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Sediment Contamination by Heavy Metals and PAH in the Piombino Channel (Tyrrhenian Sea)

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Abstract: Sediment contamination is of major concern in areas affected by heavy maritime traffic. The spatial variation and contamination of 11 trace elements and 17 PAHs in surface sediments were studied along a 31 km transect along the seaway from the port of Piombino (Tuscany) to the port of Portoferraio (Elba Island) in the Northern Tyrrhenian Sea. Heavy metal contamination was detected at sites near Piombino (Ni, Pb, Hg, Cu and Zn) and at sites near Portoferraio (Pb, Zn, Hg, Cr and Cd). Each of the 35 sampled sites showed PAH contamination, with the highest concentrations at sites near Portoferraio. The most abundant isomers detected were 2- and 4-ring PAHs. PAH ratio analysis showed a prevalence of PAHs of pyrolytic origin. High values of PAHs and heavy metals were related to high sediment water content, TOC, silt, and clay content. Arsenic increased with increasing depth. The correlation between concentrations of metals and PAHs suggests common anthropogenic sources and is of concern for possible synergistic adverse effects on the biota.

Keywords: heavy metals; polycyclic aromatic hydrocarbons; maritime traffic; Tyrrhenian Sea

1. Introduction

Sediment contamination is of major concern in areas affected by heavy maritime traffic [1–3]. In the Mediterranean Sea, the highest concentrations of persistent pollutants were reported in areas influenced by harbour and industrial activities [1]. The erosion and resuspension of sediments caused by shipping vessels [4,5] can lead to an increase in turbidity and resuspension of pollutants, with increasing toxicity for marine organisms [6]. Several studies have proved the toxicity of pollutants coming from resuspended sediments on phytoplankton and bacteria [7,8], and contaminants accumulated in marine plankton can be transferred to higher trophic levels through the food web, with potential risks also for human health [9–11]. Among others, two classes of contaminants are of particular concern: polycyclic aromatic hydrocarbons (PAHs) and heavy metals.

PAHs are an important class of persistent organic pollutants in the marine environment [12,13]. They can be released from petrogenic and natural sources, but the largest emissions of PAHs result from industrial processes and other human activities [14]. Anthropogenic sources of PAHs result from incomplete combustion (e.g., of coal, petroleum, and wood) or come from crude and refined petroleum products (oil spills, routine tanker operations, oil production, etc.). PAHs are known to have adverse effects on humans and the environment through mechanisms of carcinogenesis, mutagenesis, and teratogenesis [14,15], as well as acute and developmental toxicity, observed in the aquatic environment [16–18].



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). Their hazardous nature, semi-volatility, long-distance migration, lipophilicity, low water solubility, and adsorption to marine particles and sediments make them a group of chemicals that can accumulate throughout the food chain [19,20]. Therefore, the study of the distribution and source of PAHs in sediments is of great importance for the understanding of pollution sources [21]. According to the Italian National Act 260/10, as an implementation of the European Water Framework Directive (WFD; 2000/60/EC), 16 PAHs have been included in the list of priority pollutants for the assessment of Chemical Status.

Heavy metals in marine sediments occur at variable concentrations due to both natural processes (i.e., rock weathering, coastal and seabed erosion) and anthropogenic activities (i.e., discharge of municipal and industrial wastewaters, entry of runoff from agricultural and adjacent coastal lands, and riverine fluxes) [22,23], with human activities supplementing their natural geochemical background values [24]. Heavy metals are considered as serious contaminants in marine sediments due to their toxicity and stability to degradation; they bioaccumulate and biomagnify in the food chain and pose a risk to predators and human health [25–27].

The Tyrrhenian basin hosts large and important commercial and tourist ports (Leghorn, Piombino, Portoferraio, Civitavecchia, Naples, Palermo, Cagliari, Olbia, Bastia, just to name a few), and is affected by the consequent huge ship traffic. Nevertheless, there are few studies dealing with seafloor contamination in the Tyrrhenian basin, and most of them are concentrated in the southern part of the basin, such as the Gulf of Naples [28–30] or the mouth of the river Tiber [31]. The Piombino channel is a stretch of the sea located in the northern Tyrrhenian Sea between the Gulf of Follonica (Tuscany) and Elba Island and is among the less studied areas. This channel is affected by the heavily impacted seaway connecting the port of Piombino (continent) to the port of Portoferraio (Elba Island): in 2013, total traffic of the port of Piombino accounted for 3532 passengers and 6,074,000 tons of shipping material, whereas the total traffic of the port of Portoferraio accounted for 2839 passengers [32]. But the area has also high ecological value being part of the Ligurian Sea Cetacean Sanctuary and other protected areas and hosting an important net of *Posidonia oceanica* meadows, included in Habitat Directive (92/43/CEE) as a protected and priority habitat type.

The aim of the present work was twofold: (i) to analyse the concentrations of PAHs and trace metals in sediments from a busy seaway, and (ii) to assess the degree of contamination of the area and its potential adverse effect on aquatic fauna.

2. Materials and Methods

2.1. Study Area

The Tyrrhenian Sea (Central Mediterranean) is a deep basin delimited by the Italian coast to the east, by Corsica and Sardinia to the west, and by Sicily to the south. It has a narrow and shallow northern opening toward the Ligurian Sea (the Corsica channel) and a much larger and deeper opening in the south. The Tuscan Archipelago, of which Elba Island is the largest and most famous island both from a historical and tourist point of view, is located at the center of the northern part of the basin.

The study area was a stretch of sea (named Piombino channel) comprised between the Gulf of Follonica (Tuscany) and Elba Island, in the northern Tyrrhenian Sea (Figure 1). Along with this route, 35 sampling sites (Table A1) were chosen, subdivided into three areas, Piombino (PB), Portoferraio (PF), and offshore (OF), to study the distributional pattern of pollution by 11 trace elements and 17 PAHs. Surface sediments were collected from 2nd to 4th November 2017, during an environmental study performed by TERNA SpA related to submarine HVAC link to Elba Island and Mainland realization along the seaway from Piombino to Portoferraio.



Figure 1. Map with the location of the Piombinochannel, and the three study areas along the seaway (Piombino, PB1-10, Portoferraio, PF1-14, and offshore, PF15-25).

