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Occurrence, origin and potential ecological risk of dissolved polycyclic aromatic hydrocarbons and organochlorines in surface waters of the Gulf of Gabès (Tunisia, Southern Mediterranean Sea)

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1 **Occurrence, origin and potential ecological risk of dissolved polycyclic aromatic**
2 **hydrocarbons and organochlorines in surface waters of the Gulf of Gabès (Tunisia,**
3 **Southern Mediterranean Sea)**

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27 **Abstract**

28 We investigated the occurrence, origin, and potential ecological risk of dissolved polycyclic
29 aromatic hydrocarbons (PAHs), polychlorinated biphenyl (PCBs) and organochlorine
30 pesticides (OCPs) in 27 surface water samples collected from a highly anthropized and
31 industrialized area in the Gulf of Gabès (Tunisia, Southern Mediterranean Sea) in October-
32 November 2017. The results demonstrated a wide range of concentrations (ng L⁻¹) with the
33 following decreasing order: $\Sigma 16$ PAHs (17.6-71.2) > $\Sigma 20$ PCBs (2.9-33.7) > $\Sigma 6$ DDTs (1.1-
34 12.1) > $\Sigma 4$ HCHs (1.1-14.8). Selected diagnostic ratios indicated a mixture of both pyrolytic
35 and petrogenic sources of PAHs, with a predominance of petrogenic sources. PCB
36 compositions showed distinct contamination signatures for tetra- to hepta-chlorinated PCBs,
37 characteristic of contamination by commercial (Aroclor) PCB mixtures. The dominant OCP
38 congeners were γ -HCH, 2,4'-DDD and 2,4'-DDE, reflecting past use of Lindane and DDTs in
39 the study area. Agricultural, industrial and domestic activities, as well as atmospheric
40 transport are identified as potential sources of PAHs, PCBs and OCPs in surface waters of the
41 Gulf of Gabès. Toxic equivalents (TEQs) suggested a low carcinogenic potential for PAHs in
42 seawater samples (mean of 0.14 ng TEQ L⁻¹). Evaluation of risk coefficients revealed low risk
43 for PAHs and PCBs, and moderate to severe risk for OCPs.

44 **Keywords:** PAHs, PCBs, OCPs, ecological risk, Gulf of Gabès, Mediterranean Sea

52 1. Introduction

53 Polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and
54 organochlorine pesticides (OCPs) are widespread in the environment (e.g. water, particulate
55 matter, sediment, soil and organisms) and have been a focus of attention due to their
56 persistence nature, bioaccumulation, toxicity, carcinogenicity and long range transport (Lee et
57 al., 2016; Miraji et al., 2021; Nguyen et al., 2014; Nyarko et al., 2011). They are a serious
58 threat to both ecosystems and human health. As a consequence, many of these compounds are
59 listed as priority pollutants by international organizations (Ashraf, 2017; Guo et al., 2019;
60 Jones and de Voogt, 1999; Muir and Howard, 2006). PCBs and OCPs have been widely used
61 for industrial and agricultural purposes, while PAHs are accidental byproducts of combustion.
62 Therefore, they can be used as chemical tracers for their possible sources (Lin et al., 2013).
63 These pollutants can enter the ocean by various vectors including rivers, effluents, runoff and
64 the atmosphere (Barhoumi et al., 2018; Moreno-González et al., 2013; Tolosa et al., 1997). In
65 marine waters, they can undergo diverse processes, such as adsorption onto particles and
66 subsequent sinking to sediments, adsorption on/absorption in biota, and accumulation into the
67 food chains (Jamieson et al., 2017; Luo et al., 2014; Ma et al., 2015). Generally, their fate in
68 the environment is strongly influenced by their physical–chemical properties (Mackay et al.,
69 1992). Due to their hydrophobicity, PAHs, PCBs and OCPs have a tendency to partitioning
70 between the dissolved phase of seawater/coastal waters and particles/sediment (Mhadhbi et
71 al., 2019; Zhang et al., 2013; Rabodonirina et al., 2015). When sediment resuspension occurs
72 due to changes in hydrodynamic processes such as waves, tides and currents, trawling and
73 ship traffic, deposited contaminants in sediments may be again reintroduced to the overlying
74 water, thus causing a secondary contamination (Zhao et al., 2021). In the dissolved state, these
75 compounds may be subjected to quite strong dynamics, i.e., exchange with the atmosphere
76 (deposition, volatilization), photodegradation, interactions with dissolved and particulate

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77 organic matter, or transport with water masses. In addition, from their dissolved form, they
78 may bioconcentrate in a variety of micro- and macro-organisms *via* respiration or passive
79 diffusion (Dachs et al., 2002; Dachs and Méjanelle, 2010; Daewel et al., 2020; Duran and
80 Cravo-Laureau, 2016; Montuori et al., 2014; Salvadó et al., 2019). Therefore, the knowledge
81 of the distribution of dissolved PAHs, PCBs and OCPs is an essential step to better
82 understand and quantify their global budget and their potential impact on a marine
83 environment.

84 The Mediterranean Sea, a semi-closed aquatic ecosystem, is particularly vulnerable to
85 chemical pollution due mainly to the long history of industrialization of its basin, its high
86 population density, and its natural characteristics (such as large watersheds, high continental
87 inputs, intense and extreme meteorological events, thermohaline circulation) (The Mermex
88 group, 2011). As consequence, a multitude of chemical pollutants derived from urban,
89 agricultural and industrial activities find their way into the Mediterranean waters (Campillo et
90 al., 2019; Gómez-Gutiérrez et al., 2007; Köck-Schulmeyer et al., 2021; Sánchez-Avila et al.,
91 2012). The Gulf of Gabès (Southeastern Tunisia) is one particular ecoregion of interest within
92 the Mediterranean Sea basin due to its hydrodynamics, biogeochemical and biological
93 features (Ayata et al., 2018; D’Ortenzio and Ribera d’Alcalà, 2009). It is one of the most
94 productive areas of the Mediterranean, characterized by hydrodynamics driven by tides and
95 low bathymetry (Béjaoui et al., 2019; Hattab et al., 2013). While the Gulf of Gabès
96 contributes approximately 40% of the national fish production in Tunisia (DGPA, 2015), the
97 coastal zone of the Gulf of Gabès is subjected to high anthropogenic pressures due to
98 numerous industrial and agricultural activities within the zone. The latter are mainly
99 concentrated in great coastal urban centers (Gabès and Sfax cities), which host fishing and
100 commercial harbours, phosphoric acid production plants, municipal landfills, and various
101 industries (Fourati et al., 2018; Nedja et al., 2011; Zaghden et al., 2007). Despite its

102 ecological and biogeochemical importance within the Mediterranean Sea, and at the same
103 time, its high level of anthropogenic pressure, the Gulf of Gabès remains very poorly
104 documented in terms of concentrations of organic pollutants in the dissolved phase of
105 seawater, in particular organochlorine (OCs) compounds such as PCBs and OCPs. In this
106 context, the objectives of this work were i) to describe the spatial distribution of dissolved
107 PAHs, PCBs and OCPs (concentrations, molecular patterns) in surface coastal waters of the
108 Ghannouch-Gabès area (Gulf of Gabès, Tunisia) and compare this distribution with that of
109 core physico-chemical and biogeochemical parameters, ii) to assess the main origins/sources
110 of these organic contaminants, and iii) to determine their potential ecological risk. To our
111 knowledge, this work is the first to provide together levels of dissolved PAHs, PCBs and
112 OCPs in coastal marine waters of Tunisia.

113

2. Material and Methods

2.1. Study area

114 The Gulf of Gabès is located in the Southeastern part of Tunisia (Southern Mediterranean
115 Sea), and spreads over 400 km of coastline (more than 50% of the Tunisian coastline)
116 between the city of Chebba in the North and the Tunisian-Libyan border in the South (Fig. 1).
117 The Gulf of Gabès, which accounts for ~ 30% of the Tunisian coastal waters, is characterized
118 by a wide continental shelf, shallow waters (isobaths 50-m deep are reached 110-km
119 offshore), high amplitude (up to 2-m) tides, and hydrodynamics driven by tides and
120 anticyclonic winds (Béjaoui et al., 2019; Zayen et al., 2020). The Gulf of Gabès hosts coastal
121 cities, such as Sfax and Gabès, with concentrated industrial, harbour and agriculture activities.
122 The Ghannouch-Gabès area in the central part of the Gulf, holds one of the biggest industrial
123 zones in Tunisia. This industrial complex includes the Ghannouch commercial harbour, the
124 phosphoric acid industry of the Tunisian Chemical Group, the Gabès Fluorine Chemical

127 Industry, a thermal power plant, petrochemical infrastructures, as well as industries related to
128 agribusiness, building materials, energy production and textiles (El Zrelli et al., 2015, 2018;
129 Fourati et al., 2018). The phosphoric acid industry of the Ghannouch industrial complex has
130 been discharging the phosphogypsum, an acidic by-product of phomanufacturing, into the
131 Gabès coastal waters over the past several decades. The solid phosphogypsum issued from the
132 production of phosphoric acid, diluted with seawater to form gypsum water, is conveyed by
133 several pipes to the South of the Ghannouch commercial harbour where the gypsum water
134 mixes with the cooling water discharged by all the chemical industry units, and finally ends
135 up at sea through a unique canal (Darmoul et al., 1980). Between 1000 and 13,000 t of
136 phosphogypsum is released daily this way into the sea (Béjaoui et al., 2004; El Zrelli et al.,
137 2015). In addition to this phosphogypsum discharges, several wadis carry a variety of urban,

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2.2. Sampling and in situ measurements

701 Samples were collected at 27 stations in the Ghannouch-Gabès coastal area during the
702 MERITE cruise from 30 October to 3 November 2017. Stations were distributed along
703 coastal-offshore transects (also corresponding to North-South transects), i.e., S1-S3, S20 in
704 front of El Melah wadi (Northern Ghannouch), S4-S6, S21 in front of El Demna wadi
705 (Northern Ghannouch), S7-S12, S23 in front of Ghannouch industrial complex/commercial
706 harbour and phosphogypsum effluent outlet, S13-S15 in front of fishing harbour and Gabès
707 wadi, and S16-S18, S27 in front of Mersit wadi (Southern Gabès). Also, S19 was positioned
708 as the north most station, S22 close to the Ghannouch industrial complex, in front of Ettine
709 wadi/fluorine discharges, S24 and S25 between the Ghannouch industrial complex and the
710 fishing harbour, and S26 in front of Griaa wadi (Fig. 1, Table S1). Stations S1-S18 (> 5-m
711 depth) were sampled on board the vessel “Rahma” on 30 October (S1-S6), 1 November (S7-
712 S12) and 3 November (S13-S18). The most coastal stations S19-S27 (< 5-m depth) were

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sampled on board a zodiac-type vessel on 31 October (S23-S27) and 2 November (S19-S22). Stations S1-S11 and S16 were sampled at high tide, while stations S12-S15 and S17-S27 were sampled at low tide. Sunny weather and very calm sea conditions prevailed during the cruise, except during the sampling of stations S13-S18 (3 November) for which windy weather and rough sea conditions were encountered (Fig. 1, Table S1).

