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Long-term monitoring emphasizes impacts of the dredging on dissolved Cu and Pb contamination along with ultraplankton distribution and structure in Toulon Bay (NW Mediterranean Sea, France)

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Abstract

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2 A long-term monitoring during dredging and non-dredging periods was performed.
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4 Total and dissolved Cu and Pb concentrations, DGT-labile Pb, ultraphytoplankton abundance
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6 and structure were monitored at four sites: dredging site, dumping site (inside/outside of a
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8 geotextile bag) and reference site. During the reference period (non-dredging), an increasing
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10 contamination in Pb, Cu and a progressive shift from *Synechococcus* to photosynthetic
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12 picoeukaryotes dominance was observed from reference to dumping site. Pb concentrations
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14 were significantly higher during dredging period, pointing out sediment resuspension as Pb
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16 major source of contamination. Unlike Pb, Cu concentrations were not statistically different
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18 during the two periods. Dredging period did not impact on ultraphytoplankton abundance and
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20 structure but influence heterotrophic prokaryotes abundance. Sediment resuspension is
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22 therefore a major driver of chemical and biological qualities in Toulon Bay. Furthermore,
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24 although the geotextile bag reduces particulate transport of the dredged sediment, the
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26 transport in the dissolved phase remains a major problem.
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36 **Keywords:** sediment resuspension, dissolved metals, Chelex-DGT samplers, Toulon Bay
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1. Introduction

Coastal areas are of great interest for Humanity. Harbors, located at the continent-ocean interface, represent hotspots of copper (Cu) contamination on the coast, both in water column and sediment (Cindrić et al., 2015; Coclet et al., 2019, 2018; Tessier et al., 2011). Copper contamination from harbors has been proved to be highly toxic for planktonic microorganisms (Coclet et al., 2020, 2018). Lead (Pb) contamination is mostly archived in harbors sediment, often originating from older sources such as coal combustion, battery factories, metal smelting, leaded gasoline. However, Pb can be remobilized into the water column and be transferred to aquatic resources consumed by humans (Dang et al., 2015).

Due to sedimentation, harbors become less accessible and therefore need to be dredged to preserve or increase their level of activity. During dredging activities, sediment can become a major source of chemical contamination for the water column (Torres et al., 2009). For example, sediment resuspension could lead to an important release of metals in the water column, both in the particulate and dissolved phases (Dang et al., 2015).

The ecologically-meaningful planktonic microbial realm responds quickly to dredging and more generally to sediment resuspension. Thus, it represents a useful tool to evaluate the extent of the impact of such human-induced perturbations on the ecosystem. While physiological responses have been identified for long (Borgmann and Munawar, 1989; Munawar et al., 1985; Nayar et al., 2004) and suggest the stimulation of heterotrophic growth by increased resource availability, community structure shifts have scarcely been studied with respect to this specific kind of perturbation. Recent experimental work suggested that sediment resuspension could be responsible of large changes in ultraphytoplanktonic groups' relative abundances (Coclet et al., 2018). However, the *in situ* response of ultraphytoplankton to dredging operations monitored remains to be evaluated.

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The influence of an 18-month long dredging operation on Cu and Pb contamination and ultraplankton structure and abundance was studied in Toulon Bay (NW Mediterranean Sea, France). Due to historical (World War II) as well as current activities, Toulon Bay is subjected to strong anthropogenic pressure resulting in a strong multi-contamination in metals (Tessier et al., 2011), metalloids (Dang et al., 2014), organo-metals (Pougnnet et al., 2014) and organic contaminants (Misson et al., 2016). On the other hand, natural variations commonly observed in other coastal areas, such as salinity, appear to be of relatively low amplitude (Coclet et al., 2019, 2018). A gradient of distribution and structure of ultraplankton have been recently observed (Coclet et al., 2019, 2018) correlated with the already-known chemical contamination.

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Through sediment resuspension, we hypothesized that this dredging operation would lead to a general increase in Cu and Pb contamination in the particulate fraction, to an increase in dissolved Pb, to a development of heterotrophic prokaryotes and to a strong dominance of eukaryotic ultraphytoplankton in the bay. The spatial extent of this phenomenon could be limited by efforts made to contain suspended particles, and its significance could be challenged by the frequency of other human-induced sediment resuspension events in this harbor. In order to test this hypothesis, a long-term monitoring was performed. The dredging site, the dumping site and a control reference site located close to an aquaculture area were monthly monitored. Discrete water sampling was used to evaluate ultraplankton abundance as well as Cu and Pb concentrations partitioning between the dissolved phase and the particles. A continuous weekly monitoring of labile Pb was also weekly conducted using DGT samplers.

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In addition to focusing on the dredging period, common spatial and temporal variations of the different parameters were recorded up to 1 year before dredging and for almost 2 years afterwards, in order to be able to describe a reference period.

2. Experimental

2.1. Dredging operation and sampling sites

A long-term monitoring covering dredging and non-dredging periods was performed in Toulon Bay, known to be highly contaminated in both sediment and water compartments (Coclet et al., 2018; Tessier et al., 2011).

The dredging operation consisted in sediment suction (manually driven) at the dredging site (DS) located in the northern part of Small Bay, in an area well exposed to water currents (Figure 1). Dredged material was stored underwater in a geotextile bag of ~ 60 x 20 m at a dumping site (IDuS) located ~ 800 m away from DS, in an enclosed docking area. This bag was supposed to retain particles, but it was water permeable. Dredging operation occurred from September 2014 until January 2016. Once dredging operations were complete, the geotextile bag and the sediment it contained were removed from the dock and transferred to land for treatment of dredge spoil.

In order to evaluate the impact at short distance and taking into account a potential difference between DS and IDuS, a third site (ODuS), located few meters away from IDuS in the same enclosed dock was also sampled. Furthermore, a control reference site (RS) located 3 km away from DS, in the southern part of Small Bay (Figure 1), close to the aquaculture area was studied. DS was monitored to evaluate the potential impact of the dredging at the dredging site. IDuS was studied to assess the supposed impact of sediment resuspension. ODuS was examined to measure the impact of sediment resuspension at short distance, as well as the effectiveness of the geotextile bag in relation to both particulate and dissolved contamination export. Concerning RS, the authors are aware that it is not a pristine site since it is near the aquaculture. Nevertheless, it was considered as a reference site since it is closed to the main water renewal channel, with a huge influence of the open sea water. These four sites were monthly monitored.