The Piombino Channel is an area of particular interest, as there are numerous potentially polluting activities affecting it. In fact, this relatively narrow stretch of sea is between a highly polluted industrial site (Piombino), an equally highly polluted tourist port (Portoferraio) and is crossed daily by a number of ferries. The Site of National Interest (SIN) of Piombino consists of an industrial pole of considerable size, the port area of Piombino, characterized by commercial and tourist traffic towards the islands (Elba, Sardinia, and Corsica), and by industrial activity, mainly the steel industry. Among the pollutants deriving from steel processing, there is coal and all the products derived from its distillation, namely PAHs, tars, and heavy metals contained in ferroalloy minerals that are added to steel (e.g., Cr, V). In the industrial area subjected to remediation, main contaminants have been identified, both in soils (As, Crtot, V, Cd, Zn, Ni, Pb, Hg, Cu, PAH), and in groundwater (As, Fe, Pb, CrVI, Ni, PAH, PCB, aromatic solvents). With regards to the port of Portoferraio, its maritime traffic is almost exclusively made up of ships of the Passenger and Passenger/Ro-Ro categories, with over 9800 ships per year: the annual activity times have been calculated equal to over 20,500 h, with the mooring phase representing over 90% of the total hours.

2.2. Sample Collection and Chemical Analysis Methods

Four sediment replicate samples were collected at each site with a 0.1 m³ Van Veen grab, with the exception of sites PF15-25, in the off-shore trait, where, due to technical constraints, only one sediment sample was collected. The top 5 cm of each grab were collected into separate pre-labeled, acid-washed, and solvent-rinsed glass jars, stored onboard ship in coolers filled with ice, and then stored at -20 °C immediately upon return to the laboratory. All samples were analyzed for grain size ([33] -ASTM D 422-63 method), water content (by gravimetry), total nitrogen, and phosphorus ([34] -CNR-IRSA, 1985),

and total organic carbon (TOC-CUBE Analyzer). Trace elements (Al, As, Cd, Crtot, Hg, Fe, Ni, Pb, Cu, V, and Zn) were analyzed by inductively coupled plasma mass spectrometry (Thermo ScientificTM TQ-ICP-MS), according to the EPA method 6010D [35], with BRC-320R as certified reference material. Polycyclic aromatic hydrocarbons (PAHs) were extracted and cleaned using a Thermo ScientificTM DionexTM 350 Accelerated Solvent Extractor; analysis was performed following EPA methods 3545A [36], 3630C [37], and 8270D [38], and quantification through high resolution gas chromatography high-resolution mass spectrometer (Thermo ScientificTM TRACE 1300 GC + Thermo ScientificTM TSQ Duo TQ-MS). Spiked sediment samples were analysed with each set of samples: the relative standard deviation for duplicate spiked samples ranged from 0 to 10%, while recoveries from 80 to 110%. Ultrascientific^{®®} PAH Mix standard (Batch n. 6466630 and 6509932) was used for examining the recovery of samples and quantifying the analytes. The detection limit was 0.2 ng/g dry weight.

2.3. Data Analyses

Sediments were defined according to the grain size following the classification of Flemming (2000) [39]. Spearman rank correlation coefficient (rs, [40]) was used to check for correlations among sediment variables.

Non-parametric chi-square test applied to Kruskal-Wallis (KW) ranks was used to test for differences in PAHs and trace elements concentrations among sampling areas [41], followed by Wilcoxon rank-sum test (W) post hoc comparison test when significant differences were encountered.

To infer information about the potential sources of PAHs, we consider the diagnostic ratios most commonly used in the literature, including Flu/(Flu + Py), Ant/(Ant + Phe), following Perra et al. (2009) [42] and Han et al. (2019) [21], the relative concentrations of PAHs by aromatic groups (2-ring, 3-ring, 4-ring, 5-ring, and 6-ring PAHs), and the ratio between low molecular weight (2–3 rings) and high molecular weight (4–6 rings) PAHs (LMW/HMW).

In order to define the chemical status of the area, chemical data were confronted to the existing national thresholds, when available, according to the Italian Law (DL56/2009). This law fixes the threshold concentration of harmful elements and compounds to discriminate between Good and Moderate status, as requested by the Water Framework Directive (WFD). In order to evaluate the potential risk for benthic organisms, chemical concentrations were compared with thresholds defined by Sediment quality guidelines established by Long et al. (1995) [17]. Those guidelines define two thresholds for a chemical: Effects Range-Low (ERL) and Effects Range-Median (ERM). These values defined three ranges, rarely (below the ERL), occasionally (above the ERL, but below ERM), and frequently (above the ERM) associated with adverse effects on aquatic organisms.

Principal Component Analysis (PCA) was finally performed with spectral decomposition function in order to examine the relations among pollutants and sediment parameters. Prior calculation data were log-transformed, centred, and scaled. High collinearity levels among sediment texture and pollutants were checked with Spearman rank correlation coefficient (rs, [40]). All calculations were performed with the R package [43]. The chosen significant threshold was p < 0.05.

3. Results

Sediment grain size was variable (Table A1), ranging from sand and silty sand (>50% of sand) at sites PB1-5 and PF17-25, to clayed silt (>50% of silt) at PB7 and silty clay (>50% of clay) at PB8-9. Sediment composition varied with depth. In PB area (PB1-10) sand (rs = -0.78) and gravel (rs = -0.82) decreased and silt (rs = 0.79) and clay (rs = 0.92) increased with increasing depth; the same pattern of increasing silt (rs = 0.61) and clay content (rs = 0.85) and decreasing gravel (rs = -0.69) with depth was observed also in OS area (PF15-25). Conversely in PF area(PF1-14) there was an increase of sand (rs = 0.81) with depth. Total organic carbon (TOC) showed a positive correlation with nitrogen (Ntot, rs = 0.49), water

content (rs = 0.84), clay (rs = 0.72), silt (rs = 0.68), and a negative correlation with sand content (rs = -0.72).