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At each station, seawater samples for the analyses of dissolved organic pollutants (PAHs, PCBs and OCPs) and other biogeochemical parameters were collected at ~ 0.5-m depth using a LIMNOS water sampler (Hydro-Bios) equipped with two Schott® glass bottles of 1-L volume each. This sampler allowed the bottles to go down in water closed and thus avoid the sampling of the surface microlayer. The glass bottles were rinsed several times with sea water before filling. Once filled, they were unscrewed from the sampler, closed with their Teflon lined caps, and stored in the dark at 4 °C. Before sampling, the glass bottles were extensively washed with 1 M hydrochloric acid (HCl) and ultrapure (resistivity 18.2 MΩ cm) water, and baked at 450 °C for 6 h.

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Besides sampling, *in situ* profiles of temperature, salinity, concentration of total chlorophyll *a* (TChl*a*) and turbidity were conducted in the water column (from 0.5-m depth to the bottom) using a 19 *plus* SBE conductivity-temperature-depth (CTD) profiler (SeaBird Electronics, Inc.) equipped with a WETStar Chl*a* fluorometer (WETLabs, Inc.) and a STM turbidity meter (Seapoint, Inc.). Moreover, a Tripod multiparameter probe (Aqualabo) was deployed in surface waters (at ~ 0.5-m depth) for the measurement of temperature, salinity, pH, dissolved oxygen (O₂) concentration and redox potential (redox). Sensors were calibrated just before the cruise according to manufacturer's procedures.

54 735 **2.3. Filtration of samples**

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Once in the laboratory, seawater samples were immediately filtered under a low vacuum (< 50 mm Hg) through pre-baked (450 °C, 6 h) GF/F (~ 0.7 μm) glass fiber filters (25- or 47-mm

738 diameter, Whatman) using glassware filtration systems. For dissolved organic pollutants
739 (PAHs, PCBs, OCPs), 1 L of sample was filtered through 47-mm diameter GF/F filters.
740 Filtered samples were stored in 2.5 L glass bottles with 50 mL of dichloromethane (CH₂Cl₂)
741 at 4 °C in the dark before solvent extraction.

742 For the determination of the concentrations of suspended particulate matter (SPM),
743 particulate organic carbon and nitrogen (POC and PON), as well as δ¹³C and δ¹⁵N isotopic
744 ratios, between 250 and 1100 mL of sample were filtered with pre-weighted 25-mm diameter
745 GF/F filters (the same filter was used for SPM, POC, PON, δ¹³C and δ¹⁵N analyses). After
746 seawater filtration, filters were rinsed with ultrapure water to remove salts, and then were
747 dried at 60 °C for 24 h and stored in aluminum foil until analysis.

748 ***2.4. Analytical procedures for PAHs, PCBs and OCPs***

749 The analyzed compounds included 16 PAHs, 20 PCB congeners and 10 OCPs ([Text S1](#),
750 [Table S2](#)). The targeted PAHs were naphthalene (Nap), acenaphthylene (Acy), acenaphthene
751 (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr),
752 benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene
753 (BkF), benzo[a]pyrene (BaP), dibenz[a,h]anthracene (DahA), benzo[g,h,i]perylene (BghiP),
754 and indeno[1,2,3-cd]pyrene (IcdP). The 20 PCBs congeners were PCB 8, 18, 28, 52, 44, 66,
755 77, 101, 105, 118, 126, 128, 138, 153, 170, 180, 195, 206, 187 and 209. The 10 OCPs were
756 four hexachlorocyclohexanes (α-HCH, β-HCH, γ-HCH, δ-HCH), as well as two
757 dichlorodiphenyltrichloroethanes (2,4'-DDT, 4,4'-DDT) and their metabolites,
758 dichlorodiphenyldichloroethylenes (2,4'-DDE and 4,4'-DDE) and
759 dichlorodiphenyldichloroethanes (2,4'-DDD and 4,4'-DDD). Details on analytical procedures
760 for the determination of dissolved PAHs, PCBs and OCPs including chemicals,
761 extraction/cleanup and instrumental analysis are presented in [Text S1](#). Briefly, water samples
762 (dissolved phase) were spiked with a multi standard mixture containing surrogate standards

763 for PAHs, PCBs and OCPs, and extracted by Liquid–Liquid extraction (LLE). The extracts
764 were then purified using silica-alumina columns, internal standards were added before the
765 analysis which was subsequently performed using gas chromatography coupled with a mass
766 spectrometer (GC–MS) and an electron capture detector (GC-ECD) (Text S1, Table S2).

767 **2.5. Quality assurance/Quality control**

768 All data were guaranteed through the implementation of strict QA/QC procedures. Before
769 use, all glassware was intensively cleaned and baked at 450 °C for 6 h. All the material that
770 could not be baked was washed with 1 M HCl, ultrapure water, *n*-hexane:acetone mixture 1:1
771 (v/v) and then dried at room temperature. Procedural blanks (to determine the contamination
772 introduced during extraction and cleanup) were processed in the same way as the real samples
773 and were included with each batch of 5 samples. Most of the analytes were not detected in
774 blanks, except Nap, Acy, Ace, Flu, Phe, Ant, Flt, Pyr, BaA and Chr. Their amounts were not
775 significant relative to that of the water samples (< 7%). The recoveries of target compounds
776 were checked using standard mixtures spiked in ultrapure water (50 ng for PAHs, 20 ng for
777 PCBs and 30 ng for OCPs). They ranged from 63 to 114% (mean of 87%), 7 to 117% (mean
778 of 85%) and 20 to 115% (mean of 79%) for PAHs, PCBs and OCPs, respectively (Table S2).
779 The surrogate standard (added to perform the quantification) recoveries ranged from 69 to
780 92% and from 66 to 84% for PAHs and OCs, respectively (Table S3). Method detection limits
781 (MDLs) defined as mean blank value + 3 × standard deviation (SD) ranged from 0.04 to 2.66
782 ng L⁻¹, 0.03 to 0.42 ng L⁻¹ and 0.10 to 0.40 ng L⁻¹ for PAHs, PCBs and OCPs, respectively
783 (Table S2). For compounds that were not detected in blanks, instrument detection limits
784 (IDLs), determined as a signal-to-noise ratio (S/N) of 3, were substituted. If the concentration
785 of a given compound in a sample was below its MDL/IDL, thus this compound was
786 considered as not detected in the sample, and was assumed zero. All data reported here were
787 blank corrected.

788 2.6. Data handling and risk assessment

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2 789 The low molecular weight (LMW) PAHs (Nap, Acy, Ace, Flu, Phe, Ant) are petrogenic
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4 790 (non-combustion processes) origin while the high molecular weight (HMW) PAHs (Flt, Pyr,
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7 791 BaA, Chr, BbF, BkF, BaP, DahA, BghiP, IcdP) are rather associated with incomplete
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9 792 pyrogenic processes origin. In addition, two molecular PAH ratios, i.e., Flt/(Flt+Pyr) and
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11 793 Ant/(Ant+Phe) were used to identify the possible PAH sources (petrogenic *versus* pyrogenic)
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13 794 (Table S4). According to their degree of chlorination, the 20 PCBs can be classified into 9
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15 795 groups: di-(PCB-8), tri- (PCB-18 and -28), tetra- (PCB-52, -44, -66 and -77), penta- (PCB-
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17 796 101, -105, -118 and -126), hexa- (PCB-128, -138 and -153), hepta- (PCB-170 and -180), octa-
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19 797 (PCB-195), nona- (PCB-206) and deca-PCBs (PCB-187 and -209). Among the 20 PCB
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21 798 congeners targeted, seven PCBs (PCB-28, -52, -101, -118, -138, -153, and -180), are named
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23 799 indicators PCBs ($\Sigma 7$ IN-PCBs). Among the 10 targeted OCPs, $\Sigma 4$ HCHs represent the sum of
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25 800 α -, β -, γ - and δ -HCH and $\Sigma 6$ DDTs represent the sum of 2,4'-DDT, 4,4'-DDT, 2,4'-DDE, 4,4'
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27 801 '-DDE, 2,4'-DDD and 4,4'-DDD. Two ratios, (DDE+DDD)/ Σ DDTs and DDD/DDE, were
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29 802 used to determine the recent/historical character of DDT contamination and to determine the
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31 803 type of biodegradation mechanism (aerobic or anaerobic) to which DDT is subjected in the
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33 804 study area (Table S4), respectively.

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35 805 To estimate the environmental risk posed by PAHs in the study aquatic system, toxicity
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37 806 equivalents (TEQs) was calculated using Toxicity Equivalent Factors (TEFs) (Nisbet and
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39 807 LaGoy, 1992), as follows:

$$40 \quad \text{TEQs} = \text{TEF} \times \text{PAH concentration}$$

41
42 808 The risk quotients of negligible concentrations $\text{RQ}_{(\text{NCs})}$ and maximum permissible
43
44 809 concentrations $\text{RQ}_{(\text{MPCs})}$ (Cao et al., 2010) were calculated as follows:

$$45 \quad \text{RQ}_{(\text{NCs})} = C_{\text{PAHs}}/C_{(\text{NCs})}$$

$$46 \quad \text{RQ}_{(\text{MPCs})} = C_{\text{PAHs}}/C_{(\text{MPCs})}$$

813 Finally, the risk of PAHs on benthic species endemic in the water column ($\sum RQs$) was
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 2 814 calculated by the sum of RQs for individual PAHs, as follows:
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$$815 \quad RQ_{\Sigma PAHs(NCs)} = \sum_{i=1}^{16} RQi \left(RQ_{(NCs)} \geq 1 \right)$$

$$816 \quad RQ_{\Sigma PAHs(MPCs)} = \sum_{i=1}^{16} RQi \left(RQ_{(MPCs)} \geq 1 \right)$$

817 The interpretation of the risk classification of individual PAHs and $\Sigma PAHs$ are as follows
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 16 818 ([Cao et al., 2010](#)). In principle, $RQ_{(MPCs)} > 1$ indicates that contamination by the single PAH is
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 18 819 sever, whereas an $RQ_{(NCs)} < 1$ indicates that a single PAHs is probably of negligible concern.
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 21 820 In the case of $RQ_{(NCs)} > 1$ and $RQ_{(MPCs)} < 1$, contamination of the single PAH might be
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 23 821 considered moderate. For $RQ_{\Sigma PAHs}$, $RQ_{\Sigma PAHs(MPCs)} < 1$ indicates PAH contamination is low-
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 26 822 level, while $RQ_{\Sigma PAHs(NCs)} > 800$ would indicate severe PAH contamination. If $RQ_{\Sigma PAHs(NCs)} \geq 1$
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 28 823 or < 800 and $\sum RQ_{(MPCs)} > 1$, it likely indicates moderate PAH contamination.