2.2. Seawater sampling and samples pretreatment

Seawater was monthly sampled from September 2013 to November 2017. The monitoring of ODuS only started in July 2014. Samples were collected at 5m depth using a 4.2 L horizontal van Dorn type water sampler (Wildco, model Beta) previously acid-cleaned (10 % v/v HNO₃, pro analysis, Fisher Scientific) and fully rinsed with Milli-Q water (18.2 MΩ, Millipore) in the laboratory. On field, the water sampler was washed with acidified Milli-Q water (0.1 % v/v HCl, TraceSelect, Fluka) and then rinsed three times with seawater from the site to be sampled. One liter of seawater was stored in fluorinated ethylene propylene (FEP) bottles (Nalgene), which had previously been carefully cleaned (10 % HNO₃ (Fisher Scientific Pro), rinsed three times with Milli-Q water, filled with 0.1 % v/v HCl (TraceSelect, Fluka) and rinsed three times with seawater from the site. Back to the laboratory, these 1 L samples were immediately split under a metal-clean laminar hood: (i) one part to analyze the acid leachable metals concentrations, and (2) another part filtered over 0.2 μm (cellulose acetate syringe filter (Sartorius) pre-rinsed and conditioned following the same protocol as described for 1 L FEP bottles). To avoid adsorption onto vessels, each fraction was stored into metals clean FEP bottles. All samples (acid leachable samples and filtered samples) were then acidified with 0.2 % v/v HNO₃ s.p (Merck) before being UV-irradiated (150-W mercury lamp, Hanau, Germany) for at least 24h in order to completely decompose organic matter (Omanović et al., 2006) and assess the concentration of metals by stripping voltammetry. The total acid leachable metals concentration will further be referred as total concentrations (Cu_T and Pb_T) in this manuscript.

In addition, from January 2015 to November 2017, 10 mL seawater samples were filtered on field through a 25 mm Swinnex filter holder (Millipore, USA) equipped with a 90

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μm nylon mesh. The filtrate was fixed with 0.25 % glutaraldehyde (final concentration) and frozen at -80°C until flow cytometry analyses.

Because of the heterogeneity of the monitoring period for the biological and chemical analyses, and the absence of biological data before the beginning of the dredging operation, the data will not be presented in 3 periods representing the situation before, during and after the dredging but only in 2 periods representing on the one hand the dredging period and on the other hand the reference period, which includes all the other sampling dates.

The number of samples analyzed as well as the beginning and the end of each type of sampling are summarized in Supplementary Information 1.

2.3. DGT passive sampling

In addition to discrete water sampling, DGT passive samplers were deployed at three sites (DS, ODuS, RS) in order to constantly monitor time-weighted average concentration of Pb. It was not possible to deploy DGT sampler at IDuS because of the inability to suspend a mooring line which was not in contact with the geotextile (larger risk of fouling). Open pore Chelex-DGT samplers (DGT Research, UK) were deployed in triplicates on a weekly basis. After 7 days of deployment, DGT samplers were removed from the holder, externally rinsed with Milli-Q water on field to remove the excess of salt water, stored in plastic bags and thereafter processed in the laboratory. Chelex resin gel with accumulated Pb was removed under laminar hood with cleaned Teflon clamp, which was finally soaked in 1 M HNO_3 s.p. (Carlo Erba Normatom II quality) solution during 24h.

2.4. Chemical analyses

Determination of DGT-labile Pb concentration in DGT eluates was performed in undiluted samples using an Atomic Absorption Spectrometer (Varian, AA 240Z) coupled

1 with a graphite furnace (Varian, GTA120). The emission wavelength was set to 217 nm. For
2 each measurement, 20 μL of sample were injected with 5 μL of a matrix modifier (composed
3 of palladium nitrate and magnesium nitrate hexahydrate). Since DGT deployment was
4 dedicated to legal monitoring of Pb contamination by French Navy, concentrations were
5 determined by the Navy's laboratory (LASEM Toulon). Calculation of DGT-labile Pb
6 concentrations (referred to as Pb_L in the figures) was performed using diffusion coefficients
7 (corrected for temperature average during the deployment period) supplied by DGT-Research
8 company. The standard material used to test AAS recovery was an ICP multi-element
9 standard VI CERTIPUR Certified Material by MERCK. The recovery was $99 \pm 7.7 \%$. It
10 should be underlined that DGT blanks were also analyzed each time and blanks
11 concentrations were subtracted from sample results. Note that Pb concentration in DGT blank
12 was always less than 10 % of the measured Pb in samples. All Pb results were above the limit
13 of quantification (LOQ) value (9.6 nM).
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31 The dissolved and total concentrations of Cu and Pb were determined by Differential Pulse
32 Anodic Stripping Voltammetry (DPASV). The measurements were carried out using an AutolabIII
33 potentiostat (EcoChemie) equipped with an autosampler. A three-electrode system (663 VA Stand,
34 Metrohm) consisting of Hg working electrode (size 3), Ag/AgCl/3M KCl reference electrode and Pt
35 counter electrode was used for measurements. Deposition potential was -0.8 V during 600 s. The
36 initial and final scanning potential were set to -0.75 to 0.02 V . The peak positions were $-0.15 \pm 0.03 \text{ V}$
37 and $-0.37 \pm 0.03 \text{ V}$ for Cu and Pb respectively. Automated measurements were controlled by the
38 GPES 4.9 software (EcoChemie). Quantification of the metals concentrations was performed using the
39 standard additions method, automated using syringe burettes (Pump XE 1000, TECAN).
40 Voltammograms were processed (peak height of 2nd derivative as analytical signal characteristic)
41 using ECDSOFT software (Omanović and Branica, 1998). Additional information can be find
42 elsewhere (Cindrić et al., 2015; Louis et al., 2009; Oursel et al., 2013). All Pb and Cu results were
43 above the limit of detection (LOD) value (0.004 and 0.04 nM, respectively).
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2.5. Ultraplankton characterization and enumeration

Autotrophic prokaryotes (Synechococcus-like SYN) and photosynthetic eukaryotes were characterized and enumerated using a BD Accury™ C6 (BD Biosciences) flow cytometer, as previously described (Coclet et al., 2018; Marie et al., 2014). The differentiation between pico- and nano-planktonic cells was performed with the help of non-fluorescent size calibration beads (ThermoFisher). One photosynthetic picoeukaryotic groups (PPE) and two photosynthetic nanoeukaryotic groups (PNE) were differentiated. PNE were indexed according to their autofluorescence into highly red fluorescent photosynthetic nanoeukaryotes (HR PNE) and highly orange fluorescent photosynthetic nanoeukaryotes (HO PNE, corresponding to putative cryptophytes). Free-living heterotrophic prokaryotes (HP) were enumerated after staining with SYBR Green as previously described (Cabrol et al., 2017).

2.6. Statistical analyses

Differences in metals concentrations or in microbial groups' abundances were statistically tested through Kruskal-Wallis tests and Tukey post-hoc comparisons. These analyses were performed by using RStudio (RStudio team, packages *stats* and *PMCMR*).

In order to evaluate the links between dissolved and total concentrations of metals, Spearman's correlation analyses were performed using a homemade software named Corrdata (developed by Dr. Omanović).