PAHs concentrations are shown in Table A2. The sum of the 16PAHs (Figure 2) required by national legislation (Σ 16PAH, all analysed PAH except BeP) ranged from 9.72 μ g/Kg d.w., at PB3 to 11,689.36 μ g/Kg d.w. in PF1. The concentration of Σ 16PAH does not differ significantly among areas (KW: p > 0.05). Σ 16PAH exceeded the national threshold for good chemical status (800 μ g/Kg d.w.) in sediments of 23% of the sampling sites: 6 sites in PF, 1 in PB, and 1 in OS area, and the deviation was higher than the 20% tolerated by in Italian legislation (with the exception of site PF14). Sites PF1 and PF2 were the most contaminated, with concentrations up to 15 times higher than the national threshold and exceeding the ERL threshold. PF resulted in the most contaminated area with a total of 9 PAHs exceeding their threshold in a total of 8 sampling sites (Table A2); whereas in OS and PB areas, only 3 PAHs exceeded their thresholds in a total of 4 (PB) and 3 (OS) sampling sites. Some PAHs exceed also their ERL threshold (Table A2): all those sites were located in the PF area (PF1-3 and PF5). ERM thresholds [17] were never exceeded. The ratio Flu/(Flu + Py) was > 0.5 in sediments of 97% of sampling sites, Ant/(Ant + Phe) was always >0.1 in sediments of all sampling sites, suggesting combustion processes as the main sources. Consistently LMW/HMW ratio was generally rather low (<1), with 4-ring PAHs as the dominant species, accounting for more than 50% in many samples (Figure 3). There were, however, exceptions: at sites PB1-5 and PF18-22, the dominant species were 2-ring PAHs (47–83%), and at site PF13, there was no marked dominance of one species of PAHs, with consequent LMW/HMW ratio higher than 1. Low molecular weight PAHs are less resistant to microbial degradation; therefore, those values suggested a possible petrogenic origin, closely related to petroleum products [42].



Figure 2. Concentration of Σ 16PAH at the sampling sites (bars are standard deviation).



Figure 3. Relative concentrations (%) of PAHs by aromatic groups.

Trace elements concentration in the surface sediments at each site are shown in Table A3. PF area showed the highest concentrations of V, Pb, Ni, Cr (KW and W: p < 0.05), conversely As showed higher concentration in OS area compared to PB and PF areas (KW and W: p < 0.05). Most elements (with the exception of As and Fe) showed a similar pattern, with the most contaminated sites located close to the port in the PF area (Table A3). Cd was the only metal never exceeding any threshold (Table A3). Arsenic was the most concerning element: at all sampling sites, its concentrations exceeded both the national threshold plus the 20% tolerated by Italian legislation for good chemical status in sediments and its ERL threshold. Indeed, it exceeded also its ERM threshold at 17% of the sampling sites. As shown in Table A3, many heavy metals exceeded their national threshold for good chemical status (e.g., Cr in 77% of the sampling sites, Hg in 20%, Ni in 69%, Pb in 29%), and also their ERL (Cr, Hg, Ni, Pb, Cu, Zn) or ERM (Hg, Ni) thresholds.

All forms of PAHs were correlated among each other (p < 0.05) with the exception of naphthalene (N), showing significant correlations (p < 0.05) only with Acenaphtene (Ace) and Fluoranthene (Fl). There was a strong collinearity also among the concentration of some metals, in particular between Cu and Zn (rs = 0.93; p < 0.05), Pb and V (rs = 0.90; p < 0.05), Cu and Fe (rs = 0.89, p < 0.05). The PCA biplot (Figure 4) showed 74.2% of the total variation. The variables contributing more to the first axis were: Al, Cu, Ni, water content, clay, and Σ 16PAH, while variables contributing to the second axis were depth, As, and Ptot. Figure 5 shows the concentration of some selected pollutants (Σ 16PAH, As, Cr, Ni, Hg, V) in the Piombino Channel.



Figure 4. Correlation biplot showing relationship among concentrations of metals, the sum of 16 PAHs, and other sediment parameters. Ptot = total phosphorus, Ntot = total nitrogen, TOC = total organic carbon.



Figure 5. Concentration of selected pollutants (Σ16PAH, As, Cr, Ni, Hg, V) in the Piombino Channel.

4. Discussion

There is not much information on the contamination of sediments in the Tyrrhenian Sea. The values of pollutants such as PAHs recorded in this study are quite similar to those reported for other polluted Tyrrhenian areas, such as Naples harbour (9 to 31,774 ng/g; [28]), the whole Gulf of Naples (15.3 to 678.6 ng/g; [29]), and the mouth of the river Tiber (36.2 to 545.6 ng/g, [31]). The concentration of PAHs was highest in the area near Portoferraio port, where the concentration of Σ 16PAH decreased as we move away from the port, suggesting that those pollutants arise from port activity sources. Combustion processes are a source of PAHs, as shown by the dominance of 4-ring PAHs, low Phe/Ant ratio, high Flu/Py ratio, and low LMW/HMW ratio. Accidental leakage of oil spills [42], however, must not be overlooked to interpret high concentrations of some PAHs. Unfortunately, despite the intense maritime traffic, there is no reliable data regarding oil spillages in the area. Some PAHs (Ant, Acy, Fl, Flu, Py, BaA, Chry) exceeded their ERL threshold in the PF area (PF1-3 and PF5), but they never exceeded their ERM thresholds [17], suggesting a mild probability of adverse effects on organisms.

The spatial distribution of metals in sediments suggests their land-based sources. In general, the PF area was the most contaminated. The high levels of metals (in particular V, Pb, Ni, Cr) were likely related to anthropogenic pollution. Elba island has been one of the most important iron mining areas in Italy since Etruscan times [44]. Extraction activity ceased in 1981 and left many abandoned mines, and waste dumps, mineral deposits, and heavy metals and metalloids (As, Cr, Co, Cu, Pb, Ni, and Zn) related to iron extraction and processing, with consequent water and soil contamination [45]. Those metals likely

accumulated in coastal sediments due to weathering processes [44,46,47]. In the same way, metals (Ni, Pb, Hg, Cu, and Zn) found in sediment in the PB area can be attributed to the steel industry and power plants located in the industrial area, as suggested by previous studies [46,48].