824 Regarding OCP and PCBs, the risk assessment was estimated similarly using the following
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 33 825 equations:
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$$826 \quad RQ = C_{TCS}/C_{QV}$$

$$827 \quad RQ_{CCCs} = C_{TCS}/C_{QV(CCCs)}$$

$$828 \quad RQ_{CMCs} = C_{TCS}/C_{QV(CMCs)}$$

829 where, C_{TCS} is the concentration of targeted contaminants, C_{QV} , is the corresponding quality
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 45 830 values of certain OCPs and PCBs, RQ_{CCCs} is the risk quotient for continuous criteria
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 48 831 concentrations (CCCs), whereas RQ_{CMCs} is the risk quotient for maximum criteria
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 50 832 concentrations (CMCs) ([USEPA, 2006](#)).

833 **2.7. Other analyses**

834 After 24 h in the desiccator, 25-mm GF/F filters were reweighed by the same balance that
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 59 835 was used for pre-weighting. The SPM concentration was then calculated as the difference

836 between filter weight before and after sample filtration, normalized to the filtration volume
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2 837 (Neukermans et al., 2012). POC and PON concentrations, and $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ stable isotopic
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4 838 ratios were determined by high combustion method with mass spectrometer (Raimbault et al.,
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7 839 2008). Measurements of fluorescent dissolved organic matter (FDOM) at 254-nm excitation
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9 840 wavelength and 360-nm emission wavelength were performed on unfiltered (raw) seawater
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11 841 samples (~ 500 mL) with a UV EnviroFlu-HC fluorometer (TriOS Optical sensors) (Tedetti et
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13 842 al., 2010). Here, fluorescence data are provided in relative unit (RU).

17 843 **2.8. Statistics**

20 844 Statistical analyses, including Pearson correlations and a principal component analysis
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22 845 (PCA) based on Spearman's rank order correlation matrix, were performed using XLSTAT
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24 846 2013.5.01. Colour maps of physico-chemical parameters and pollutant concentrations were
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26 847 carried out using Ocean Data View (ODV) software version 4.6.5 (<http://odv.awi.de>). The
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28 848 spatial interpolation/gridding of data was conducted using Data-Interpolating Variational
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30 849 Analysis (DIVA) (Barth et al., 2010; Troupin et al., 2012).

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37 851 **3. Results**

41 852 **3.1. Spatial distribution of physico-chemical and biogeochemical parameters**

43 853 The spatial distribution of temperature, salinity, pH, FDOM, O_2 concentration, and redox
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45 854 potential in surface waters in the study area (at ~ 0.5-m depth) is shown Fig. 2. Temperature
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47 855 did not display any strong variability (from 21.48 °C at S4 to 23.43 °C at S22), nor any clear
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49 856 trend relative to the phosphogypsum effluent outlet (S23, Fig. 2a). However, salinity, pH,
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51 857 FDOM and O_2 concentration exhibited a distribution pattern related to the effluent. Indeed,
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53 858 the most coastal stations, particularly stations S23, S24 and S25 (the three stations located
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55 859 between the two harbours and directly under the influence of the effluent) presented the
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860 highest salinity values (40.11-40.95), the lowest pH values (7.49-7.82), the highest FDOM
861 values (1089-1410 RU), and the highest O₂ concentrations (9.6-10.6 mg L⁻¹) (Figs. 2b-e).
862 Stations S22 and S26, situated North and South of these 3 stations, respectively, also showed
863 higher values of FDOM (634-824 RU), O₂ concentration (8.6-9.8 mg L⁻¹), and lower values of
864 pH (8.17-8.29) relative to the other stations. In contrast, the lowest values of salinity (<
865 39.40), FDOM (< 600 RU), O₂ concentration (< 8.0 mg L⁻¹), and the highest values of pH (>
866 8.30) were encountered mostly in the Northern and offshore sites (S1-S11) (Figs. 2b-e).
867 Redox potential displayed the lowest values (< 200 mV) along the transect in front of
868 Ghannouch commercial harbour/phosphogypsum discharges (S7, S8, S10, S11), whereas the
869 highest values (> 260 mV) were found in the Southern part (S13-S18) (Fig. 2f).

870 The coastal stations S23-S25 were also characterized by the highest POC and PON
871 concentrations (73.2-89.6 μM and 8.9-10.9 μM, respectively), the highest SPM values (15.0-
872 16.8 mg L⁻¹), and the lowest δ¹⁵N values (-1.2-1.9‰) (Table S5). On the other side, the
873 lowest concentrations of POC, PON and SPM were recorded in the Northern and offshore
874 sites (S3, S4, S8, S11) (Table S5). Also, salinity, FDOM, O₂ concentration, POC, PON and
875 SPM concentrations were positively (linearly) correlated to each other (r = 0.65-0.97, n = 15,
876 p < 0.0001-0.009), while these latter were all negatively correlated to pH (r = -0.70-0.93, n =
877 15, p < 0.0001-0.004). Turbidity and TChl_a concentration, albeit not measured in stations
878 S19-S27, showed higher values in the Southern stations (S13-S18) than in the Northern ones
879 (Table S5). Overall, these results highlight coast-offshore and South-North gradients of the
880 different physico-chemical and biogeochemical parameters, with a substantial influence of the
881 phosphogypsum effluent outlet on the local biogeochemistry.

882 **3.2. Spatial distribution of total dissolved PAHs, PCBs and OCPs**

883 The spatial distribution of concentrations of Σ16 PAHs (sum of 16 compounds), Σ20 PCBs
884 (sum of 20 congeners) and Σ10 OCPs (sum of 10 compounds) in surface waters (at ~ 0.5-m

885 depth) is provided [Fig. 3](#). Detailed information about individual compounds is given in [Table](#)
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2 886 [S6](#). Concentration of $\Sigma 16$ PAHs was the highest in the vicinity of Ghannouch industrial
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5 887 complex: at stations S8 and S9 (62.7-71.2 ng L⁻¹), in front of Ghannouch
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7 888 harbour/phosphogypsum discharges, at station S22 (64.9 ng L⁻¹), in front of Ettine
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10 889 wadi/fluorine discharges, at stations S4, S19, S20 (40.5-59.8 ng L⁻¹), Northern Ghannouch,
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12 890 and at station S24 (46.1 ng L⁻¹), between Ghannouch and Gabès harbours ([Fig. 3a](#), [Table 1](#)).
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14 891 Concentration of $\Sigma 20$ PCBs showed a quite different distribution with the highest values
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16 892 found off Ghannouch-Gabès area (at S9: 33.7 ng L⁻¹ and S11: 31.4 ng L⁻¹), Northern
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19 893 Ghannouch (at S1: 22.1 ng L⁻¹), and in front of fishing harbour and Gabès wadi (at S13: 28.3
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22 894 ng L⁻¹) ([Fig. 3b](#), [Table 1](#)).

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24 895 For OCPs, concentration of $\Sigma 4$ HCHs displayed a distribution pattern relatively similar to
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26 896 that of PAHs, with the highest values observed around the Ghannouch industrial complex and
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29 897 Northern Ghannouch: at stations S8 (14.8 ng L⁻¹), S22 (9.4 ng L⁻¹), S19 and S20 (6.4-7.0 ng
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31 898 L⁻¹) ([Fig. 3c](#), [Table 1](#)). Interestingly, $\Sigma 16$ PAH and $\Sigma 4$ HCH concentrations were positively
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34 899 correlated ($r = 0.72$, $n = 27$, $p < 0.001$). The highest concentration of $\Sigma 6$ DDTs observed was
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36 900 in Northern Ghannouch (at S4-S6 and S19: 11.3-13.2 ng L⁻¹), and in front of phosphogypsum
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39 901 discharges (at S8: 12.0 ng L⁻¹), as well as Southern Gabès, in front of Griaa wadi (at S26: 10.9
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41 902 ng L⁻¹) and in front of Mersit wadi (at S16: 11.7 ng L⁻¹) ([Fig. 3d](#), [Table 1](#)). It appeared that the
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44 903 spatial distributions of PAHs, PCBs, HCHs and DDTs were relatively different from those of
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46 904 the physical and physico-chemical parameters described above (i.e., salinity, pH, FDOM, O₂
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48
49 905 concentration and redox potential) because the contaminant concentrations exhibited less
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51 906 pronounced coast-offshore gradients, and were less driven by the phosphogypsum effluent
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54 907 outlet. PAHs and OCPs (particularly HCHs) nonetheless seemed to show more similarities
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56 908 with physico-chemical parameters than PCBs, especially regarding their higher concentrations
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58 909 in the Ghannouch/effluent zone.

910 3.3. Molecular distribution of dissolved PAHs, PCBs and OCPs

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3 911 Among the 16 PAHs targeted in this study, four were not detected in the samples: BaP,
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5 912 DahA, IcdP, and BghiP (Fig. 4a). The most abundant PAHs were Nap (on average 63% of
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7 913 total PAHs, ranging from 27% at S7 to 82% at S19) followed by Phe (on average 12%,
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10 914 ranging from 7% at S8, S19, S22 to 25% at S4), and Flt (on average 7%, ranging from 1% at
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12 915 S8, S22 to 36% at S7), while other compounds only accounted for 0-5% on average of total
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15 916 PAHs (Fig. 4a). With respect to size, the concentrations of LMW-PAHs in surface waters of
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17 917 the Gulf of Gabès ranged from 15.8 to 62.4 ng L⁻¹ and for HMW-PAHs, the range was 1.4 to
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20 918 23.3 ng L⁻¹. The Flt/(Flt+Pyr) ratio was 0.56 on average and ranging in values from 0.40
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22 919 (S10) to 0.90 (S7), while the Ant/(Ant+Phe) ratio was 0.07 on average with values ranging
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25 920 from 0.01 (S1) to 0.21 (S8) (Table 1).

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27 921 Of the 20 targeted PCB congeners, 11 were not detected in the samples. These were PCB-
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29 922 8, -44, -66, -77, -105, -126, -170, -187, -195, -206, -209 (Fig. 4b). The relative abundance of
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32 923 the 9 congeners detected was highly variable from station to station. PCB-180 alone
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34 924 accounted for 27% of the PCBs on average, with minimal contribution of 1% (at S10) and
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37 925 maximal contribution of 71% (at S26). PCB-18 and -52 were also detected in large amounts.
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39 926 The two congeners represented 18 and 19% of PCBs on average, respectively, with minimal
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42 927 values of 0% (at S3 and S26 for PCB-18, and at S12, S14 and S15 for PCB-52) and maximal
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44 928 values of 41% (at S20 for PCB-52) or 59% (at S14 for PCB-18). PCB-101, -28 and -153
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47 929 displayed lower mean relative abundances representing 14, 10 and 6% (respectively) while
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49 930 the sum of the remaining congeners (PCB-118, -128, -138) only depicted 6% of total PCBs
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51 931 (Fig. 4b).