2.7. Meteorological data

In order to evaluate the potential influence of wind onto Cu and Pb concentration in the Bay, wind characteristics were collected from a nearby weather station (Toulon – La Mitre station, 43.104065 N, 5.931631E). Daily average wind intensity and direction were

1 recovered from Meteo France. More information on the treatment of these data and on the
2 comparison with Cu and Pb data is available in Supplementary Information SI 2.
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6 7 3. Results and discussion

8 9 3.1. Metal concentrations in the Bay during the reference period

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11 A north-south decreasing trend (i.e., IDuS, ODuS, DS, RS) of both total and dissolved
12 metal concentrations was observed in the Bay for the non-dredging period (Figure 2), as
13 previously observed either punctually or during a one-month monitoring (Coclet et al., 2019,
14 2018). Cu_T and Pb_T as well as Cu_D (dissolved Cu) and Pb_D (dissolved Pb) concentrations were
15 higher in the northern part of the Bay with a 4- or 5- fold increase in ODuS when compared to
16 RS (Table 1). It should be underlined that Pb_T increased in a higher extend than Cu_T . The
17 present study clearly demonstrates that this spatial trend of water column contamination
18 remains stable at a pluri-annual time scale (Figure 3). For all sampling dates, Pb_D was higher
19 at ODuS than DS, and concentration at RS were always the lowest. Concerning Pb_T , Cu_D and
20 Cu_T this observation was true 20 times out of 21. Moreover, they were always higher in RS
21 when compared to open Mediterranean seawater (Table 1). DS presented intermediate
22 concentrations between RS and ODuS (Table 1). This spatial trend clearly reflects the
23 anthropization of the northern part of the Bay in which various industrial and harbor sources
24 contributing to the contamination are located (Coclet et al., 2018; Dang et al., 2015; Tessier et
25 al., 2011). The difference between DS and ODuS could be linked to their respective
26 hydrodynamics, DS being located in a rather open part whereas ODuS was located at the
27 bottom of a very enclosed dead-end, thus submitted to limited water exchanges. Furthermore,
28 the enclosed dock of ODuS is subjected to high maritime traffic of numerous navy boats, thus
29 potentially exposed to higher contamination by sediment resuspension (Supplementary
30 Information 3). Similar concentration gradients were observed in other studies of semi-
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1 enclosed Bays and were explained by combinations of point contaminant sources, the
2 hydrodynamics of the zone, diffusive fluxes from sediment, as well as sediment resuspension
3 events (Cindrić et al., 2015; Lares et al., 2009; Sañudo-Wilhelmy and Flegal, 1996).
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7 In the studied area, the proportion of Pb_D in the water column was steady (56 ± 3 % on
8 average), demonstrating the absence of any spatial trend in Pb partitioning between the
9 dissolved and particulate phases. On the contrary, Cu_D proportion exhibited variations among
10 sites and was lower in the northern part (68 % at DS and 70 % at ODuS) compared to RS (81
11 %). Dissolved concentrations for both metals were within the range found for anthropized
12 costal zones (Cindrić et al., 2015; Oursel et al., 2014, 2013). Furthermore, Cu_D/Pb_D ratio was
13 higher at RS (20) compared to DS and ODuS (respectively 16 and 15). Cu_T/Pb_T ratio was also
14 higher at RS (14) than in DS and ODuS (12). Due to the higher Pb_T increase (as quoted
15 before), this ratio decrease tends to point out sediment as the main source of Pb contamination
16 at every sites, resulting in a different contamination level of the water due to different
17 sediment contamination levels (Tessier et al., 2011) and/or sediment grain size distribution
18 (Wafo et al., 2016) which impacts resuspension. The contribution of a higher maritime traffic
19 in the northern part of Small Bay to the frequency of sediment resuspension remains to be
20 determined. Furthermore, a probable higher dilution of resuspended sediment and diffusive
21 flux can be hypothesized in RS compared to DS and in DS compared to ODuS according to
22 hydrodynamics, ODuS being very enclosed and RS being close to the main connection
23 between Small Bay and Large Bay.
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49 As observed for Pb_D and Pb_T concentrations, DGT-labile Pb concentrations showed a
50 decreasing trend from ODuS towards RS (Figure 2). DGT-labile Pb concentration is
51 considered as the potentially bioavailable fraction. This relation is supported by a few studies
52 which found out that the accumulated metal (including Pb) in biota are better correlated with
53 DGT-labile fraction than with dissolved fraction (Kim et al., 2016).
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The main benefit of using DGT passive samplers is to provide continuous and time integrated level of metals concentration over the deployment period. No correlation was found between DGT-labile Pb and Pb_D (Spearman's correlation test, $p > 0.05$) in each single site, whereas DGT-labile Pb and Pb_D were correlated when all sites were considered together (Spearman's correlation test, $\rho = 0.565$, $p < 0.05$). Moreover, the concentration of DGT-labile Pb for some periods was higher than the corresponding Pb_D concentration. Taken together, these observations demonstrate that monthly discrete sampling was too occasional compared to one-week-long DGT deployments and not adequate to represent the DGT-deployment period. Such discrepancy was already observed for other dynamic systems (Cindrić et al., 2017). It suggests a strong short-term variability of Pb concentration within the Bay, which is also obvious on temporal profiles of DGT-labile Pb concentrations (Figure 3). Considering that one of the main Pb sources for the water column in the Bay appears to be sediment resuspension (Dang et al., 2015), this high short-term variability could reflect the very punctual character of sediment resuspension (in both space and time). No link between Pb concentration increases and meteorological features could be highlighted during this monitoring (Figure SI2. 2) and the influence of river inputs can be neglected according to previous studies and very low discharge (Nicolau et al., 2012). As a result, it could be hypothesized that Pb contamination events of the water column by sediment resuspension occurs along with human activities rather than natural climatic events in Toulon Bay, even in the absence of any dredging operation. Further work is needed to identify which activity could be responsible. The link with the gathering of most of the maritime traffic in the northern part of the Bay should be investigated in priority.

3.2. Distribution and structure of ultraplankton during the reference period

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The abundance of heterotrophic prokaryotes (HP) ranged between 0.9×10^5 and 2.1×10^6 cells.mL⁻¹. These abundances are in the classical range generally observed for coastal Mediterranean sites (Gregori et al., 2001; Šantić et al., 2012; Suffridge et al., 2018) and of previous observations in Toulon Bay (Coclet et al., 2019). Neither a clear seasonal trend nor a temporally stable difference between sites as demonstrated for metals could be observed. Indeed, although HP abundance was sometimes enriched in the northern enclosed area when compared to the southern site as previously observed during a short-term monitoring (Coclet et al., 2019) where similar HP abundances were also regularly observed at all sites during our longer term monitoring. As a consequence, the average HP abundance was not statistically different between sites (Kruskal-Wallis tests, $p > 0.05$, Figure 4). Since this situation is quite different from the metals' contamination spatial trend, which was observed at almost all the sampling dates, the metals' contamination trend can clearly not be the main driver of HP abundance dynamics in Toulon Bay. Longer-term temporal processes such as seasonal variations in water-temperature (Fuhrman et al., 2008; Gilbert et al., 2012), resource availability (Chow et al., 2013; Gilbert et al., 2012) or biotic interactions related to the structure of both phytoplanktonic and zooplanktonic communities (Gilbert et al., 2012; Goni-Urriza et al., 2018) could represent more important drivers of HP abundance in Toulon Bay.