The comparison with the metals background level of the area shows that the highest values of Cu, Pb, and Zn found in the present study were above the reference value for silty unpolluted sediments reported in 1991 for the northern Tyrrhenian (Cu: 33 mg/kg, Pb: 36 mg/kg, Zn: 101 mg/kg, [48]), confirming the anthropogenic source of pollutants. Moreover, the same work stated that the northern Tyrrhenian was known to be contaminated by Pb and reported enrichment of Pb in surface sediments [47,48]. The presence of Zn in the area was related to the phyllosilicate fraction [48], so its presence at sites near Portoferraio (PF area) could be related to the marine erosion of greenstone outcrops along the Elba shoreline [48]. Nevertheless, Leoni and Sartori (1997) [47] suggested a likely anthropogenic source of Zn in the area since they found a vertical trend in sediment concentration of Zn, with surface layers enriched with Zn. Conversely, concentrations of Cu in the area were related to carbonates and organic matter, and no enrichment of this element was previously reported for surface sediments [48]. This suggests that the few peaks of concentration of the present work (PB10 and PF5) were likely related to the numerous mineral deposits present in the catchment area of the Elba-Argentario basin [48]. The highest values of Cr and Ni found in the present work were in line with those reported for the central part of the Gulf of Follonica (Cr: 96–146 ppm, Ni: 20–88 ppm, [47]). In that area, the high concentrations of Cr and Ni were related with the phyllosilicate fraction, without a vertical trend in the sediment column; therefore, they were ascribed to natural sources and sediment composition [47]. Recent work confirmed that high levels of Cr and Ni along the central coast of Tuscany closely reflect the geology of river catchments, thus indicating a geogenic origin [49]. The values of Hg found in the present work are generally consistent with mild pollution observed in areas distant from the main sources [50]. Northern Tyrrhenian has been historically contaminated by Hg due to different sources, such as the cinnabar deposits of Mount Amiata, south of the studied area [50,51], and different industrial plans such as the oil-refinery of Leghorn and Rosignano Solvay, north of the studied area [50]. In the present work the values of As largely exceeded reference values reported in 1991 for silty unpolluted areas in the northern Tyrrhenian (24–38 mg/kg, [48]), suggesting a likely anthropogenic enrichment due to maritime and industrial activities. Previous studies indicated multiple anthropogenic As sources, such as agricultural [48] and industrial [47] activities. However, high values of As were known to characterize most of Tuscany's continental shelf [47,48,52], with anomalies most likely due to the basin's hydrodynamic conditions, which favor the sinking of the finest-grained particulates in the center of the basin and the combined effects of anthropogenic contamination and the influx of natural As-bearing debris from eastern Elba Island mineral deposits [47,48]. Recent studies confirmed that the high As concentrations in the region reflect leaching of felsic volcanic and plutonic parent rocks and hydrothermal products related to the Tuscan and Roman magmatic provinces cropping out in local river catchments [49].

From the ecotoxicological point of view, Cd was the only metal never exceeding any threshold, therefore not posing any threat to benthic fauna. Conversely, As was the most concerning element, exceeding its national threshold for good chemical status in sediments, and its ERL threshold at all sampling sites, and at some sites even its ERM threshold, suggesting likely adverse effects on benthic organisms. Concentrations of Cr, Pb, Zn, and Cu exceeded their ERL threshold, while As, Hg, and Ni exceed even their ERM thresholds at some sites, raising concern of possible negative effects on biota. Some metals, such as Cu, Cr, and Zn, are essential for biological function in trace concentrations [53], while high concentrations of the essential elements have adverse effects on living organisms [54]. In particular, Cu and Zn are essential elements for plankton life [53], and their pollution is mostly associated with sewage outfalls [55]. Conversely, other elements, including As and Cd, are not metabolized and have toxic effects also at very low levels [56]. Hg and Pb

have no biological function as well. Hg is toxic even at very low concentrations and can accumulate through the trophic chain posing risks for human health [57–59].

Grain size is a major factor in controlling sedimentary heavy metal concentrations [60–62] because the different specific surface areas of the sediment particles have different adsorption capacities [63,64]. Heavy metals in marine sediments have a particular affinity for finerparticle grain sizes and bind to the surface of fine particles by different processes [24,65]. The marine depositional areas of silt and clay represent the principal heavy metals archive and reflect the pollution levels in the water column [66]. The degree of correlation between metals and other elements can give us additional indications regarding the origin of the metals [55,60]. The correlation of metals and PAHs concentrations suggested common anthropogenic sources from combustion processes, confirming the hypothesis of anthropogenic enrichment of surface sediments at sites with also high concentrations of PAHs (PB10, PF1-7). The high levels of TOC found at sites with high concentrations of PAHs could reduce the bioavailability of those pollutants [14]. Some investigations found an inverse correlation between the rate of uptake of PAHs and total organic carbon (TOC) content of sediment, implying that as TOC increases, less of the compound is soluble and available for uptake into the organisms [67,68]. Interestingly, this does not imply necessarily a straightforward relation between TOC and PAHs accumulation in biota and sediment. Some authors found that PAHs accumulation decreased with increasing TOC [69,70], while others found the accumulation to increase with increasing TOC, even if the rate of uptake was lower [68]. The collinearity among concentrations of most metals and PAHs suggested possible synergic negative effects on biota. This is particularly of concern since this effect could be amplified by sediment resuspension caused by maritime traffic in the area.

Author Contributions: M.M. coordinated the team of authors. M.F. designed the sampling campaign. G.T., M.L. and M.M.L. collected and analyzed samples. P.V. wrote the first version of the manuscript, M.M., M.C., S.A. and S.A.A. revised and edited further versions of the manuscript. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

 Table A1. Coordinates (WGS84 GMS) and main characteristics of sampling sites: depth (m), grain size (gravel, sand, silt and clay, %), % water (%), total phosphorous (Ptot, mg/kg d.w.), total nitrogen (Ntot, mg/kg d.w.), total carbon (TOC, %). ND = no data available.

 Lensitude (T)
 Depth area
 Sand
 Silt
 Clar
 % Water (%), total phosphorous (Ptot, mg/kg d.w.),