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54 932 Among OCPs of interest, 2,4'-DDT, 4,4'-DDT and 4,4'-DDD were not at all detected. 2,4'-
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56 933 DDD and 2,4'-DDE compounds were major species, with average contributions to total OCPs
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59 934 of 31 and 28%, respectively, minimal contributions of 0% (at S12, S15) and 4% (at S7, S18),
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2 936 respectively, and maximal contributions of 64% (at S7) and 51% (at S11), respectively (Fig.
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4 937 4c). They were followed by γ -HCH, which represented on average 19% with minimal and
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6 938 maximal values of 4% (at S11, S14) and 51% (at S12), respectively. The sum of the
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8 939 remaining compounds (α -HCH, β -HCH, δ -HCH, 4,4'-DDE) accounted on average for 22% of
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10 total OCPs (Fig. 4c). The DDD/DDE ratio was quite variable, ranging from 0.21 (S11) to 3.74
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12 940 (S7), with mean and median values of 1.30 and 0.80, respectively (Table 1). Since 2,4'-DDT
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14 941 and 4,4'-DDT were not detected, the (DDE+DDD)/ Σ DDTs ratios were all equal to 1 (data not
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16 942 reported in Table 1).

19 943 **3.4. PCA classification of stations**

22 944 Spearman PCA was applied on various parameters including salinity, pH, O₂
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24 945 concentration, FDOM, as well as concentrations of Σ 16 PAHs, Σ 20 PCBs and Σ 10 OCPs (Fig.
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26 946 5). The first principal component (PC1), which explained 39% of total variability within
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28 947 samples, mainly received the contribution of variables salinity, pH, FDOM and in a lesser
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30 948 extent O₂ concentration and Σ 10 OCPs. The second principal component (PC2) accounted for
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32 949 21% of total variability within samples and was mostly related to variables Σ 16 PAHs, Σ 20
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34 950 PCBs, followed by O₂ concentration and Σ 10 OCPs. PCA allowed the discrimination of
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36 951 different groups of stations. Group n°1 (S2, S3, S5, S7, S10, S12, S14, S17, S18, S21 and
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38 952 S27) is rather characterized by high values of pH, low values of salinity, O₂ concentration and
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40 953 FDOM, and low concentrations of total dissolved PAHs, PCBs and OCPs. Group n°2 (S1, S6
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42 954 and S11) is depicted by quite high values of pH, low values of salinity, O₂ concentration and
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44 955 FDOM, and high concentrations of total dissolved PAHs, PCBs and OCPs. Group n°3 (S4,
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46 956 S8, S9, S13, S16, S19, S20, S22) is defined by lower values of pH, higher values of salinity,
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48 957 O₂ concentration and FDOM, and high concentrations of total dissolved PAHs, PCBs and
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50 958 OCPs. Finally, group n°4 is characterized by roughly low values of pH, high values of
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2 960 salinity, O₂ concentration and FDOM, and low concentrations of total dissolved PAHs, PCBs
3 and OCPs (Fig. 5).

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6 7 962 **4. Discussion**

8 9 10 963 **4.1. Concentration levels**

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13 964 The concentrations of total dissolved ($\Sigma 16$) PAHs (17.6-71.2 ng L⁻¹) recorded here in the
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15 965 surface waters of the Ghannouch-Gabès area (Fig. 3a, Table 1) were of the same order of
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18 966 magnitude but were lower than those recently measured in the neighboring area by Fourati et
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20 967 al. (2018) (i.e., 9-198 ng L⁻¹; Table S7). This difference may be due to the fact that this latter
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22 968 study targeted more compounds ($\Sigma 29$ PAHs) including alkylated compounds, which are
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25 969 recognized to account for a large part of the pool of dissolved PAHs in natural waters (Guigue
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27 970 et al., 2014; Wang et al., 1999). Also, dissolved PAH concentrations were in the lower range
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30 971 of those reported in coastal Bohai Bay, China (48-607 ng L⁻¹ for $\Sigma 16$ PAHs) (Tong et al.,
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32 972 2019), various coastal areas of the Southern, Northern and Northwestern Mediterranean Sea,
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35 973 such as the Gulf of Tunis, Tunisia (139-1008 ng L⁻¹ for $\Sigma 24$ PAHs) (Mzoughi and Chouba,
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37 974 2011), the Marseilles coastal area, France (1.9-151 ng L⁻¹ for $\Sigma 17$ PAHs) (Guigue et al., 2011,
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39 975 2014) and the Alexandria coast, Egypt (13-120 ng L⁻¹ for $\Sigma 7$ PAHs) (El Nemr and Abd-Allah,
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41 976 2003), but were higher than those observed in the Open Western and Eastern Mediterranean
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44 977 Sea (0.16-9 ng L⁻¹ for $\Sigma 19$ PAHs) (Berrojalbiz et al., 2011), the Northwest Atlantic coast of
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47 978 the Iberian Peninsula (9.3 ± 0.6 ng L⁻¹ for $\Sigma 16$ PAHs) (Rocha and Rocha, 2021), the European
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49 979 Atlantic coastline, Portugal (8.3-9.5 ng L⁻¹ for $\Sigma 16$ PAHs) (Rocha et al., 2021) and Barcelona,
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52 980 Spain and Banyuls-sur-Mer, France (0.4-8.6 ng L⁻¹ for $\Sigma 15$ PAHs) (Garcia-Flor et al., 2005)
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54 981 (Table S7).

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57 982 The concentrations of total dissolved ($\Sigma 20$) PCBs (2.9-33.6 ng L⁻¹) we determined here in
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59 983 the Gulf of Gabès (Fig. 3b, Table 1) were in most cases higher than those recorded in other
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1 984 environments, such as the Open Baltic Sea ($0.02 \pm 0.01 \text{ ng L}^{-1}$ for $\Sigma 7$ PCBs) (Cornelissen et
2 985 al., 2008) and the Tokyo Bay, Japan ($0.04\text{-}0.6 \text{ ng L}^{-1}$ for $\Sigma 209$ PCBs) (Kobayashi et al., 2010).
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4 986 However, they were of the same order of magnitude than those recorded in the Singapore's
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7 987 northeastern and southwestern coastlines ($0.22\text{-}20.41 \text{ ng L}^{-1}$ for $\Sigma 8$ PCBs) (Basheer et al.,
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9 988 2003), and they were lower than those found in the Daya Bay, China ($91.1\text{-}1355.3 \text{ ng L}^{-1}$ for
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12 989 $\Sigma 12$ PCBs) (Zhou et al., 2001), and the Panama Canal and California Coast, USA (*bdl*- $18,450$
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14 990 ng L^{-1} for $\Sigma 19$ PCBs) (Menzies et al., 2013) (Table S7). Urbanization, human activities and
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16 991 particularly, intense industrialization in this area may explain the higher PCB concentrations
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19 992 compared to the other environments.

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22 993 The concentrations of total dissolved ($\Sigma 10$) OCPs ($5.3\text{-}26.7 \text{ ng L}^{-1}$) found here (Fig. 3c and
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24 994 d, Table 1) were in the same range than those measured in Hangzhou Bay, China ($1.3\text{-}26 \text{ ng}$
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26 995 L^{-1} for $\Sigma 10$ OCPs) (Li et al., 2016) and Singapore's northeastern and southwestern coastlines
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28 996 ($4.9\text{-}22 \text{ ng L}^{-1}$ for $\Sigma 12$ OCPs) (Basheer et al., 2003), but lower than that recorded in Jinzhou
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30 997 Bay, China ($4.2\text{-}136.8 \text{ ng L}^{-1}$ for $\Sigma 19$ OCPs) (Yao et al., 2013) (Table S7). Large differences
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33 998 in pesticide concentrations in global surface waters may be explained mainly by differences in
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36 999 agricultural activities, given the strong connection between OCPs and agriculture (Montory et
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39 1000 al., 2017). Although all these comparisons should be taken with caution due to the different
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41 1001 number of compounds analyzed, the tendency that emerges is that the concentrations we
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44 1002 measured in this work in the Ghannouch-Gabès area (Southern Gulf of Gabès) were in the
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46 1003 mid-range of those reported in other coastal environments (Table S7).

48 49 1004 **4.2. Sources/origins**

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52 1005 Compositional profiles for PAHs in water samples are illustrated in Fig. 4a. LMW-PAHs
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54 1006 were abundant at all sampling sites, representing on average $> 80\%$ of $\Sigma 16$ PAHs; HMW-
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57 1007 PAHs, on the other hand, were present in low concentrations and contributions to $\Sigma 16$ PAHs.
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1008 This is typical for surface waters because of the relatively high solubility of LMW-PAHs (log
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21009 K_{ow} is 3-5 for LMW and 5-7 for HMW) (Moeckel et al., 2014; Shen et al., 2021).
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51010 The evaluation of PAH sources by sampling site was achieved by considering published
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71011 indicators (Table 1) and the analysis of bivariate cross plots, which reflect characteristic
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101012 PAHs ratios (Fig. S1). Because PAHs naturally partitioned between dissolved and particulate
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121013 water phases in line with their molecular weight/hydrophobicity, the dominance of the LMW
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141014 compounds in the present samples (especially Nap and Phe), which are much more soluble
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171015 than the HMW ones, cannot be used as an origin/source criterion. For that, isomeric
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191016 compound ratios, Ant/(Ant+Phe) and Flt/(Flt+Pyr) are more reliable, although they might be
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221017 affected by weathering processes between the emission points and the sampling area
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241018 (Katsoyiannis and Breivik, 2014). According to Table 1 and Table S4, PAHs were found to be
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271019 both pyrolytic and petrogenic origin, with a predominance of petrogenic sources (63% of the
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291020 stations had a ratio of Flt/(Flt+Pyr) between 0.4 and 0.5, and 89% of the stations had a ratio of
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321021 Ant/(Ant+Phe) < 0.1) (Ravindra et al., 2006; Soclo et al., 2000; Yunker et al., 2002). The plot
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341022 of isomeric ratios, Ant/(Ant+Phe) versus Flt/(Flt+Pyr) (Fig. S1), confirm the presence of the
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361023 last referred sources in surface waters. This reflects the multitude of sources of PAHs in this
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391024 anthropized coastal area of the Gulf of Gabès through the utilization of petroleum products
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411025 and incomplete combustion of fossil fuels and biomass (Stogiannidis and Laane, 2015).
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441026 Petrogenic PAHs could be introduced *via* accidental oil spills, discharges from routine ship
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461027 and harbour activities/operations, and urban runoff, while pyrogenic PAHs may enter *via*
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491028 exhausts and residues issued from the combustion related to the various urban, industrial and
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511029 agriculture activities occurring in this region. Moreover, PAHs, particularly LMW-PAHs
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541030 prevailing in this study, can reach surface waters of the Gulf of Gabès *via* atmospheric
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561031 transport over long distances from the Gabès city and other areas. Interestingly, the stations
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581032 located either in front of (S7-S9, S23) or close to (S22, S24) Ghannouch industrial
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1033 complex/commercial harbour and phosphogypsum effluent outlet (Fig. 1, Table S1) presented
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2 1034 dissolved PAHs mainly of pyrogenic (combustion) origin, as seen from their Ant/(Ant+Phe)
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5 1035 (0.12-0.21) and Flu/(Flu+Pyr) ratios (0.43 and 0.90) (Table 1, Table S4). Moreover, these
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7 1036 stations (S7-S9, S22-S24) were marked by higher concentrations of total dissolved PAHs
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10 1037 (19.5-71.2 ng L⁻¹; mean and median: 49 and 56 ng L⁻¹) compared to the other stations (17.6-
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12 1038 59.8 ng L⁻¹; mean and median: 31 and 28 ng L⁻¹). This could be the sign of a specific PAH
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14 1039 fingerprint due to the whole Ghannouch industrial complex activity.