Similarly, range of ultraphytoplanktonic community's abundance (4.5×10^3 to 6.8×10^4 cell.mL⁻¹) was similar to other observations reported for Mediterranean coastal sites (Gregori et al., 2001; Thyssen et al., 2014) or previously in Toulon Bay (Coclet et al., 2018) and did not show any clear temporal or spatial trend. When considering the different phytoplanktonic subgroups, SYN (Synechococcus-like) and PPE (picoeukaryotes-like) were the most abundant, representing at least 70 % of the community in 95 % samples. SYN abundance demonstrated a decreasing trend towards the northern enclosed ODuS whereas PPE abundance appeared stable across sites (Figure 4). As a consequence, a notable

1 community structure shift was observed from a SYN-dominated (49.1 % on average)
2 ultraphytoplanktonic community in RS to a PPE-dominated (56.3 % on average) community
3 in ODuS. The proportion of these two most abundant ultraphytoplanktonic groups was highly
4 variable through time. However, the proportion of SYN was higher in RS than in ODuS for
5 13 of the 16 sampling dates. Similarly, the proportion of PPE was lower in RS than in ODuS
6 for 75 % sampling dates. Concerning nanophytoplanktonic groups, HR PNE (highly red
7 fluorescent photosynthetic nanoeukaryotes) represented on average 10 % of the community's
8 abundance and appeared to be slightly more abundant in ODuS than in RS. Finally, HO PNE
9 (highly orange fluorescent photosynthetic nanoeukaryotes) was the least abundant group (6 %
10 on average) and displayed no spatial trend. The spatial differences mainly driven by
11 picophytoplanktonic groups are in agreement with previous observations in Toulon Bay either
12 at high-spatial but low-temporal frequency (Coclet et al., 2018) or low-spatial and similar
13 temporal frequency (Delpy et al., 2018).

3.3. Dredging significantly affected Cu and Pb contamination in the water column at large spatial scale

34 The spatial trend of Cu and Pb concentrations during the dredging period was similar
35 to the one observed during the reference period (Figure 2). A significant increase in Pb_T, Pb_D
36 and DGT-labile Pb concentrations between the dredging period and the reference period was
37 observed in all sites (Kruskal-Wallis tests, $p < 0.05$). RS site was subjected to the highest
38 relative increases in Pb concentrations (Table 2). Van den Berg et al., (2001) and Slotton and
39 Reuter, (1995) showed that there was no increased Pb_D during their monitoring of a dredging
40 activity, hypothesizing a relatively slow oxidation of metal sulfides and a fast re-oxydation of
41 Mn and Fe which precipitate Cu and Pb under oxy-hydroxides particulate forms. Based on
42 their hypothesis, during this present monitoring it could be assumed that metal sulfides
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1 undergone a fast oxidation which was not counterbalanced by Mn and Fe re-oxidation or that
2 the amount of oxy-hydroxides formed were not sufficient to precipitate all the remobilized Pb.
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4 Contribution of the organic matter on this remobilization remains to be determined.
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7 When comparing the dredging period to the reference period, the increase in Pb_D and
8 Pb_T concentrations ranged between 0.2 and 2.28 nM for RS, DS and ODuS (Table 2). During
9 the dredging period, the increases in Pb concentrations between ODuS and IDuS were far
10 higher, with around 26 nM for Pb_D and around 356 nM for Pb_T. Since an increase of Pb
11 concentrations between the dredging and the reference period was observed, along with an
12 increase in Pb concentrations between IDuS and ODuS, it can be concluded that sediment
13 resuspension by the dredging operation increased water column Pb contamination at the scale
14 of the whole studied area. The dispersion of remobilized Pb appeared nevertheless strongly
15 limited by the use of the geotextile bag. This is in agreement with the hypothesis of sediment
16 resuspension as the major source of Pb contamination in the water column of Toulon Bay
17 previously proposed by Dang et al., (2015).
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34 Pb_D fraction was globally homogeneous during the dredging period in RS, DS and
35 ODuS (58 ± 3 % on average). It was the same to that determined during the reference period
36 (56 ± 3 % on average). However, at IDuS, Pb_D percentage dropped down to 8.1 %. This could
37 be explained by a far higher particles concentration inside the dumping site, for which Pb is
38 known to have a high affinity. Moreover, the increase in Pb_D concentrations in ODuS and at
39 the other sites suggests a Pb_D export outside of the geotextile bag. This export at rather long
40 distance is not surprising, considering the main hydrological features of the Bay as previously
41 detailed, RS being localized on the main water exchange area with the open coastal part
42 (Dufresne et al., 2018).
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55 No significant difference in Cu_T and Cu_D concentrations was observed between the
56 dredging and the reference period in all sites (Kruskal-Wallis tests, p > 0.05) (Table 2). Cu_T
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1 concentrations appeared slightly higher and highly variable in IDuS compared to ODuS
2 during the dredging period. Taken altogether, these observations indicate that sediment
3 resuspension can be a source of Cu contamination in Toulon Bay water column, but this
4 contamination is lower than that of Pb and the used geotextile bag seems efficient in limiting
5 the dispersion of Cu remobilized during the dredging operation. As observed for Pb, Cu_D
6 fraction was globally homogeneous in RS, DS and ODuS ($77 \pm 6 \%$) during the dredging
7 period and close to that determined during the reference period (around 70 %). However, at
8 IDuS, the percentage of Cu_D dropped down to 7.2 % but no significant correlation could be
9 observed between Cu_D and Cu_T. On the contrary, Cu_D was quite well correlated to Cu_T at all
10 other sites (0.961, 0.939 and 0.718 at RS, DS and ODuS respectively) during both reference
11 and dredging periods. At last, Cu_D decreased in IDuS compared to ODuS during the dredging,
12 whereas Cu_T increased (Figure 2 and Table 2). This might correspond to very important
13 quantities of suspended matter at IDuS site but, contrarily to what was observed for Pb, no
14 export of Cu_D outside of the geotextile bag could be detected. This could be explained by Cu
15 higher affinity (compared to Pb) to organic ligands, which could have trapped Cu in IDuS
16 because of more numerous organic active sites for Cu surface complexation/adsorption.
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39 Cu_D/Pb_D was higher at RS (15) than in DS (13) and ODuS (11) (Table 2). Contrarily,
40 Cu_T/Pb_T was homogeneous at RS, DS and ODuS (10 ± 0.5) and lower compared to the
41 reference period. These ratios were lower during the dredging period compared to the
42 reference period due to higher Pb concentrations during the dredging period. This observation
43 confirms the different impacts of sediment resuspension on Cu and Pb contamination of the
44 water column, in both the dissolved and particulate phases.
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54 In spite of homogeneous Cu_T/Pb_T, Cu_T and Pb_T were spatially highly variable during
55 the investigated period. It could increase up to 90-fold for Cu and to 1,600-fold for Pb
56 between RS and IDuS (Figure 3). Although suspended matter was not evaluated, such
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1 increase was likely to be due to the increase of suspended sediment particles since the
2 difference between IDuS and ODuS in average reached a 7.5 factor for Cu and a 55 factor for
3 Pb, respectively, although the two sites were only separated by a few meters and a geotextile
4 bag. In plus of being spatially variable during this period, Cu_T, Pb_T and Pb_D concentrations
5 were highly variable in time (Figure 3). As an example, for the most extreme variations at
6 IDuS, Cu_T, Pb_D or Pb_T could increase by a 35-fold, 16-fold or even 645-fold between two
7 consecutive samplings, respectively. These increases could sometimes be synchronized at all
8 sites, suggesting possible short-term water exchange between sites through hydrodynamics.
9 However, those synchronized periods were quite rare, suggesting the predominance of local
10 impact in the short-term. The significant increase of all Pb concentrations during the dredging
11 period nonetheless suggests a cumulative effect of the dredging operation in the long-term. It
12 should be noticed that there were more strong wind events during the dredging period (around
13 7 by month in average) than during the reference period (around 3 by months in average)
14 (Supplementary Information 2). Nevertheless, no co-variation was found between wind
15 direction or intensity and Cu or Pb concentration, suggesting that the wind was not
16 responsible for the high variability of those concentrations in the Bay during the dredging
17 period and emphasize the influence of the dredging onto the water column contamination.
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42 Thus, although precautions were taken to limit contaminated particles dispersion at a
43 large scale, the underwater storage of dredged sediment in a permeable compartment still lead
44 to a significant environmental impact, especially since DGT-labile Pb, and thus potentially
45 bioavailable Pb, was released in the water column.
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54 3.4. Dredging accentuated the spatial trends of ultraplankton structure