_		Longitude (E)	Latitude (N)	Depth	Gravel	Sand	Silt	Clay	% Water	Ptot	Ntot	TOC
	PF1	10°19′10.21″	42°48′13.33″	1	1.1	13	42.1	43.9	59.9	233.4	237.7	4.4
	PF2	10°19'15.95"	42°48'12.09"	2	0	12.5	46.6	40.9	55.7	258.1	284.5	4.7
	PF3	10°19′25.22″	42°48'11.11"	2.7	1.3	9.5	46.3	42.9	52.7	251.7	267.5	4.9
	PF4	10°19'34.19"	42°48'10.62"	2.9	14.5	40.4	27.3	17.9	43.5	172.2	131.2	3.6
	PF5	10°19′42.25″	42°48'12.92"	4.3	7	41.2	37.7	14.1	55.6	264.6	171.0	4.2
	PF6	10°19′42.25″	42°48'12.92"	12.4	ND	ND	ND	ND	56.4	198.8	151.0	4.1
	PF7	10°20′16.16″	42°48′33.57″	28	ND	ND	ND	ND	56.5	268.2	246.7	4.2
ronoierraio	PF8	10°20'32.81″	$42^{\circ}48'44.10''$	31	6.2	31.3	44.8	17.7	41.1	196.8	152.9	2.4
	PF9	10°20′49.50″	42°48′54.67″	36	ND	ND	ND	ND	21.0	129.1	138.5	0.3
	PF10	10°21′05.02″	42°49′05.71″	40.5	ND	ND	ND	ND	20.1	127.3	151.6	0.3
	PF11	10°21′06.21″	42°49′21.71″	55	15.7	46	23.7	14.6	31.6	251.5	152.6	0.7
	PF12	10°21′18.09″	42°49′35.41″	54.5	ND	ND	ND	ND	25.2	289.9	135.3	0.6
	PF13	10°21′29.84″	42°49′49.05″	62	ND	ND	ND	ND	24.5	230.4	156.9	0.3
	PF14	10°21′41.72″	42°50′02.68″	65	4.7	43.8	32.8	18.7	28.9	222.8	151.2	0.7
	PF15	$10^{\circ}22'14.01''$	42°50′56.34″	71.5	4.1	36.4	31.3	28.1	40.2	567.8	106.5	0.96
	PF16	10°22′42.92″	42°51′50.07″	71.5	2.8	46	25	26.1	36.0	538.6	128.7	0.9
	PF17	10°23′12.63″	42°52′43.59″	70.5	7.7	58.4	15.6	18.3	31.2	548.1	150.7	0.5
	PF18	10°23′12.63″	42°52′43.59″	68.5	1.8	83.2	6.8	8.2	35.4	474.5	94.1	1.2
	PF19	10°25′26.55″	42°53′35.76″	64.5	1.4	90.5	3.1	5	29.7	450.1	91.4	0.2
Off-shore	PF20	10°26′42.53″	42°53′49.65″	53	17.7	73.1	4.4	4.7	28.3	438.5	84.2	1.5
	PF21	10°27′58.62″	42°54′03.76″	46	12	79.6	4	4.4	32.8	381.5	22.4	0.2
	PF22	10°29′14.67″	42°54′19.76″	38.5	11.4	78.2	4.5	5.9	28.0	362.5	64.5	1.4
	PF23	10°30′30.97″	42°54′29.46″	38	20.1	69.2	5.8	4.9	23.5	324.8	63.7	0.2
	PF24	10°30′52.71″	42°54′29.14″	31.5	28.3	65.2	3.2	3.3	18.1	298.8	99.1	0.3
	PF25	10°31′14.63″	42°54′29.24″	31	9.7	80.3	5.3	4.6	26.2	334.1	106.4	0.3
	PB10	10°33′59.84″	42°55′09.93″	26	0	18.9	46.4	34.7	39.5	439.1	147.6	1.6
	PB9	10°34′41.51″	42°55′52.54″	21.2	0	3.6	44	52.4	37.9	203.2	131.7	1.5
	PB8	10°35′39.73″	42°56′32.97″	14.5	0	1.4	48.1	50.5	45.5	323.8	135.7	1.2
	PB7	10°35′34.70″	42°56′41.53″	11.1	0	3.1	60.7	36.2	41.0	283.7	132.5	1.9
Piomhino	PB6	10°35′32.08″	42°56′46.11″	10.5	0.4	43.9	30.5	25.2	30.0	162.8	58.5	0.7
riombino	PB5	10°35′32.21″	42°56′46.11″	9	0	89.8	4.1	6.1	24.6	116.4	143.2	1.1
	PB4	10°35′32.43″	42°56′52.65″	7	4	86	5.6	4.5	16.9	103.0	45.0	0.2
	PB3	10°35′31.98″	42°56′55.87″	5.6	0.4	59.7	37.2	2.6	19.1	73.7	58.0	0.2
	PB2	10°35′31.59″	42°56′59.31″	4.2	11.8	83.6	1.7	2.9	17.8	63.0	185.4	0.2
	PB1	10°35′36.48″	42°57′02.61″	3.7	2.8	92.6	2.4	2.3	15.0	56.7	174.6	< 0.08

Table A2. Concentrations of PAH (Units: μg/kg d.w.). National Thresholds (NT, According to DL56/2009), ERL and ERM (according to Long et al., 1995) are also reported. Naphthalene (N), Anthracene (Ant), Phenanthrene (Phe), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Fl), Fluoranthene (Flu), Pyrene (Py).Benz[a]anthracene (BaA), Chrysene (Chry), Benzo[b]+[j]fluoranthene (BbjFlu), Benzo[a]pyrene (BaP), Benzo[k]fluoranthene (BkFlu), Indeno [1,2,3-cd]pyrene (InPy).Benzo[ghi]perylene (BPl), Dibenzo[a,c] + [a,h]anthracene (DBA), Benzo[e]pyrene (BeP).