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17 1040 The dissolved phase was mostly enriched by PCBs displaying a low degree of chlorination,
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19 1041 i.e., tri- (CB-18, CB-28; 28% of total PCBs on average), tetra- (CB-52; 19% of total PCBs on
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22 1042 average), penta- (CB-101; 19% of total PCBs on average), and hexa-CBs (CB-153; 19% of
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24 1043 total PCBs on average) (Table 1, Fig. 4b), which is in agreement with previous observations
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27 1044 and may be explained by their higher solubility in water compared to PCBs of higher degree
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29 1045 of chlorination (Li et al., 2016; Montuori et al., 2020; Moret et al., 2005). Less chlorinated
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32 1046 PCBs are more amenable to long range atmospheric transport than heavier ones (Wania and
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34 1047 MacKay, 1995). This suggests that atmospheric transport could be the most important PCB
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36 1048 source in the studied region. Furthermore, Tri-CBs, which are mainly used as an insulating
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39 1049 liquid for transformer oils and power capacitors (Tang et al., 2020), can leaked into the soil
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41 1050 and end up in surface waters *via* surface runoff and land use (Duan et al., 2013). Surprisingly,
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44 1051 CB-180 which is a heptachloro-congener, characterized by a higher number of chlorine atoms
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46 1052 (and thus a lower solubility in water), was one of the most abundant PCBs in our samples
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49 1053 (27% of total PCBs on average). Generally, CB-180 is not detected or detected at very low
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51 1054 levels in the dissolved phase of natural waters (Montuori et al., 2020; Moret et al., 2005;
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53 1055 Rabodonirina et al., 2015). Indicator PCBs were detected in the surface waters of the Gulf of
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56 1056 Gabès with detection frequency varied from 19 to 100%, and concentrations ranged from 1.8
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58 1057 to 28.4 ng L⁻¹. The levels of Σ7 IN-PCBs were strongly correlated with total PCBs (Σ20
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1058 PCBs) (Pearson $r = 0.97$, $p < 0.0001$) and a conversion factor of 4 can be used to estimate
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21059 total PCB concentrations ($\Sigma 209$ PCBs) from 7 IN-PCBs (i.e., $\Sigma 209$ PCBs = 4×7 IN-PCBs).
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51060 Similar conversion factor was applied by previous studies for sediment and road dust (Anh et
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71061 al., 2019; Hoai et al., 2010). PCBs are used in industry as a heat exchanger liquid in
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101062 transformers and capacitors, and as additives in paints, carbonless copy papers and plastics.
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121063 As in numerous other countries, equipment containing PCBs have been widely used in
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141064 Tunisia from the 1970s to late 1980s (an import ban of those equipment being occurred in
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171065 1986). Generally, sources of PCBs in marine environment are wastes of electrical
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191066 transformers, oil spillage, and any historical use of PCB (Mitra et al., 2019). Recently, PCBs
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221067 have been recorded in Tunisian (Bizerte) coastal areas: in surface sediments, mussels, fishes
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241068 and atmospheric particles (Barhoumi et al., 2014a, b, c; Barhoumi et al., 2018; Ben Ameur et
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271069 al., 2013). In these compartments (sediments, fishes, atmosphere), the presence of tetrachloro-
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291070 , pentachloro-, hexachloro- and even heptachloro-congeners was consistent with a
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321071 contribution of commercial (Aroclor) PCB mixtures (Takasuga et al., 2006). In the same way,
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341072 our PCB molecular distribution may reflect the influence of commercial PCB mixtures.

361073 Various approaches based on the ratio of parent DDTs and their metabolites have been
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391074 used to assess whether the DDT contamination in a given area can be considered recent, and
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411075 whether the degradation of DDTs occurred rather under aerobic or anaerobic condition (Hitch
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441076 and Day, 1992; Hong et al., 2003; Lee et al., 2001; Liu et al., 2008) (Table S4). Here, since
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461077 parent DDTs were not detected at all in our samples, the ratios (DDE+DDD)/ Σ DDTs were
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491078 always equal to 1. This point outs the historical/ancient character of the DDT contamination
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511079 in the Gulf of Gabès. Moreover, more than half of stations displayed DDD/DDE ratio < 1
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541080 (Table 1), which suggests that DDT was rather biotransformed into DDE under aerobic
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561081 conditions (Galanopoulou et al., 2005; Mostafa et al., 2007). This is in accordance with the
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581082 oxidizing conditions prevailing in surface waters of the Gulf of Gabès, as indicated by O_2

1083 concentrations and redox values (Table S5). Therefore, our results indicate that DDTs origin
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2 1084 from historical load since only their metabolites have been detected. Prior to their ban in
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5 1085 Tunisia early 1984s (APEK, 2005), DDTs were largely used as pesticides in agriculture in the
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7 1086 environment of the Gulf of Gabès, thus the current flux may come from places where they
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10 1087 have remained *via* runoff and wadis or the sediment resuspension. The highest DDT
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12 1088 concentrations measured at stations S4 (12.1 ng L⁻¹), S5 (11.35 ng L⁻¹), S6 (13.23 ng L⁻¹) and
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14 1089 S19 (11.5 ng L⁻¹) located in front of El Demna and El Melah wadis (Northern Ghannouch),
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17 1090 and station S16 (11.66 ng L⁻¹) located in front of Mersit wadi (Southern Gabès), may confirms
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19 1091 the last statement. In addition, antifouling paints used in boat maintenance could be another
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22 1092 source of DDTs in surface water of the Gulf of Gabès, evidenced by the high DDT
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24 1093 concentration measured at station S8 (11.97 ng L⁻¹) located in front of Ghannouch
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26
27 1094 harbour/phosphogypsum discharges.

29 1095 HCHs usually have two formulations: technical-grade (predominantly constituted by α -
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31 1096 HCH) and lindane (> 99% γ -HCH) (Iwata et al., 1993). The isomer ratio α -/ γ -HCH can be
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33
34 1097 used as an indicator of the HCH sources, i.e., technical HCH has α -HCH/ γ -HCH ratios
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36 1098 ranging between 3 and 7, whereas those of lindane are < 1.0 or close to 1.0 (Willett et al.,
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38
39 1099 1998). α -HCH was found in only three sites, in the Gulf of Gabès, whereas γ -HCH was
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41 1100 detected at all sites (Table S6), thus yielding ratios lower than technical HCHs. This reflects
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43
44 1101 the fact that HCHs were predominantly of lindane origin. Lindane was usually used in
45
46 1102 agriculture for seed treatment for beans, barley and wheat, as well as a lesser degree as a
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48
49 1103 pharmaceutical to treat lice and scabies (Barhoumi et al., 2016). Given their high vapour
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51 1104 pressure (Shunthirasingham et al., 2010), γ -HCH was probably entered the Gulf *via*
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54 1105 atmospheric transport. Additionally, high detection frequency (85%) of β -HCH was observed,
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56 1106 suggested that HCHs were applied in these areas in the past and had degraded, because β -
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58 1107 HCH is more stable than other HCH isomers and accumulates in the environment over time
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1108 (Malik et al., 2009). Direct discharges, surface runoff from surrounding agricultural soils,
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2 1109 along with long-range transport may account for the HCHs pollution in this area.
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5 1110 As shown in Fig. 4c, DDTs and HCHs displayed different patterns. This could indicate
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7 1111 different types of human activities (agriculture) prevailed in the area, as well as different input
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10 1112 sources, e.g., wet and dry deposition sources, surface runoff, and atmospheric transport. DDT
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12 1113 and HCH patterns could be also influenced by the physico-chemical properties of isomer
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14 1114 compounds along with environmental conditions. In addition, redox potential, topography,
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17 1115 and hydrodynamic condition together with other factors play important roles in controlling the
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19 1116 distribution and fate of DDTs and HCHs (Su et al., 2006).
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22 1117 ***4.3. Potential influence of the industrial effluent and ecological risk assessment*** 23

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25 1118 Our results show a significant influence of the phosphogypsum effluent on the local
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28 1119 physico-chemistry and biogeochemistry, with the highest values of salinity, FDOM, O₂, SPM,
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30 1120 POC, PON, and the lowest values of pH observed in the most coastal stations located between
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32 1121 the Ghannouch and Gabès harbours (S23-S25; Fig. 1), and the opposite pattern (i.e., the
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34
35 1122 lowest values of salinity, FDOM, O₂, SPM, POC, PON, and the highest values of pH) found
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37 1123 in the Northern and offshore sites, leading to marked coast-offshore and South-North
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40 1124 gradients (Fig. 2, Table S5). Hence, the influence of the phosphogypsum effluent mostly
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42 1125 appeared South of the Ghannouch industrial complex, in the inter-harbour (Ghannouch-
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45 1126 Gabès) zone. These observations are in accordance with recent works, which pointed out a
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47 1127 substantial effect of the Ghannouch phosphogypsum effluent on seawater or surface sediment
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50 1128 chemistry, with lower pH values and higher concentrations in various major, trace and metal
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52 1129 elements in stations situated close to/South of the discharge outlet, and in the inter-harbour
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54 1130 area (El Kateb et al., 2018; El Zrelli et al., 2015, 2018). The fact that the phosphogypsum
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57 1131 followed a North to South direction, and accumulate in the area between Ghannouch and
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59 1132 Gabès harbours is consistent with results from Zayen et al. (2020), who reported, during our
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1133 sampling period and in our sampling area, the occurrence of a tidally-averaged coastal current
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21134 flowing from the North to the South, very likely in relation with wind and the regional
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51135 current. Dissolved organic contaminants (PAHs, PCBs and OCPs) did not closely follow the
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71136 spatial distribution of salinity, FDOM, O₂, SPM, POC and PON (Fig. 3), neither were
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101137 correlated to these parameters as seen from the PCA (Fig. 5). They displayed less marked
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121138 coast-offshore and North-South gradients (particularly for PCBs and DDTs), although PAHs
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141139 and HCHs presented higher concentrations in the Ghannouch/effluent zone (Fig. 3). This was
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171140 due to the fact that dissolved PAHs, PCBs and OCPs in coastal surface waters of the Gulf of
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191141 Gabès very likely originated from various sources including probably the phosphogypsum
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221142 effluent but also many others as industrial effluents, wadis, runoffs, sediment resuspension,
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241143 and the atmosphere with aerosol deposition and gas exchanges (Barhoumi et al., 2018; Fourati
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271144 et al., 2018; Zouch et al., 2018).

