9	164.4	165.5	0.70819	0.70805	0.70832	Bouchaou et al., 2017
	Atlantic water at Oualidia	SW	73.0	–	–	0.70916	–	–	*This study*
Corbières region, France	Quaternary aquifer	GW	17.1	1.8	85.5	0.70916	0.70814	0.71174	Petelet et al., 1998; Petelet-Giraud et al., 2016
	Pliocene aquifer	GW	7.8	4.7	15.2	0.71055	0.70987	0.71155	Petelet-Giraud et al., 2016
	Cretaceous aquifer	GW	4.2	1.2	9.0	0.70847	0.70753	0.70908	Khaska et al., 2013
	Jurassic aquifer	GW	9.6	0.3	53.0	0.70860	0.70788	0.70978	Aquilina et al., 2002; Petelet-Giraud et al., 2016
	Mediterranean water at Salses-Leucate	SW	69.7	–	–	0.70917	–	–	*This study*

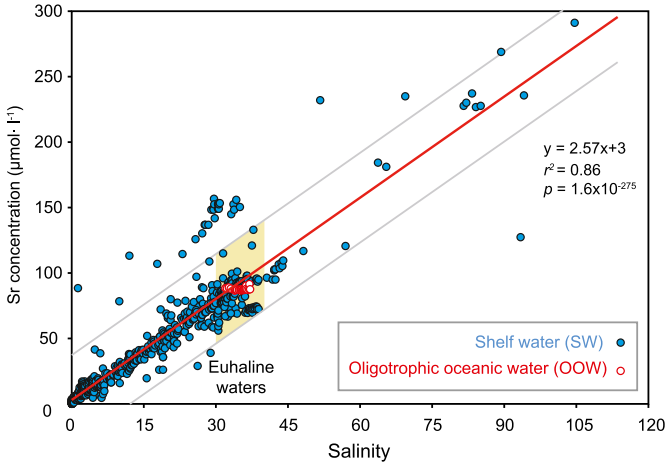

Fig. 5. Sr concentration of shelf (SW) and oligotrophic oceanic (OOW) waters as a function of salinity. The OOW data are from de Villiers (1999) and the SW data are compiled from various sources (see Supplementary data). The red and grey lines represent the linear regression and the 95% confidence interval of Sr concentration values, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

et al., 2014). This situation applies to the Oualidia lagoon, with the exception that tides are an important factor in the local hydrodynamics (Hilmi et al., 2017). Based on numerical modelling of tidal currents at Oualidia, the time necessary for full lagoon water renewal was estimated to be one or a few days in close vicinity of oceanic inlet increasing up to 15 to 30 days in the upstream channel and salt marshes, respectively (Hilmi et al., 2005, 2009, 2017). As a result, the duality and rhythmicity between SGD discharges and seawater tidal influxes explain the $^{87}Sr/^{86}Sr$ gradient observed in the Oualidia lagoon from the oceanic inlet towards the interior of the lagoon.

In epeiric seas, similar water mass restriction processes appear relevant to explain the variability of seawater $^{87}Sr/^{86}Sr$ ratios. The Mediterranean basin is very interesting in this respect as SGD with elevated Sr concentrations (5–12 $\mu mol \cdot l^{-1}$) and low $^{87}Sr/^{86}Sr$ ratios (i.e., 0.7078–0.7080) prevail all along the northern Mediterranean margin (Rodellas et al., 2015; Trezzi et al., 2017). However, only the eastern Mediterranean waters are significantly affected by these unradiogenic terrestrial Sr inputs (Fig. 3A). We propose that this spatial heterogeneity can be due to an efficient buffering of SGD in the western Mediterranean areas due to Atlantic OOW inflows from the strait of Gibraltar. Indeed, the renewal time of intermediate and deep waters is estimated to be 2.5 to 5 times faster in the western Mediterranean basin than it is in the eastern basin owing to the Sicily Strait (Tanhua et al., 2013).