			Ν	Ant	Phe	Acy	Ace	Fl	Flu	Ру	BaA	Chry	BbjFlu	BaP	BkFlu	IndPy	BPI	DBA	BeP
	NT		35	45	_	_	_	_	110	_	_	_	_	30	20	70	55	_	_
	ERL		160	85,3	240	44	16	19	600	665	261	384	—	430	—	—	—	—	—
	ERM		2100	1100	1500	640	500	540	5100	2600	1600	2800	_	1600	_	_	_	—	_
	DE4	avg	1.4	320.2	190.1	137.6	3.5	20.2	2409.9	1773.3	1567.9	2176.1	2139.2	115.0	695.6	67.3	50.6	22.5	565.2
	LLL LLL LLL LLL LLL LLL LLL LLL LLL LL	sd	0.4	535.2	318.1	225.1	5.6	29.8	4081.1	2987.4	2631.4	3653.1	3546.8	187.1	1148.1	114.8	87.3	37.9	945.2
	PE2 avg	avg	11.4	125.3	68.9	120.7	1.7	6.1	959.3	838.4	583.6	940.4	1326.1	64.4	431.8	45.7	36.1	14.3	354.0
	ГГД	sd	18.7	205.6	112.4	198.1	1.4	6.0	1610.7	1403.9	964.3	1566.0	2195.7	102.8	710.5	78.7	62.1	24.4	591.6
	PE3	avg	0.3	49.7	32.7	46.6	0.6	3.0	383.7	336.0	269.7	433.3	605.3	31.6	204.6	22.0	17.2	7.2	108.1
	115	sd	0.1	77.2	51.8	71.8	0.7	4.9	620.3	544.7	426.6	696.9	948.7	47.2	319.1	35.4	28.6	11.4	168.6
	PF4 avg sd	avg	3.6	9.7	5.2	12.9	0.8	2.2	68.3	63.6	48.3	71.0	115.0	7.2	39.3	3.1	2.0	1.2	23.2
		sd	5.8	10.7	4.3	18.1	0.5	1.8	98.7	90.3	69.0	106.1	177.0	9.0	58.8	5.0	3.2	1.7	33.1
	PF5 avg	avg	46.8	120.6	72.3	102.0	12.0	20.7	613.5	569.4	413.9	640.9	767.9	44.1	258.6	28.7	24.1	9.5	162.3
	115	sd	9.4	151.2	66.0	112.2	12.0	16.5	627.1	612.0	439.9	777.3	928.9	50.2	306.0	39.3	33.8	12.4	190.0
	PE6	avg	<0.2	5.6	6.2	3.8	1.2	1.6	21.1	19.5	16.7	24.7	37.1	3.6	14.0	0.8	0.2	0.4	9.0
	110	sd	ND	6.3	8.2	4.8	1.8	2.4	30.3	29.7	23.4	38.0	56.4	2.8	19.5	1.0	0.0	0.3	11.3
	PF7	avg	0.3	47.7	32.1	35.6	5.6	9.4	237.8	212.0	181.4	315.6	324.2	19.4	121.4	20.2	15.6	6.3	72.3
Portoferraio		sd	0.2	75.9	48.8	55.3	5.9	11.4	378.8	336.3	280.0	501.7	493.3	28.1	185.0	24.7	19.1	7.8	110.0
Tontorentato	PF8 av	avg	< 0.2	2.9	1.8	2.3	0.3	0.4	12.2	10.7	10.6	15.9	16.5	2.3	6.6	< 0.2	< 0.2	< 0.2	7.4
	110	sd	ND	1.5	1.0	1.7	0.2	0.3	11.2	9.6	8.8	14.1	14.0	1.1	4.7	ND	ND	ND	6.3
	PF9	avg	6.2	2.6	2.1	1.4	0.8	1.7	5.4	7.3	4.5	13.2	5.9	1.2	2.8	<0.2	<0.2	< 0.2	2.5
	11/	sd	10.4	3.1	2.5	1.9	0.1	1.6	8.1	9.5	6.3	17.3	9.0	0.6	3.5	ND	ND	ND	2.9
	PF10	avg	< 0.2	2.8	3.5	1.2	1.0	1.1	10.0	12.2	6.4	22.3	10.3	1.4	3.3	< 0.2	< 0.2	< 0.2	2.9
	1110	sd	ND	3.0	4.5	1.6	0.4	0.5	16.1	17.0	10.0	7.6	13.8	0.6	4.1	ND	ND	ND	3.3
	PF11	avg	20.8	35.8	22.2	18.5	2.2	4.8	125.0	115.1	93.0	141.0	141.1	9.1	47.2	4.3	3.3	1.5	34.5
		sd	19.7	57.8	34.0	27.6	0.1	0.6	207.5	189.0	153.9	234.4	232.9	12.6	75.7	7.1	5.4	2.2	54.1
	PF12	avg	0.7	1.6	1.8	0.8	0.4	1.0	2.6	2.1	2.6	2.7	3.1	1.4	1.9	< 0.2	<0.2	< 0.2	1.9
	1112	sd		0.6	0.9	0.3	0.0	1.0	2.4	2.4	2.5	2.3	3.3	0.5	1.3	ND	ND	ND	1.1
	PF13	avg	0.3	1.3	2.0	0.6	0.6	1.5	0.9	0.3	0.8	1.1	0.6	1.3	1.1	<0.2	<0.2	<0.2	1.2
	1110	sd	0.0	0.2	0.1	0.2	0.6	1.8	0.2	0.0	0.2	0.1	0.1	0.3	0.2	ND	ND	ND	0.2
	PF14	avg	11.4	33.7	95.5	5.5	2.0	12.6	210.2	164.7	110.6	107.0	90.5	6.5	31.7	2.3	1.5	0.9	22.0
		sd	15.1	55.3	161.6	7.8	1.4	14.0	357.5	281.0	186.1	181.6	152.2	8.6	51.5	3.6	2.3	1.2	33.1

Table A2. Cont.