55 The highest HP abundances observed during the whole monitoring were recorded
56 within the geotextile bag, at IDuS. Moreover, at all sites, a slight but significant HP
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1 enrichment (Kruskal-Wallis tests, $p < 0.05$) was recorded during the dredging period when
2 compared to the reference period (Figure 4). HP enrichment was previously observed along
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4 with sediment resuspension in experimental conditions (Goni-Urriza et al., 2018). Yet, the
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6 present work transposes this observation *in situ* and demonstrates that although precautions
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8 were taken, the dredging methodology used was not enough to avoid impact on the biota.
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12 Regarding the ultraphytoplanktonic community, no significant difference in total
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14 abundance was observed and none of the four considered subgroups demonstrated noticeable
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16 variation during the dredging (Kruskal-Wallis tests, $p > 0.05$). At IDuS, while the mean total
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18 abundance was not different from the other sites (Kruskal-Wallis tests, $p > 0.05$), PPE
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20 abundance was on average the highest among all sites whereas SYN abundance was on
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22 average the lowest (Figure 4). As a consequence, the shift in biological structure observed
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24 from RS to ODuS during the reference period was still observed during the dredging and even
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26 accentuated within the geotextile bag where the highest proportion of PPE was observed 65%
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28 of the sampling dates. This suggests that sediment resuspension and particles concentration in
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30 IDuS accentuated the structure shift from the reference situation in RS. Coclet et al., (2018)
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32 demonstrated by experimental approaches that such structure shift could be linked to metals'
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34 contamination as well as to sediment resuspension. While metals' addition led to a strong
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36 decay of the ultraphytoplanktonic community, exposition to sediment elutriate temporally
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38 promoted phytoplanktonic growth (Coclet et al., 2018). Lafabrie et al., (2013) also reported
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40 similar temporal stimulation of pico and nanoeukaryotic phytoplanktonic groups by sediment
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42 resuspension. Hence, the spatial community structure shift observed during this long-term
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44 study in the shallow and enclosed Toulon Bay could well be linked to sediment resuspension
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46 by different human activities.
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57 Although not determined in this study, the ultraphytoplanktonic biovolume must have
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59 been higher in the northern enclosed area and especially within the geotextile bag since the
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1 dominance of smaller SYN cells ($\leq 1\mu\text{m}$) was replaced by the dominance of larger PPE cells
2 ($\geq 2,5\ \mu\text{m}$ during this monitoring) without any variation in total cells' number. As a
3
4 consequence, higher primary production could explain the larger abundances of HP, which
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6 are known to strongly rely on phytoplanktonic resources in the marine environment (Buchan
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8 et al., 2014; Luria et al., 2017; Williams et al., 2013). The role of sediment resuspension in
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10 providing organic resources for the HP community cannot be excluded since sediment
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12 resuspension can release significant organic carbon amounts (Guigue et al., 2017), fueling HP
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14 growth at short time scales (Goni-Urriza et al., 2018). Moreover, the mixotrophic potential of
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16 PPE (Hartmann et al., 2013) could also contribute to their success in case of sediment-
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18 resuspension-related HP growth, like the one observed in the present study. The respective
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20 influences of phytoplanktonic structure shifts and organic carbon release by sediment
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22 resuspension requires further investigation to assess a potential trophic alteration of the
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24 planktonic food web by dredging activities, as observed in other coastal contexts (Jonsson et
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26 al., 2017).
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34 4. Conclusion

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37 The long-term monitoring during the reference period emphasized the consistency of
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39 the previously-observed spatial gradient of metals' contamination in Toulon Bay. Moreover,
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41 although investigated here on a smaller area, the dominance of PPE over SYN in
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43 ultraphytoplanktonic community of the most contaminated sites also appeared recurrent in the
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45 long term. Temporal ultraphytoplankton and HP abundance dynamics however appeared to be
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47 driven by other parameters than metals' contamination.
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53 During a rather long dredging operation, the use of a combination of sediment
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55 aspiration and immersion of dredged material underwater in a geotextile bag appeared
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57 efficient in preventing dumped sediment dispersion and water column contamination by
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59 contaminated particles at a large spatial scale. However, in spite of the use of this protocol,
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1 statistically significant increases in Pb concentrations and lability were observed at large
2 spatial scale (several km away from the dredging and dumping sites) during the dredging
3 operation under the probable influence of hydrodynamics in the area. Impacts on the biota of
4 the dredging operation consisted in stimulating HP abundance through ecological processes
5 that remain to determine.
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12 In the dumping site created for this dredging operation, the highest metals
13 concentrations, HP abundances and PPE proportion were observed thus appearing to extend
14 the limit of both chemical and biological spatial trends observed during the reference period.
15 Taken together, these observations point toward a strong potential influence of sediment
16 resuspension in the ecological functioning of the highly anthropized Toulon Bay.
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25 The simultaneous characterization of energetic (light, O₂) and nutritive (inorganic and
26 organic nutrients) resources as well as a better characterization of the speciation of Pb
27 controlling its lability represent further direction to better evaluate the ecological processes
28 underlying these observations. Moreover, evaluating the quantity, mineralogy, grain size
29 distribution and contamination of particles along with physico-chemical characteristics of the
30 water (redox potential, pH and oxygen availability) at a higher temporal resolution are needed
31 for a better understanding of the processes leading to metals' remobilization.
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43 This present study clearly points out the necessity to take precautions during dredging
44 activities in order to minimize the contamination in the dissolved fraction which is often
45 disregarded. Indeed, the impact in the particulate phase seemed to have been under control,
46 nevertheless, the dissolved phase was impacted by export of contaminant through the
47 geotextile bag. Therefore, techniques to reduce the impact of harbors activities onto the
48 environment need to be improved.
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13 hydrodynamics.
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27 This paper is dedicated to our friend and mentor, Cédric Garnier, an outstanding scientist who
28 tragically passed away and without whom this work would not have been possible.
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Figures and tables captions