291145 Concentrations of the targeted PAH, PCB and OCP molecules in surface waters of the Gulf
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321146 of Gabès were generally low. As a consequence of their environmental persistence,
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341147 bioaccumulation and toxicity, the continued exposure to these POPs can pose adverse effects
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361148 to animals and human health, even at very low levels (Kim et al., 2018). Therefore, the
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391149 potential risk of those chemicals on aquatic organisms was evaluated. TEFs were applied here
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411150 to quantify the current mixture of PAHs carcinogenicity in seawater samples (Table 2). The
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441151 results show that in terms of total TEQ_{ΣPAHs}, the measured values were found in the range
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461152 between 0.04 and 0.51 ng TEQ L⁻¹ (mean of 0.14 ng TEQ L⁻¹) which were lower than the
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491153 threshold values established for BaP in water, suggesting that at the ongoing concentrations
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511154 the evaluated mixture does not seem able to start carcinogenic responses in aquatic organisms
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531155 (Abdel-Shafy and Mansour, 2016). TEQ_{ΣPAHs} observed in surface waters of the Gulf of
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561156 Gabès is comparable to that found in the Atlantic Iberian northwest coastline (Rocha and
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581157 Rocha, 2021).

1158 It is also important to evaluate if the measured concentrations of PAHs pose ecological
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21159 risks. As shown in [Table 2](#), $RQ_{(MPCs)}$ of individual PAHs in waters from the study area were
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51160 all < 1 , indicating relatively low ecosystem risk ([Cao et al., 2010](#)). $RQ_{(NCs)}$ of Chr was < 1
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71161 indicating almost no risk to the ecosystem, whereas $RQ_{(NCs)}$ of the other PAHs was > 1 in at
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101162 least 7% of samples indicating moderate risks. Concerning the mix of the twelve PAHs
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121163 ($RQ_{\Sigma PAHs}$), as the value of $RQ_{\Sigma PAHs(NCs)}$ are ≥ 1 or < 800 and those of $RQ_{\Sigma PAHs(MPCs)}$ are ≈ 0 , it
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141164 is concluded that, overall, the analyzed PAHs have a low risk to aquatic organisms.

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171165 Regarding organochlorines, it can be seen that all RQ_{CMCs} values for individual OCPs were
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191166 < 1 and $RQ_{CCCs} > 1$ suggesting that risk from measured OCPs is moderate to severe, however,
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221167 RQ_{CCCs} for PCBs are < 1 indicating a low risk ([Table 3](#)).

241168 25 26 271169 **5. Conclusion**

291170 The results obtained in this study provided the first detailed analysis of dissolved PAH and
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321171 OCs (including PCBs and OCPs) distributions in surface water collected from 27 stations in
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341172 the Gulf of Gabès (Tunisia). PAH levels were lower than those found in other coastal waters
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361173 around the world, while OC levels were moderated. The tendency that emerges is that the
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391174 concentrations we measured in this work in the Ghannouch-Gabès area (Southern Gulf of
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411175 Gabès) were in the mid-range of those reported in other coastal environments. They displayed
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441176 less marked coast-offshore and North-South gradients (particularly for PCBs and DDTs),
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461177 although PAHs and HCHs presented higher concentrations in the Ghannouch/effluent zone.
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491178 Risk assessment based on toxic equivalents (TEQs) indicated that DDTs could induce the
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511179 highest risks for benthic organisms, whereas ecological risks from PAHs and PCBs were
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541180 potentially lower. This work provides baseline information for further studies about this Gulf.
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561181 Since PAHs and OCs levels were only studied in autumn, more studies throughout the whole

1182 year with increasing samples sizes are needed to fully understand the levels of PAHs and OCs
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21183 and the ecological risk assessment of this area.
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13
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171188 **CRedit authorship contribution statement**

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21
221190 Investigation, Methodology, Data curation, Writing - original draft, and Writing - review &
23
24
251191 editing. **Lobna Jlaiel:** Methodology and Data curation. **Catherine Guigue:** Writing – review
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271192 & editing. **Lassaad Chouba:** Conceptualization and Data curation. **Soufiane Touil and Sami**
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29
301193 **Sayadi:** Supervision

331194 **Declaration of competing interest**

34

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1626 **Figure captions**

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5 1628 **Figure 1.** Location of the study stations in the Ghannouch-Gabès coastal area in the Gulf of
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7 1629 Gabès (Tunisia, Southern Mediterranean Sea). The detailed characteristics of the stations are
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9 1630 given in [Table 1](#).

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12 1631 **Figure 2.** Spatial distribution of a) temperature, b) salinity, c) pH, d) fluorescent dissolved
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14 1632 organic matter (FDOM) at 254-nm excitation wavelength and 360-nm emission wavelength
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17 1633 (in relative unit, RU), e) dissolved oxygen (O₂) concentration, and f) redox potential in coastal
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20 1634 surface waters (at ~ 0.5-m depth) of the Gulf of Gabès. Spatial interpolation was made using
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22 1635 Data-Interpolating Variational Analysis (DIVA) method from Ocean Data View (ODV)
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25 1636 software version 4.6.5. <http://odv.awi.de>.

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27 1637 **Figure 3.** Spatial distribution of concentrations of total dissolved a) PAHs (sum of 16
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30 1638 compounds), b) PCBs (sum of 20 congeners), c) HCHs (sum of 4 compounds), and d) DDTs
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32 1639 (sum of 6 compounds) (in ng L⁻¹) in coastal surface waters (at ~ 0.5-m depth) of the Gulf of
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35 1640 Gabès. Spatial interpolation was made using Data-Interpolating Variational Analysis (DIVA)
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37 1641 method from Ocean Data View (ODV) software version 4.6.5. <http://odv.awi.de>.

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40 1642 **Figure 4.** Relative abundance (in %) of a) 12 PAHs, b) 9 PCBs congeners, and c) 7 OCPs in
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43 1643 each station.

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45 1644 **Figure 5.** Principal component analysis (PCA), based on the Spearman 's correlation matrix,
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48 1645 applied on various parameters measured in coastal surface waters (at ~ 0.5-m depth) of the
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51 1646 Gulf of Gabès, i.e., salinity (Sal), pH, dissolved oxygen concentration (O₂), fluorescent
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53 1647 dissolved organic matter (FDOM), and concentrations of total dissolved PAHs (Σ PAHs),
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55 1648 PCBs (Σ PCBs) and OCPs (Σ OCPs). Projection of variables (green lines) and samples (blue
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58 1649 circles) on the first factorial plane (PC1 *versus* PC2). Four groups of stations have been
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60 1650 highlighted (G1-G4).

1651 **Table captions**

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4 1653 **Table 1.** Total concentrations of dissolved PAHs, PCBs, OCPs, HCHs, DDTs and associated
5 ratios from seawater samples collected at ~ 0.5-m depth in coastal area of the Gulf of Gabès
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7 1654 (Tunisia, Southern Mediterranean Sea).
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12 1656 **Table 2.** Toxic equivalence quantities (TEQs) and mean values of $RQ_{(NCs)}$ and $RQ_{(MPCs)}$ of
13 PAHs from seawater samples collected at ~ 0.5-m depth in coastal area of the Gulf of Gabès
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15 1657 (Tunisia, Southern Mediterranean Sea).
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17 1658