			Ν	Ant	Phe	Acy	Ace	Fl	Flu	Ру	BaA	Chry	BbjFlu	BaP	BkFlu	IndPy	BPI	DBA	BeP
	NT ERL ERM		35 160 2100	45 85,3 1100	 240 1500	- 44 640	_ 16 500	_ 19 540	110 600 5100	 665 2600	_ 261 1600		_ _ _	30 430 1600	20 	70 — —	55 — —	 	_ _ _
	PF15	avg	26.9	59.0	67.3	25.1	6.6	13.4	265.5	212.1	168.8	218.3	229.7	13.8	79.3	7.6	4.9	2.3	58.6
	PF16	avg	29.9	18.0	14.5	10.2	4.8	6.6	73.7	61.9	51.9	74.1	81.6	5.4	28.9	1.7	0.5	0.7	22.1
	PF17	avg	4.1	7.3	6.3	5.7	1.3	4.1	30.2	25.7	24.1	32.3	41.7	3.3	15.6	< 0.2	< 0.2	< 0.2	11.7
	PF18	avg	30.5	2.0	2.2	2.0	1.9	4.4	4.0	3.0	3.0	3.3	5.4	1.7	3.0	< 0.2	< 0.2	< 0.2	2.6
	PF19	avg	30.5	1.6	1.7	1.4	0.9	2.7	2.9	2.4	2.6	2.4	4.1	1.4	2.3	< 0.2	< 0.2	< 0.2	2.1
Off-shore	PF20	avg	5.3	1.3	1.4	0.9	< 0.2	1.8	1.6	0.9	1.1	0.9	1.8	1.2	1.5	< 0.2	< 0.2	< 0.2	1.5
	PF21	avg	38.4	1.9	1.9	1.7	1.9	5.1	2.1	1.0	1.3	1.1	1.5	1.7	1.7	< 0.2	< 0.2	< 0.2	1.7
	PF22	avg	9.1	1.3	1.4	1.2	1.4	3.8	1.8	1.0	1.3	1.2	2.0	1.2	1.5	< 0.2	< 0.2	< 0.2	1.5
	PF23	avg	2.0	2.2	2.7	1.2	0.4	2.2	7.1	5.5	5.3	6.5	7.0	1.4	3.1	< 0.2	< 0.2	< 0.2	2.8
	PF24	avg	< 0.2	1.4	0.9	0.8	< 0.2	< 0.2	4.5	3.3	4.3	4.3	6.0	1.3	2.6	< 0.2	< 0.2	< 0.2	2.4
	PF25	avg	0.4	1.6	1.9	1.1	< 0.2	1.5	5.9	5.1	4.0	4.4	5.9	1.4	2.9	< 0.2	< 0.2	< 0.2	2.5
	DD 10	avg	27.6	38.1	77.0	17.1	7.6	15.2	178.1	149.6	128.1	169.1	248.8	13.8	83.2	14.9	11.0	4.2	40.9
	PB10 s	sd	38.2	43.8	105.8	21.5	10.1	18.5	230.6	192.2	164.3	220.7	318.8	17.0	108.1	9.4	7.0	2.4	50.6
	DBO	avg	18.2	3.6	5.6	1.8	1.5	3.0	19.3	21.4	12.6	17.0	18.0	2.3	6.1	< 0.2	< 0.2	< 0.2	5.8
	PB9 sd	sd	24.5	4.2	7.2	1.7	1.0	2.5	36.1	35.9	21.8	30.5	31.7	1.8	9.6	ND	ND	ND	8.2
	PB8 avg	9.9	8.1	12.8	5.3	1.3	4.2	86.6	82.0	72.2	92.4	114.3	7.5	36.9	6.5	4.2	1.7	27.7	
	T Do	sd	19.4	5.4	7.1	4.1	0.8	2.3	95.9	88.2	82.9	104.7	127.5	7.3	41.7	4.6	3.6	1.0	29.0
	DB7	avg	22.0	10.9	21.2	4.8	1.7	8.1	60.0	63.3	2.1	54.1	57.8	4.3	18.9	0.9	0.5	0.4	14.9
	T D7	sd	33.2	11.3	22.5	5.0	1.5	6.5	73.0	84.5	2.2	65.1	71.5	4.0	23.3	1.4	0.7	0.5	17.0
	PR6	avg	22.8	17.9	29.8	5.3	2.6	6.2	90.5	95.7	1.7	72.1	65.8	4.7	23.4	2.9	0.7	1.0	15.1
Piombino	1 00	sd	9.1	26.5	46.9	5.9	2.1	4.6	135.8	109.9	2.6	100.0	82.8	4.7	28.7	2.2	1.0	0.7	17.6
Tiomonio	PB5	avg	32.1	1.7	1.9	1.3	1.4	3.5	2.8	3.5	2.1	3.2	3.3	1.2	1.7	< 0.2	< 0.2	< 0.2	1.7
	1 00	sd	10.3	0.7	0.8	0.3	0.5	1.7	1.8	0.1	1.6	2.2	1.9	0.4	0.7	ND	ND	ND	0.7
	PB4	avg	39.7	1.0	0.9	0.7	1.3	0.5	1.0	0.8	0.7	1.3	1.2	1.0	1.1	< 0.2	<0.2	< 0.2	1.1
	101	sd	47.3	0.2	0.3	0.3	0.6	0.2	0.4	0.2	0.4	0.6	1.0	0.2	0.4	ND	ND	ND	0.4
	PB3	avg	4.4	0.8	0.6	0.3	0.4	0.2	0.7	0.3	0.4	0.3	0.6	0.8	0.7	<0.2	<0.2	<0.2	0.8
		sd	8.5	0.2	0.3	0.1	0.3	0.1	0.4	0.2	0.1	0.1	0.6	0.1	0.3	ND	ND	ND	0.3
	PB2	avg	10.4	1.2	1.1	0.8	1.5	2.6	0.9	0.3	0.7	0.3	0.6	1.2	1.0	<0.2	<0.2	<0.2	1.1
		sd	11.8	0.1	0.1	0.4	0.6	1.4	0.3	0.2	0.1	0.1	0.4	0.1	0.1	ND	ND	ND	0.1
	PB1	avg	20.5	1.0	0.9	0.8	0.8	2.2	0.7	0.3	0.6	0.2	0.7	1.0	0.8	<0.2	<0.2	<0.2	0.9
		sd	15.8	0.2	0.2	0.1	0.4	0.6	0.3	0.2	0.2	0.2	0.5	0.2	0.2	ND	ND	ND	0.2

			Al	As	Cd	Cr tot	Cu	Fe	Hg	Ni	Pb	V	Zn
	NT		_	12	0.3	50	_	_	0.3	30	30	_	_
	ERL		_	8.2	1.2	81	34	_	0.15	20.9	46.7	_	150
	ERM		_	70	9.6	370	270	_	0.71	51.6	218	_	410
	DE1	avg	2.50	53.69	0.28	98.11	29.90	2.33	0.47	72.39	111.80	83.05	276.78
	PFI	sd	0.66	9.56	0.40	7.17	18.01	0.46	0.62	5.66	144.60	12.72	380.91
	PF2	avg	2.31	49.10	0.19	111.56	26.76	2.42	0.68	85.62	90.47	85.92	223.16
		sd	0.57	4.21	0.27	12.39	10.89	0.68	1.01	8.02	124.96	3.59	310.57
	PF3	avg	2.47	52.95	0.25	107.63	30.41	2.44	0.39	78.53	147.12	89.34	320.95
		sd	0.63	5.53	0.31	22.07	15.49	0.74	0.35	13.74	176.62	6.53	397.61
	PF4	avg	1.40	51.16	0.05	65.81	17.20	1.62	0.21	46.71	43.54	70.24	82.77
		sd	1.11	22.36	0.03	38.52	13.99	0.98	0.29	29.11	61.66	22.82	112.94
	PF5	avg	1.67	72.06	0.08	85.04	37.87	2.36	0.76	64.38	104.47	75.43	248.91
		sd	0.81	16.71	0.05	31.15	34.03	0.77	0.90	15.37	115.96	19.08	296.86
	DEC	avg	1.35	59.81	0.04	70.07	16.61	1.59	0.09	54.91	15.32	86.66	36.46
	PF6	sd	0.59	3.51	0.00	20.69	4.12	0.31	0.06	14.49	9.70	7.75	16.17
	PF7	avg	1.64	50.72	0.05	105.29	25.94	2.27	0.34	88.64	66.51	76.90	105.49
Destation		sd	0.37	8.13	0.04	18.95	9.07	0.51	0.42	14.76	75.19	3.67	98.58
Portoferraio	PF8	avg	1.13	56.44	0.03	80.55	13.38	1.69	0.08	60.12	12.35	52.16	36.56
		sd	0.49	3.63	0.00	20.41	4.37	0.37	0.08	25.20	4.89	13.58	11.98
	DEO	avg	0.74	34.57	0.03	71.41	11.15	1.32	0.03	35.62	9.30	31.22	33.13
	PF9	sd	0.38	15.48	0.01	28.94	7.21	0.49	0.03	22.45	5.83	7.51	11.79
	DE10	avg	0.68	36.87	0.01	63.74	8.69	1.38	0.02	26.07	6.27	30.16	29.55
	PFIU	sd	0.15	14.51	0.01	13.16	1.68	0.32	0.02	4.79	3.95	7.54	10.68
	DF11	avg	1.36	37.87	0.03	64.87	11.09	1.83	0.04	38.63	6.64	43.72	41.40
	PFII	sd	0.27	13.12	0.01	9.71	0.99	0.17	0.02	4.98	2.68	11.99	4.10
	DE10	avg	1.59	36.70	0.03	89.61	14.34	1.98	0.05	43.36	14.35	47.70	43.54
	PF12	sd	0.30	13.59	0.01	19.37	4.64	0.44	0.03	7.54	8.87	7.27	15.73
	DE12	avg	1.60	30.80	0.04	108.61	12.59	1.63	0.02	46.02	6.23	45.46	32.19
	PF13	sď	0.91	5.09	0.02	27.13	0.95	0.31	0.01	10.54	0.98	11.86	9.34
	DF14	avg	1.36	44.46	0.02	75.21	10.52	1.93	0.09	37.65	45.44	91.32	55.30
	1714	sd	0.27	12.31	0.02	21.05	4.57	0.51	0.08	8.00	42.36	85.30	24.42