Figure. 1 Location of the monitoring sites and the main anthropogenic activities in Toulon Bay. Sampling sites are represented with the following marking, cross: dredging site (DS), closed star: inside the dumping site (IDuS) (inside the geotextile bag), closed circle: outside the dumping site (ODuS) (outside the geotextile bag), closed triangle: reference site (RS) (near the fish farming area). Note that ODuS is deliberately moved to the right for more visibility on the map, however only a few meters separate it from IDuS.

Figure. 2 Spatial variations of Cu and Pb concentrations (nM) during the two define periods (during the reference and the dredging periods). Pb_L stands for DGT-labile Pb in the manuscript. No data were collected for Pb DGT-labile fraction at IDuS because no DGT could be deployed at this site. Note that concentration scale is given in log-units. On the boxplots, the boxes represent the first (Q1) and third (Q3) quartiles, the band inside the boxes represent the median values. The whiskers represent the minimum and the maximum values whereas closed circles represent outliers.

Figure. 3 Spatial and temporal variation of Cu_T (A), Pb_T (B), Cu_D (C), Pb_D (D) and Pb_L (E). Pb_L stands for DGT-labile Pb in the manuscript. No data were collected for Pb DGT-labile fraction at IDuS because no DGT could be deployed at this site. Note that Cu_T, Pb_T and Pb_D concentrations scale is given in log-units. The period between the two black lines represent the dredging period.

Figure. 4 Spatial variations of microbial groups' abundance (cell.mL⁻¹) during the two define periods (during the reference and the dredging periods). SYN: Synechococcus-like cells, PPE: Photosynthetic picoeukaryotes-like cells. On the boxplots, the boxes represent the first (Q1) and third (Q3) quartiles, the band inside the boxes represent the median values. The whiskers represent the minimum and the maximum values whereas closed circles represent outliers.

Table 1 Ranges and average values of dissolved and total Cu and Pb concentrations (in nM) at the three sites monitored apart from the dredging period.

Table 2 The ranges and average values of dissolved and total Cu and Pb concentrations (in nM) at the four sites monitored during the dredging period. Values in parentheses correspond to percentage and absolute (in nM) concentrations' increase compared to the reference period. Note that for IDuS no DGT were deployed. The stars (*) mean that there is a significant difference between the reference and the dredging period. ND stands for non-determined.

Supplementary material

Element	Site	Min	Average	Max
Cu _D	RS	3.40	9.35 (a)	19.97
	DS	5.50	15.33 (b)	28.88
	ODuS	12.93	36.20 (c)	73.16
Cu _T	RS	4.52	11.57 (a)	22.16
	DS	10.43	22.58 (b)	37.83
	ODuS	18.11	51.83 (c)	95.14
Pb _D	RS	0.13	0.48 (a)	1.05
	DS	0.36	0.98 (b)	1.66
	ODuS	1.65	2.37 (c)	3.90
Pb _T	RS	0.30	0.82 (a)	1.42
	DS	0.75	1.83 (b)	3.70
	ODuS	2.15	4.27 (c)	9.74
Pb _L	RS	0.12	0.77 (a)	1.71
	DrS	0.65	1.44 (b)	3.47
	ODuS	2.22	2.75 (c)	3.98

Table 1 Ranges and average values of dissolved and total Cu and Pb concentrations (in nM) at the three sites monitored apart from the dredging period.

Fraction	Site	Min	Average	Max
Cu _D	RS	4.78	12.74 (36%; 3.39)	31.98
	DS	5.75	20.72 (35%; 5.39)	37.52
	ODuS	18.63	41.17 (14%; 4.97)	60.70
	IDuS	17.63	31.84	65.60
Cu _T	RS	6.24	15.25 (32%; 3.68)	36.24
	DS	7.19	25.23 (12%; 2.65)	41.54
	ODuS	25.54	59.34 (14%; 7.51)	109.85
	IDuS	34.01	440.00	3362.34
Pb _D	RS	0.35	0.86 (80%; 0.38) *	1.81
	DS	0.52	1.58 (61%; 0.6) *	2.60
	ODuS	1.49	3.66 (54%; 1.29) *	7.79
	IDuS	2.22	29.32	81.31
Pb _T	RS	0.47	1.49 (82%; 0.67) *	3.12
	DS	0.73	2.59 (42%; 0.76) *	5.38
	ODuS	1.91	6.55 (53%; 2.28) *	14.86
	IDuS	3.99	362.39	2978.76
Pb _L	RS	0.45	0.97 (26%; 0.20) *	3.09
	DS	0.79	1.65 (15%; 0.21) *	3.09
	ODuS	2.00	4.63 (68%; 1.88) *	10.45
	IDuS	ND	ND	ND

Table 2 The ranges and average values of dissolved and total Cu and Pb concentrations (in nM) at the four sites monitored during the dredging period. Values in parentheses correspond to percentage and absolute (in nM) concentrations' increase compared to the reference period. Note that for IDuS no DGT were deployed. The stars (*) mean that there is a significant difference between the reference and the dredging period. ND stands for non-determined.