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20 1659 **Table 3.** Mean values of RQ (CCCs) and RQ (CMCs) for OCPs and PCBs from seawater
21 samples collected at ~ 0.5-m depth in coastal area of the Gulf of Gabès (Tunisia, Southern
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23 1660 Mediterranean Sea).
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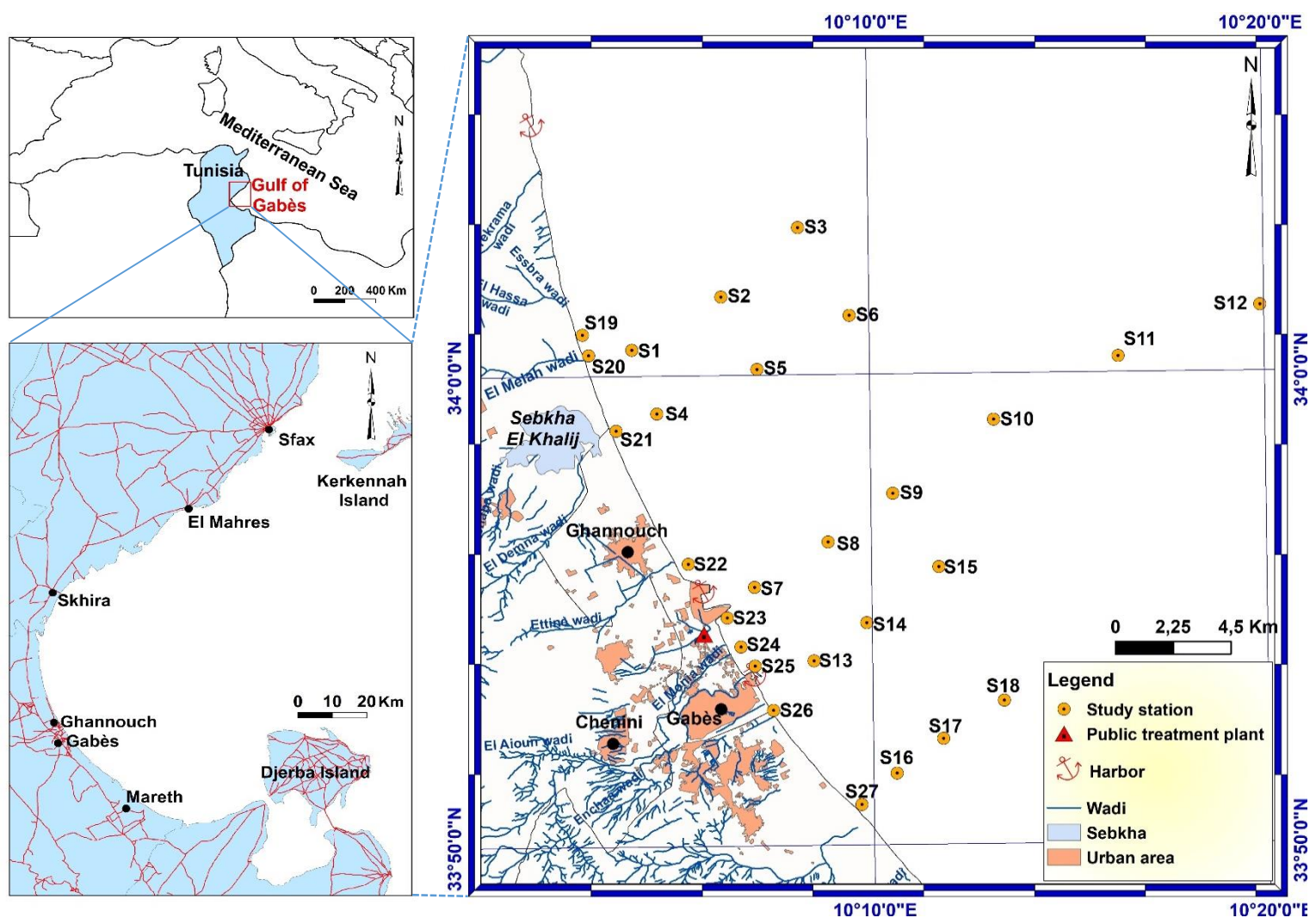
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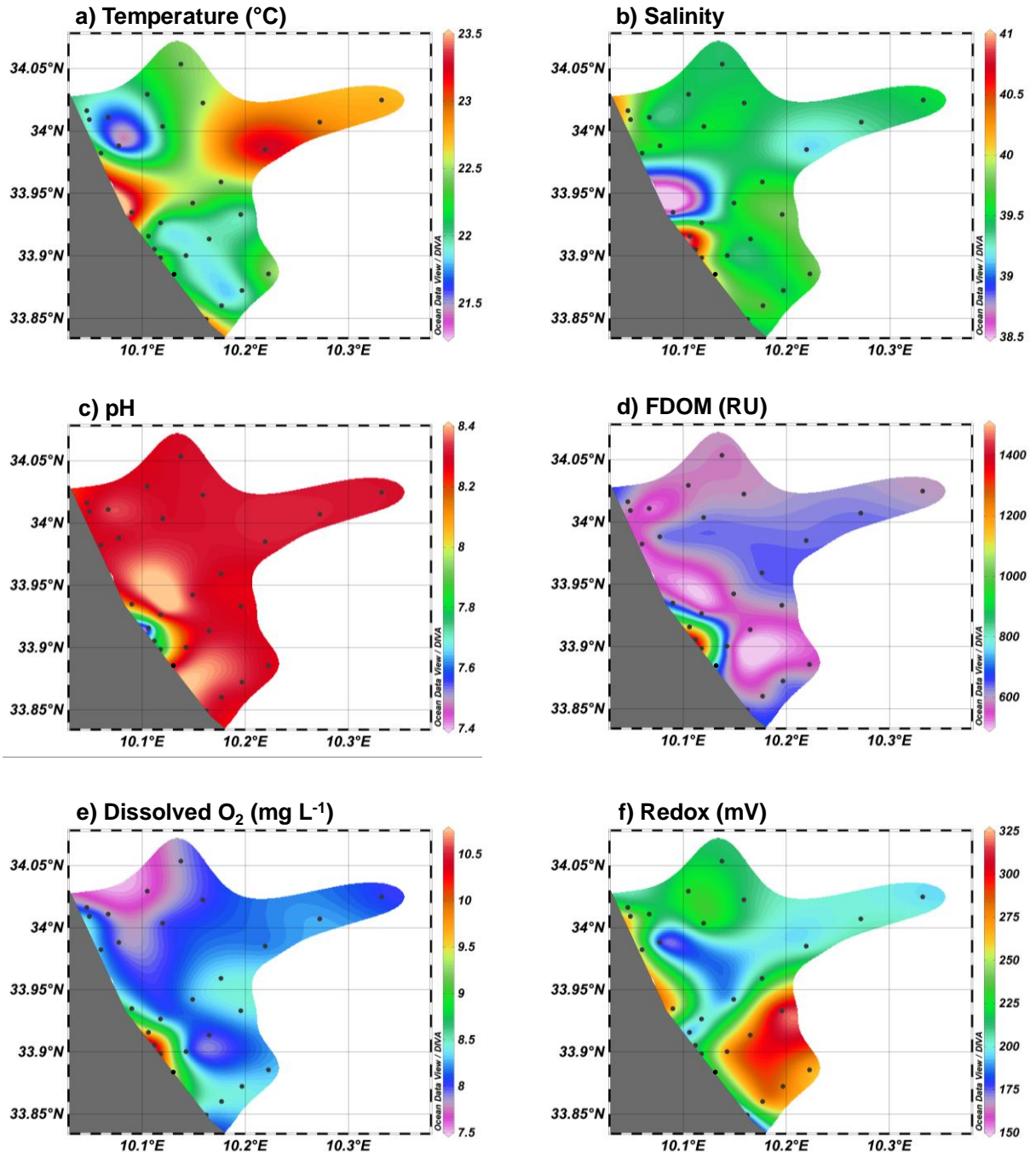
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Figure 1



1695 **Figure 2**



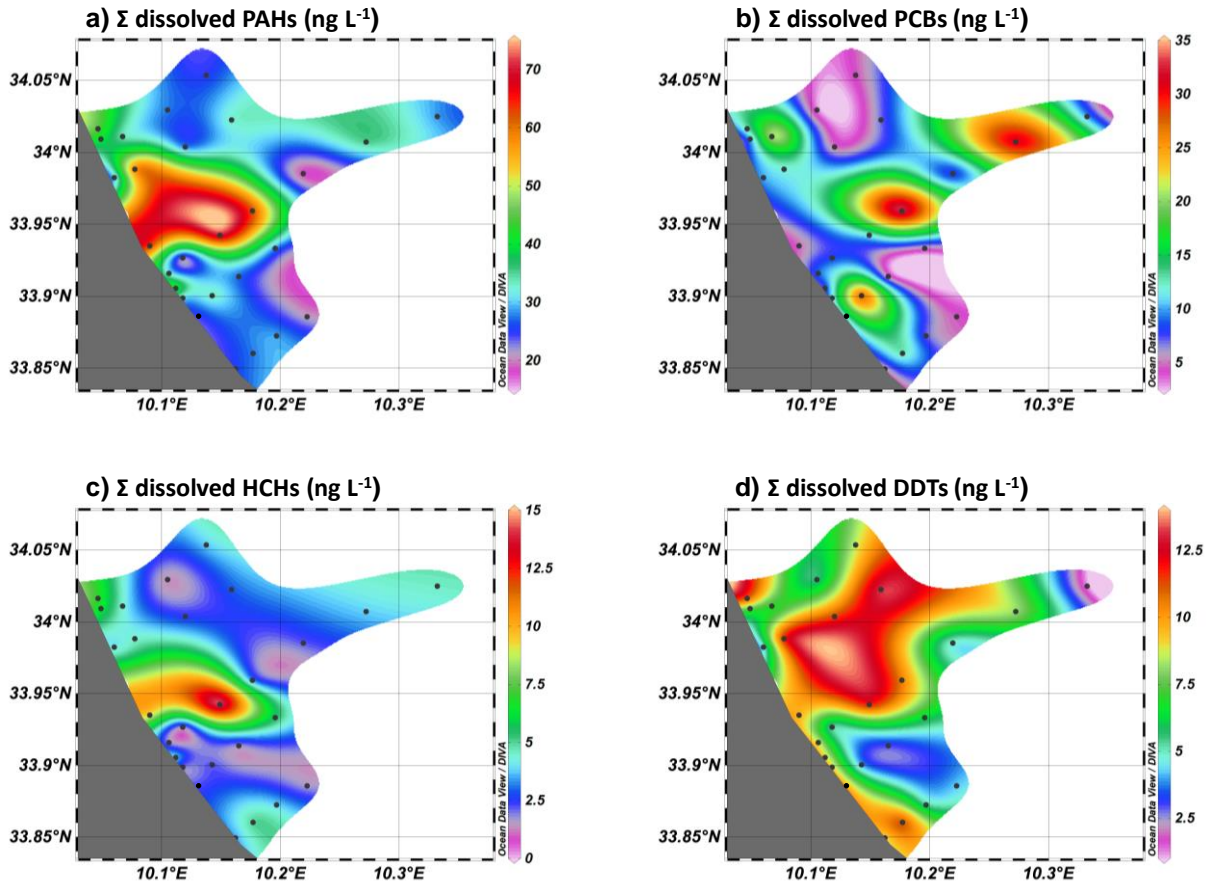
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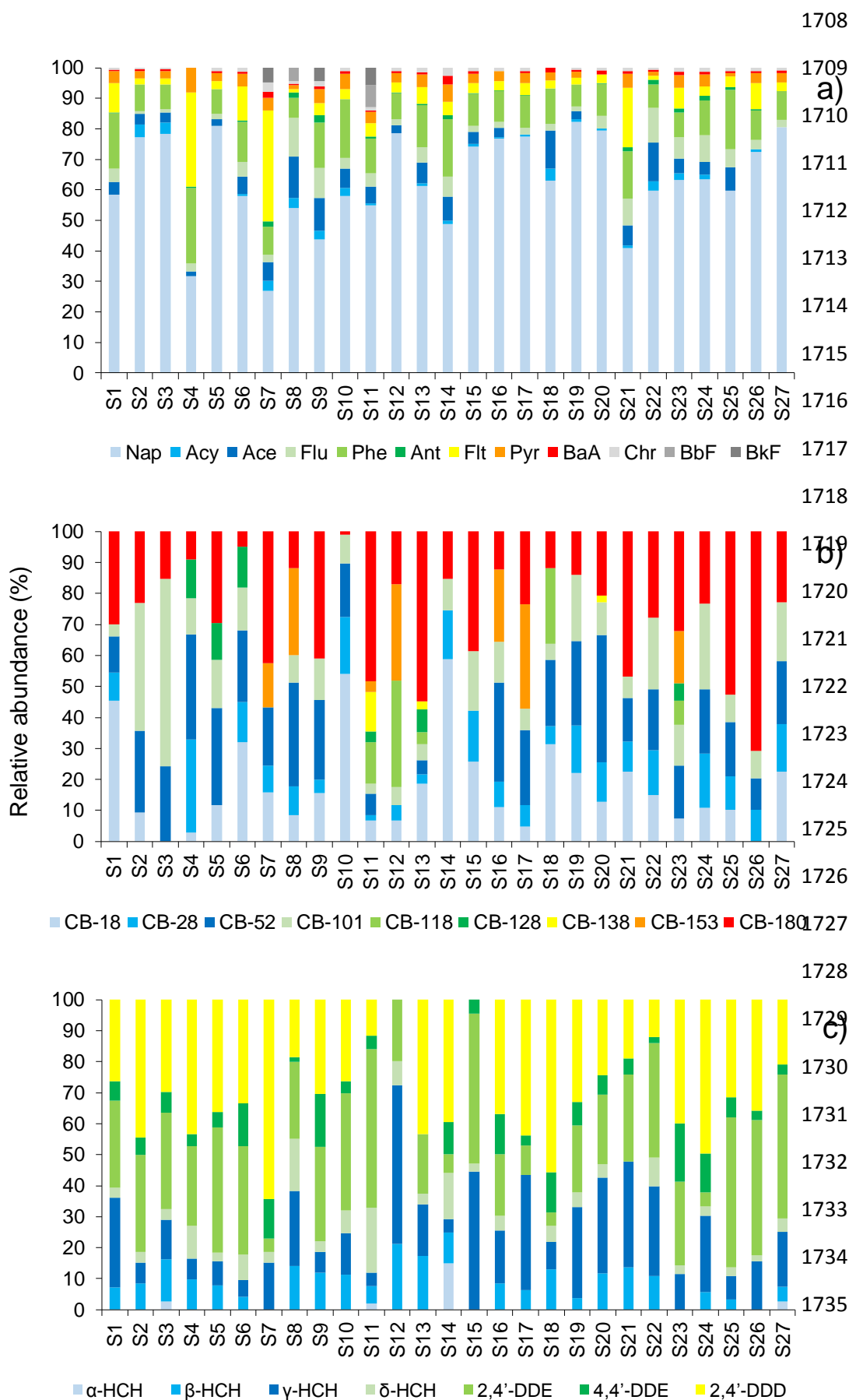
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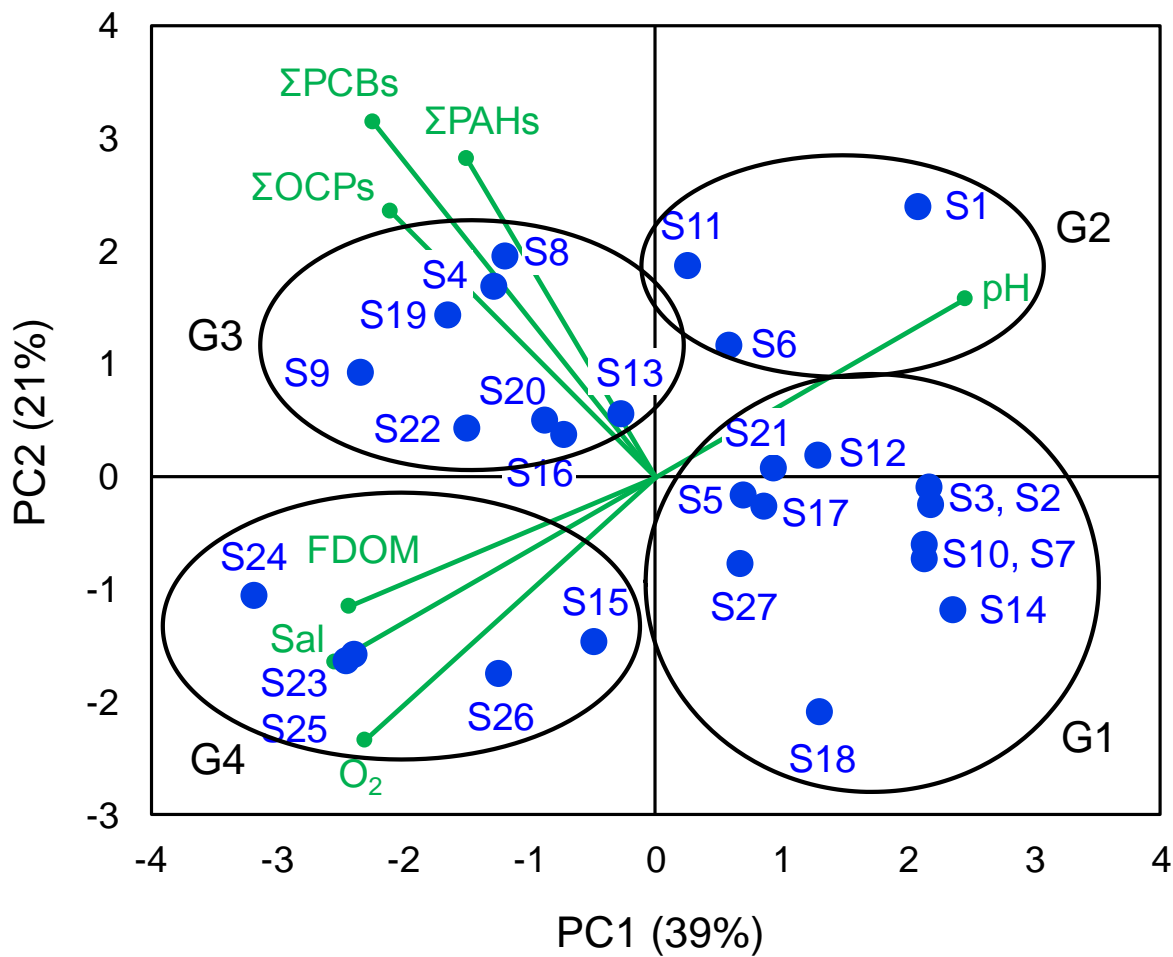
1700 **Figure 3**