By summarizing, we identify significant deviations from the OOW $^{87}Sr/^{86}Sr$ ratio in various marine contexts where the interplay between oceanic and terrestrial Sr inputs is a key parameter. This relative variability attests that, depending on their isolation, shelf domains may be regarded as dynamic and transitory Sr sub-reservoirs not always representative of the global ocean reservoir for which the modern Sr residence time is estimated around 2.5 Myr (Hodell et al., 1990). This spatial decoupling in the Sr budget was suggested by de Villiers (1999) for the upper ocean by estimating partial Sr residence times of only 5800–700,000 years in the upper 400 m of the water column due to a group of celestite ($SrSO_4$)-secreting radiolarians (i.e., Acantharia). In semi-enclosed coastal domains with transient dynamics like lagoons or large epeiric seas, we expect that the partial residence time (sensu Lin and Liu, 2019) could be in the same order of magnitude as water renewal time in marginal basins (i.e., one day to half a month for studied lagoons up to 30–130 years, 2–40 years, and 5–625 years in the Mediterranean, Baltic and Black Sea basins, respectively) (Lee et al., 2002; Tanhua et al., 2013; Omstedt et al., 2014; Hilmi et al., 2017).

Finally, it worth noting that the Sr concentrations measured in the studied marine areas are relatively low (i.e., 60–70 μmol·l^{-1}) compared with those of OOW (i.e., 80–90 μmol·l^{-1}) (Fig. 2B). However, these results are in agreement with the large variability of coastal Sr concentrations (i.e., ± 20 μmol·l^{-1}) reported in worldwide coastal contexts (Fig. 5), sometimes with important annual fluctuations and local heterogeneities in euhaline waters (Ladouche et al., 2000; Brunskill et al., 2003; Elsdon and Gillanders, 2006). This suggests that the slightly non-conservative behaviour of Sr in seawater could be locally enhanced in some coastal and hemipelagic domains. Whether perennial or temporary, the cause of Sr depletions in euhaline waters – already reported in the Salses-Leucate lagoon (Ladouche et al., 2000) but also sporadically observed along the Panama coast, in the Venice lagoon (Italy) or in the Gulf of Papua (Papua New Guinea) (Brunskill et al., 2003; Beck et al., 2013) - remains obscure. We hypothesize that, in some water mass restriction contexts, three factors could be involved and combined: 1) binding of Sr at the surface of sedimentary particles depending on grain size, iron and manganese oxides, and organic matter (Takada et al., 2014); 2) fast biological adsorption rates by specific organisms (i.e., see Bowen, 1956, de Villiers, 1999); or 3) influences of saline SGD (i.e., similar to superficial porewaters) slightly depleted in Sr and with isotopic ratios close to the one of seawater (e.g., Kastner et al., 1990).

6. Conclusion

In this study, we analyzed and compared the $^{87}Sr/^{86}Sr$ ratios and the major and trace element concentrations of seawaters and shells from marginal and hemipelagic contexts with different degrees of water mass restriction. Homogeneous $^{87}Sr/^{86}Sr$ ratios typical of OOW (i.e., 0.709172 ± 0.000023) are recorded in the Tatakoto atoll and along the BSM transect. This suggests that, in open shelf context without any river inputs, alternative Sr source like SGD or particulate dissolution are too limited and likely buffered by oceanic Sr inputs. This comforts the use of carbonate fossils from these domains to infer past OOW compositions. However, slight isotopic shifts may be observed in the foreshore area (e.g., BSM beach) that we interpret as resulting from rainwater mixing, local groundwater discharges or particle dissolution in the surf zone. Elevated coastal water retention times could also account for sporadic anomalous $^{87}Sr/^{86}Sr$ ratios in open shelves. In semi-enclosed domains, we report variable Sr isotope patterns between the studied karstic lagoons, with homogeneous seawater $^{87}Sr/^{86}Sr$ ratios typical of OOW (i.e., 0.709155 to 0.709170) in the Salses-Leucate lagoon (except close to groundwater discharges where the values drop to 0.708903) and an important $^{87}Sr/^{86}Sr$ gradient from OOW values close to the Atlantic inlet to less radiogenic values of 0.707957 in the upstream parts of the Oualidia lagoon. We relate these differences to two main factors. First, we note that, despite lower fluxes, the Oualidia lagoon is supplied by SGD with unradiogenic $^{87}Sr/^{86}Sr$ ratios and very high Sr concentrations (up to 150 μmol·l^{-1}) linked to the dissolution of Mesozoic evaporites. These inputs lead to maximal contributions of 60% to the local Sr budget. The second parameter concerns the leaky, restricted or choked morphology of lagoons which controls the oceanic Sr inputs and thus buffers more or less any terrestrial Sr inputs through water mass homogenization. These two parameters appear also relevant to explain the $^{87}Sr/^{86}Sr$ gradient prevailing from west to east in the Mediterranean basin.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemgeo.2020.119874.

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