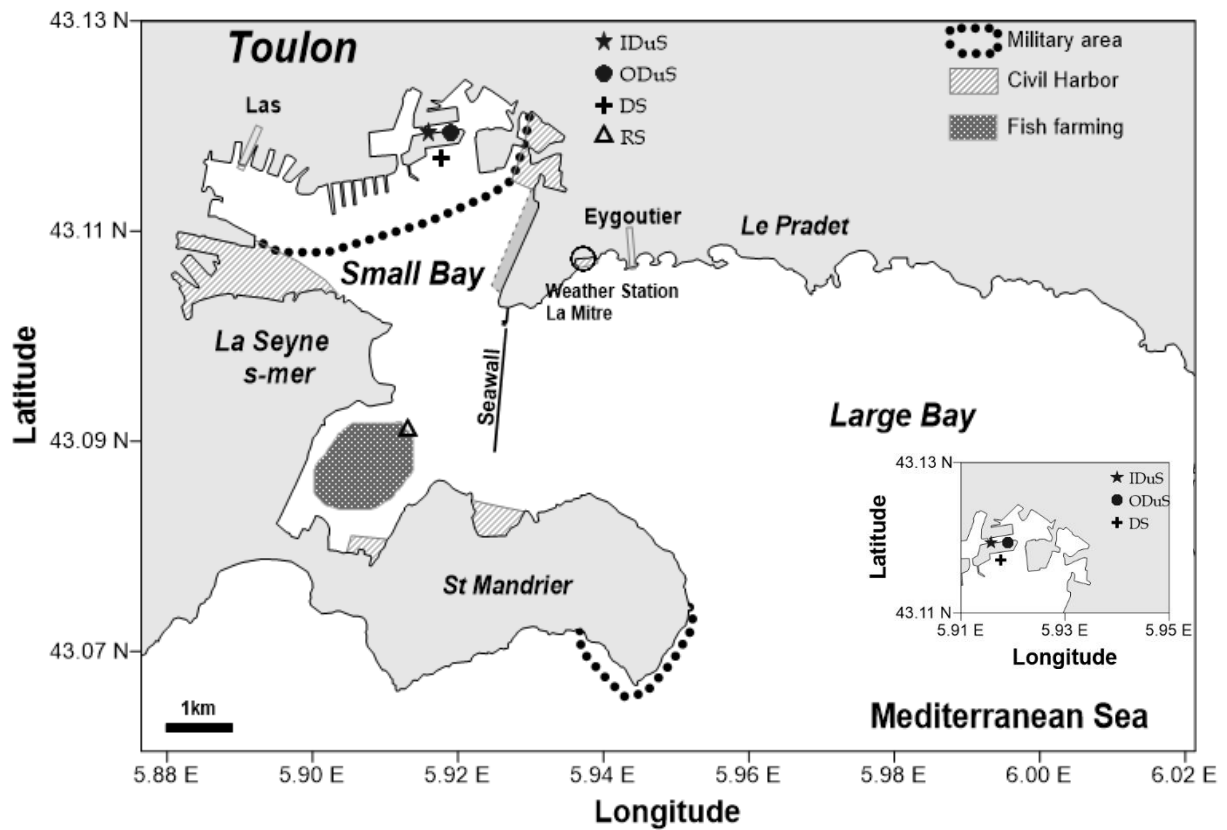


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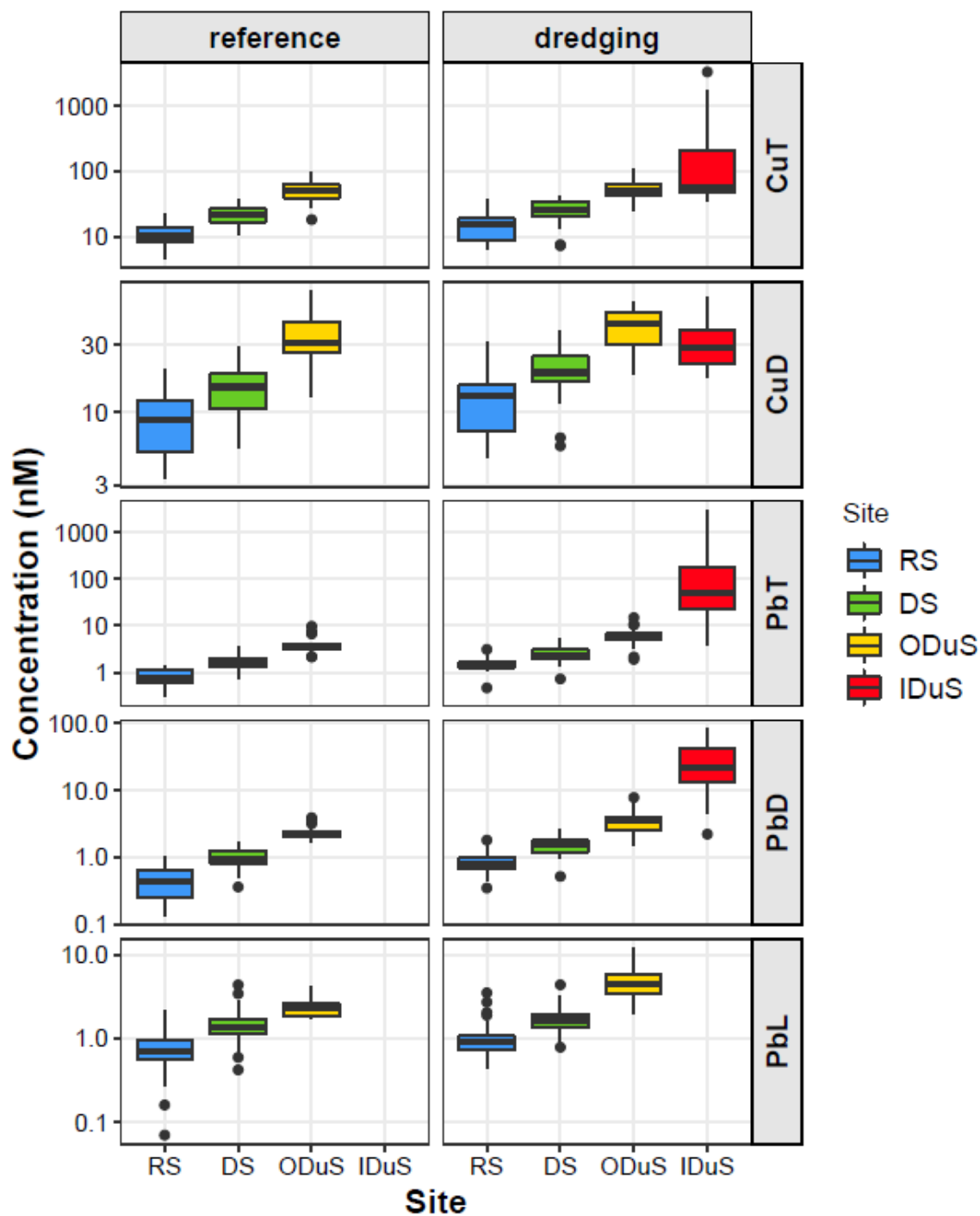