1707 **Figure 4**



1736 **Figure 5**



1744 **Table 1.** Total concentrations of dissolved PAHs, PCBs, OCPs, HCHs, DDTs and associated
 1745 ratios from seawater samples collected at ~ 0.5-m depth in coastal area of the Gulf of Gabès
 1746 (Tunisia, Southern Mediterranean Sea).

Stations	PAHs			OCs				
	Σ16 PAHs (ng L ⁻¹)	Flt/(Flt+Pyr)	Ant/(Ant+Phe)	Σ20 PCBs (ng L ⁻¹)	Σ10 OCPs (ng L ⁻¹)	Σ4 HCHs (ng L ⁻¹)	Σ6 DDTs (ng L ⁻¹)	DDD/ DDE
S1	32.7	0.71	0.01	22.0	11.0	4.3	6.6	0.77
S2	27.7	0.43	nd	2.9	6.8	1.3	5.5	1.21
S3	25.0	0.43	nd	4.3	12.7	4.1	8.6	0.78
S4	59.8	0.79	0.02	11.6	16.6	4.5	12.1	1.48
S5	27.1	0.50	nd	4.6	13.9	2.6	11.3	0.80
S6	32.8	0.73	0.04	6.9	16.1	2.9	13.2	0.69
S7	19.5	0.90	0.16	8.1	6.0	1.1	4.9	3.74
S8	71.2	0.49	0.21	11.2	26.7	14.8	12.0	0.71
S9	62.7	0.47	0.15	33.6	13.5	3.0	10.5	0.64
S10	17.6	0.40	nd	7.3	6.7	2.2	4.6	0.63
S11	35.3	0.54	0.05	31.4	12.3	4.0	8.2	0.21
S12	28.3	0.52	0.04	9.6	5.6	4.5	1.1	nd
S13	33.5	0.55	0.03	28.3	6.1	2.3	3.8	2.24
S14	26.8	0.44	0.06	4.3	5.3	2.3	2.9	2.38
S15	27.5	0.51	0.03	4.7	11.8	5.6	6.2	nd
S16	27.4	0.49	0.03	13.3	16.7	5.1	11.7	1.13
S17	29.0	0.55	0.03	6.7	8.9	3.8	5.0	3.41
S18	20.6	0.48	nd	4.6	5.9	1.6	4.3	3.25
S19	45.0	0.53	nd	12.7	18.5	7.0	11.5	1.13
S20	40.5	nd	0.02	8.5	13.7	6.4	7.3	0.85
S21	27.7	0.81	0.08	10.1	8.2	3.9	4.3	0.57
S22	64.9	0.48	0.18	4.6	19.0	9.4	9.7	0.31
S23	30.9	0.61	0.15	9.4	10.4	1.5	8.9	0.87
S24	46.1	0.43	0.12	6.9	13.7	4.6	9.1	2.93
S25	28.1	0.73	0.04	10.7	10.7	1.5	9.2	0.57
S26	22.9	0.72	0.04	7.6	13.3	2.3	10.9	0.77
S27	25.3	0.47	0.02	4.8	12.3	3.6	8.7	0.42
Min	17.6	0.40	0.01	2.9	5.3	1.1	1.1	0.21
Max	71.2	0.90	0.21	33.6	26.7	14.8	13.2	3.74
Mean	34.7	0.56	0.07	10.8	11.9	4.1	7.9	1.30
Median	28.3	0.51	0.04	8.1	12.3	3.8	8.6	0.80

nd: not determined.

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Table 2. Toxic equivalence quantities (TEQs) and mean values of $RQ_{(NCs)}$ and $RQ_{(MPCs)}$ of PAHs from seawater samples collected at ~ 0.5-m depth in coastal area of the Gulf of Gabès (Tunisia, Southern Mediterranean Sea).

PAHs	TEQs (ng L ⁻¹)				RQs for PAHs								
	TEFs	Min	Max	Mean	NECs	Min	Max	Mean	$RQ_{(NCs)} > 1^*$	MPCs	Min	Max	Mean
Nap	0.001	0.005	0.039	0.021	12	0.436	3.236	1.776	93	1200	0.004	0.032	0.018
Acy	0.001	0.000	0.002	0.001	0.7	0.000	3.469	0.741	30	70	0.000	0.035	0.007
Ace	0.001	0.000	0.010	0.002	0.7	0.000	13.770	2.830	81	70	0.000	0.138	0.028
Flu	0.001	0.000	0.009	0.002	0.7	0.332	12.960	2.708	56	70	0.003	0.130	0.027
Phe	0.001	0.002	0.015	0.004	3	0.592	4.949	1.389	59	300	0.006	0.050	0.014
Ant	0.001	0.000	0.002	0.001	0.7	0.000	2.304	0.409	15	70	0.000	0.023	0.004
Flt	0.01	0.005	0.185	0.028	3	0.154	6.159	0.766	19	300	0.002	0.062	0.008
Pyr	0.001	0.000	0.005	0.001	0.7	0.000	6.849	1.654	81	70	0.000	0.069	0.017
BaA	0.1	0.000	0.078	0.027	0.1	0.000	7.771	2.655	89	10	0.000	0.078	0.027
Chr	0.01	0.000	0.011	0.004	3.4	0.000	0.311	0.104	0	340	0.000	0.003	0.001
BbF	0.1	0.000	0.310	0.030	0.1	0.000	31.000	2.082	7	10	0.000	0.310	0.021
BkF	0.1	0.000	0.277	0.029	0.4	0.000	6.926	0.531	11	40	0.000	0.069	0.005
TEQs _{∑PAHs}		0.040	0.510	0.137	$RQ_{∑PAHs(NCs)}$	6.550	73.170	17.646		$RQ_{∑PAHs(MPCs)}$	0.070	0.730	0.176

Percent of stations with $RQ_{(NCs)}$ of PAHs >1.

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1753 **Table 3.** Mean values of RQ (CCCs) and RQ (CMCs) for OCPs and PCBs from seawater
 1754 samples collected at ~ 0.5-m depth in coastal area of the Gulf of Gabès (Tunisia, Southern
 1755 Mediterranean Sea).

	γ -HCH	DDEs	DDDs	PCBs
CMCs (ng L ⁻¹)	950	1100	1100	-
CCCs (ng L ⁻¹)	-	1	1	4
MEC*	2.286	4.295	3.568	10.771
RQ _{CMCs}	0.002	0.004	0.003	-
RQ _{CCCs}	-	4.295	3.568	0.769

*Average Measured Concentration for OCPs and PCBs in this study (ng L⁻¹).