Table A3. Trace Metals Concentration (Average and Standard Deviation). Units: mg/kg d.w. (Al and Fe: % d.w.).National Thresholds (NT, According to DL56/2009), ERL and ERM (according to Long et al., 1995) are also reported.

			Al	As	Cd	Cr tot	Cu	Fe	Hg	Ni	Pb	V	Zn
	NT		_	12	0.3	50	_	_	0.3	30	30	_	_
	ERL		_	8.2	1.2	81	34	_	0.15	20.9	46.7	_	150
	ERM		_	70	9.6	370	270	-	0.71	51.6	218	_	410
	PF15	avg	1.74	39.74	0.05	81.47	28.63	3.12	0.25	61.49	40.91	67.19	124.96
	PF16	avg	1.38	50.87	0.04	109.47	20.85	2.81	0.18	50.29	25.91	59.12	89.05
	PF17	avg	0.92	60.99	0.04	66.73	25.59	2.83	0.17	43.81	34.96	54.52	84.13
	PF18	avg	0.36	54.25	0.02	37.77	8.46	1.84	0.04	19.08	10.89	39.77	44.82
	PF19	avg	0.47	85.26	0.04	67.15	26.17	3.48	0.05	30.97	22.84	51.54	70.13
Off-shore	PF20	avg	0.42	73.67	0.02	28.47	11.47	1.57	0.06	14.75	10.97	43.69	31.03
	PF21	avg	0.27	73.14	0.01	15.38	4.87	1.05	0.04	7.65	8.52	39.51	16.79
	PF22	avg	0.48	63.50	0.02	28.13	4.60	1.13	0.06	16.68	10.89	43.74	18.57
	PF23	avg	0.53	75.88	0.02	48.01	4.52	1.49	0.05	34.72	8.57	45.22	24.41
	PF24	avg	0.73	66.44	0.02	76.02	5.80	1.50	0.13	37.25	8.64	50.98	32.07
	PF25	avg	0.37	81.82	0.02	34.17	4.54	1.47	0.04	25.22	9.48	51.45	30.92
	DD10	avg	2.47	54.32	0.16	67.61	38.72	3.50	0.33	63.91	43.83	56.12	197.86
	PB10	sd	0.32	12.09	0.09	7.44	2.86	0.17	0.22	9.80	44.20	14.61	123.26
	DPO	avg	1.46	41.87	0.07	51.70	20.82	2.02	0.18	35.34	4.29	36.33	62.59
	P D9	sd	0.88	17.59	0.05	8.01	15.19	1.10	0.13	11.17	3.53	19.37	49.05
	DDO	avg	2.22	44.85	0.10	54.11	30.04	2.66	0.32	43.86	23.23	46.42	93.56
	PB8	sd	0.79	4.94	0.03	4.69	11.90	0.74	0.12	12.76	14.17	9.49	41.75
		avg	2.20	54.36	0.14	59.28	30.77	2.73	0.22	43.52	19.37	56.90	102.91
	PB/	sd	1.18	13.66	0.11	10.38	19.85	1.01	0.16	15.03	17.57	11.27	75.60
	DDC	avg	1.01	29.21	0.05	63.78	13.05	1.51	0.11	27.01	10.31	26.81	50.99
D' 1'	P D0	sd	0.54	4.94	0.02	10.10	8.23	0.58	0.02	7.73	10.35	9.42	38.21
Piombino	DDE	avg	1.18	25.96	0.04	89.23	11.23	1.52	0.05	32.08	4.09	29.52	42.41
	PD3	sd	1.16	4.87	0.02	13.87	11.07	1.02	0.03	16.95	3.22	17.36	26.47
	DD4	avg	0.31	20.94	0.02	49.21	2.94	0.70	0.02	13.50	1.82	12.95	13.06
	PD4	sd	0.10	6.18	0.01	25.56	1.28	0.27	0.01	5.68	0.86	3.88	10.75
	DD2	avg	0.38	30.99	0.02	60.94	2.69	0.86	0.02	17.88	1.69	14.09	16.34
	PD3	sd	0.16	14.01	0.00	31.96	1.15	0.31	0.01	4.16	1.22	4.84	11.11
	DDO	avg	0.42	26.51	0.03	62.68	2.84	0.79	0.02	18.72	1.93	15.61	13.33
	PB2	sd	0.12	8.57	0.01	21.73	1.32	0.30	0.01	4.34	1.12	4.58	9.36
	DD1	avg	0.39	20.07	0.02	39.38	2.91	0.75	0.02	13.35	2.39	13.60	12.76
	LR1	sd	0.14	10.12	0.00	13.35	1.60	0.35	0.01	2.71	0.62	3.08	9.69

Table A3. Cont.

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