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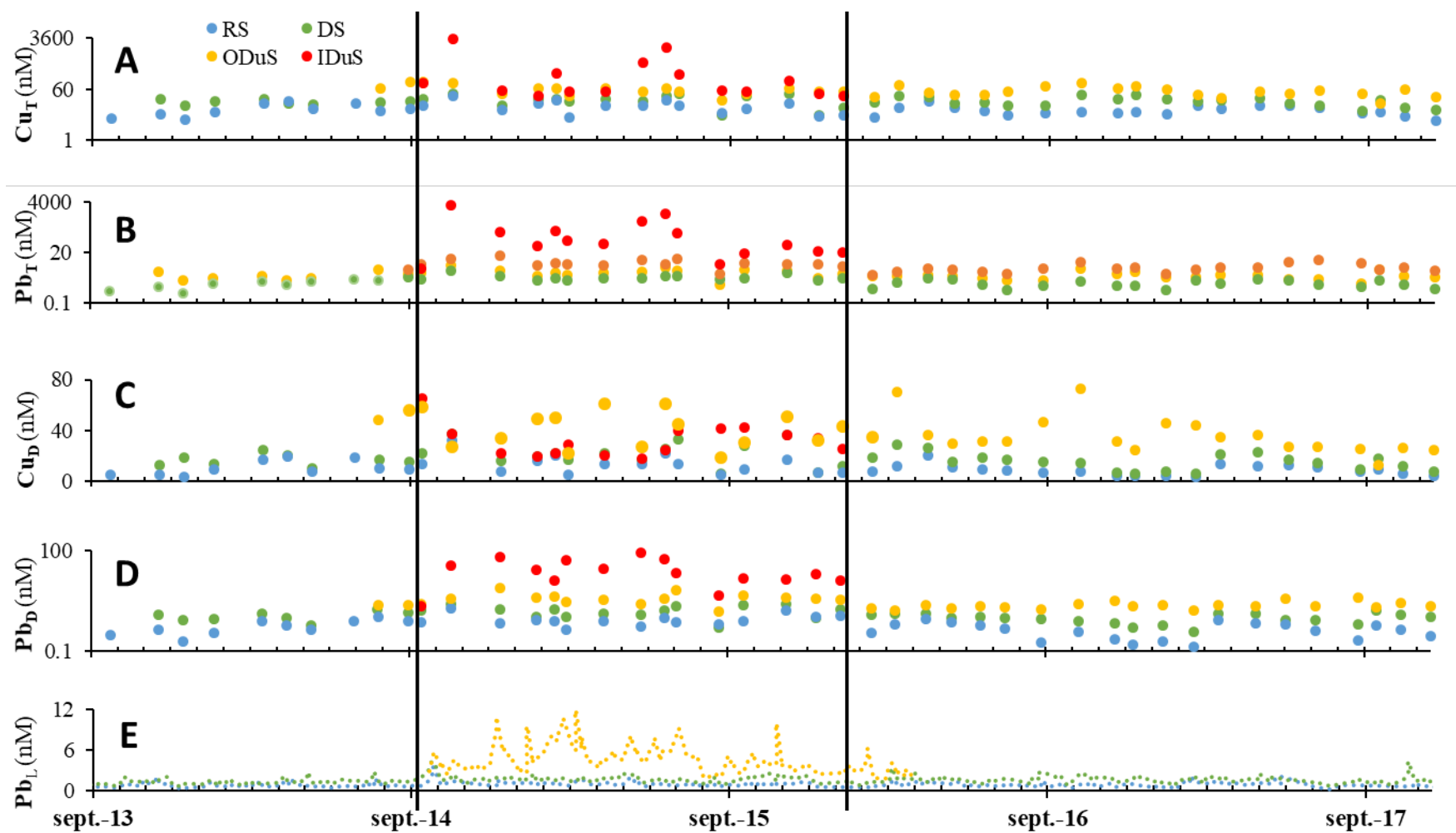


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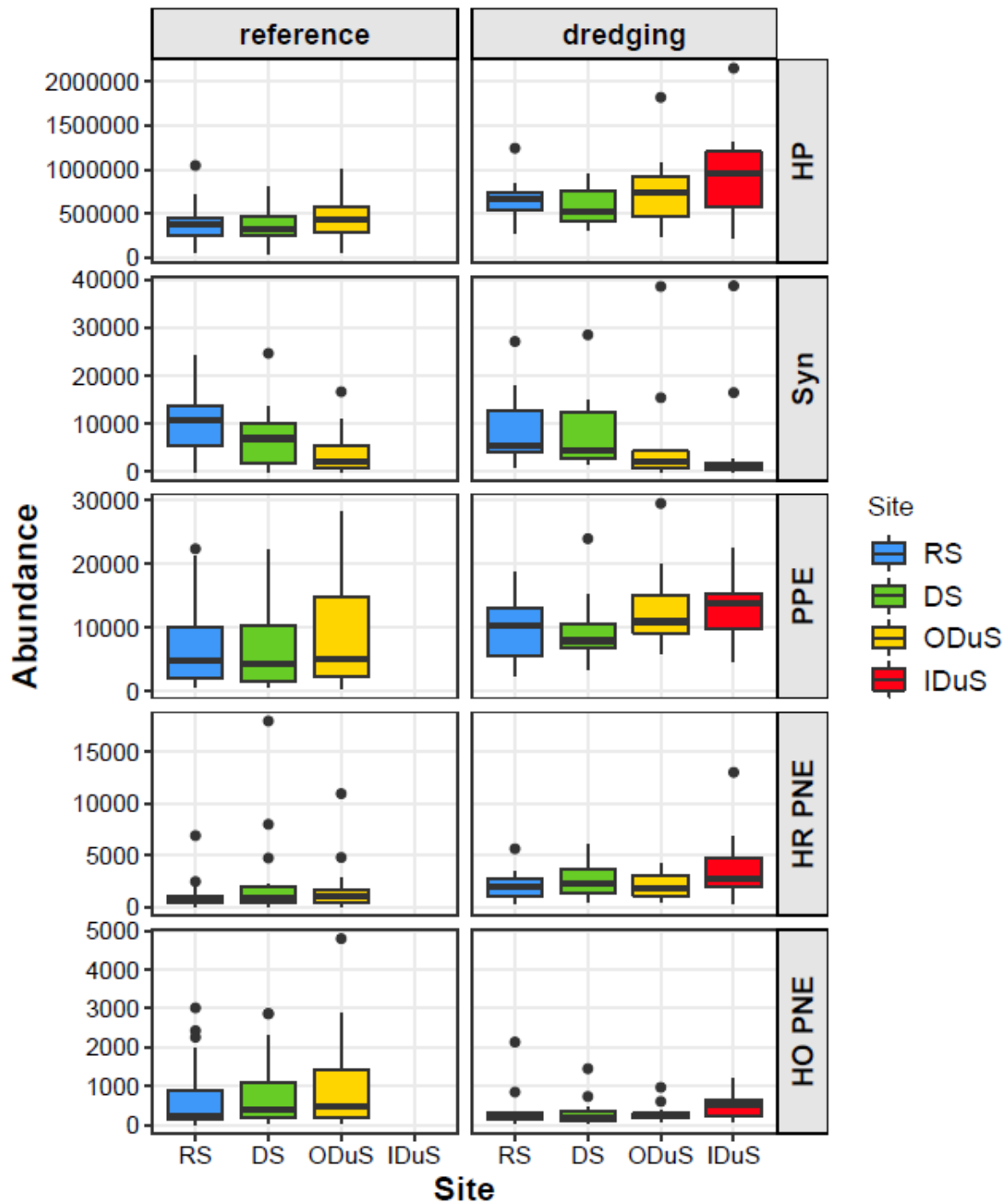


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Supplementary Data

[Click here to download Supplementary Data: Supplementary material.pdf](#)

*Declaration of Interest Statement

Declaration of interests

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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