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Organic pollutants (PCBs, PCDD/Fs, PAHs) and toxic metals in farmed mussels from the Gulf of Naples (Italy): Monitoring and human exposure

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ABSTRACT

In this study, we analyzed contaminant levels in mussels (*Mytilus galloprovincialis*) farmed into the waters of the Gulf of Naples and Domitio littoral (Italy), areas heavily influenced by human activities, especially the highly productive zones such as the harbor area. This marine environment is known to be polluted by a mixture of inorganic and organic contaminants: heavy metals (lead, cadmium and mercury), persistent organic pollutants (PCB and dioxins) and polycyclic aromatic hydrocarbons (PAHs).

In order to protect the health of consumers, mollusks farmed in the Gulf of Naples were analyzed to verify the presence of these contaminants. For the monitoring program, the mussels were repeatedly sampled during a period of four years (2016–2019).

The results demonstrate the substantial food safety of the mussel farmed in this area with regard to heavy metals and organochlorinated persistent pollutants. The presence of PAHs linked to seasonal phenomena and due to the contamination of marine sediments remains a critical factor.

1. Introduction

The monitoring of marine chemical pollution and its impact on human health and ecosystems are priority actions within the recent European Directives (EC, 2008). Different anthropogenic activities, such as industry, maritime traffic, agriculture, fisheries and the dense urban settlements can be considered potential sources of seawater pollution and a threat for fishery resources and coastal ecosystems (Piazzese et al., 2019).

The Gulf of Naples, is a marginal basin of the southeastern Tyrrhenian Sea, that is semi-enclosed by the Bay of Pozzuoli in the northern part, the Bay of Naples in the northeastern, and the Gulf of Castellammare in the southeastern part (Tornero and Ribera d'Alcalà, 2014). In particular, this area has not only a heavy commercial and touristic traffic due to harbor of Naples, but also a high population density that are a source of municipal and commercial discharges. Moreover, the Gulf of Pozzuoli is in particular characterized by intensive fishing as well as mussel cultivation and production facilities (Arienzo et al., 2017; Esposito et al., 2017; Qu et al., 2018). Therefore, due to human activities, coastal areas receive potentially hazardous chemicals, which could affect the environmental quality of marine ecosystems and, as a consequence, the human health (Fasano et al., 2018).

Among ubiquitous chemical contaminants there are heavy metals, as lead (Pb), cadmium (Cd) and mercury (Hg), and organic substances, such as, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), regulated by the Stockholm Convention, which aims to remove or restrict the production and use of Persistent Organic Pollutants (POPs) (Stockholm Convention, 2001; Chiesa et al., 2016). PCBs and PCDD/Fs are POPs widespread in the abiotic and biotic environmental compartments as complex mixtures of their congeners. They enter the aquatic environment through atmospheric deposition, urban and industrial discharge as well as riverine inputs and, because of their high stability, low volatility and lipophilic nature, they have a strong tendency to accumulate and persist in the suspended matter, sediments and marine organisms (Wang et al., 2016; Panseri et al., 2019) which represents a risk for human health (Okay et al., 2009).

Incomplete combustion of organic matter, especially fossil fuels, oil spills and its products, as well as their post-depositional

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transformation result in production of polycyclic aromatic hydrocarbons (PAHs).

Most of these organic chemicals show immunotoxic and carcinogenic activities and they can also cause endocrine disruption, reproductive, neurobehavioral and developmental disturbances (Barone et al., 2014; Wang et al., 2016). Similarly, heavy metals are a widespread class of carcinogenic and mutagenic pollutants, that increase incidence of chronic diseases (Liang et al., 2019).

Therefore, the environmental contamination and the resulting bioaccumulation and biomagnification of toxic substances in marine biota, results in a food safety problem due to the trophic transfer of these substances through food chain (Barbosa et al., 2019).

In order to ensure an efficient protection of public health, the European Commission has set maximum limits for certain contaminants in foodstuffs (EC, 2006) including seafood.

Bivalve mollusks, are good bioindicators used to assess levels and trends of coastal chemical contamination, due to their filter-feeding behavior and sedentary life, that leads to the accumulation of xenobiotics in their tissues proportionally with levels in seawater (Andral et al., 2004; Trinchella et al., 2013; Galgani et al., 2014; Carella et al., 2018; Fiorito et al., 2019; Grbin et al., 2019). For this purpose, the use of mussels, also the transplanted specimens, from an unpolluted area is a good strategy for biomonitoring environmental status of the coastal waters (Kucuksezgin et al., 2013; Spada et al., 2013; Bajt et al., 2019).

In Campania region of Southern Italy, *Mytilus galloprovincialis* represents among bivalve molluscs, the most important species from the point of view of production and consumption, therefore it is subject to intense control aimed to protect human health.

The aim of this study was to investigate the levels of some contaminants, monitoring heavy metals, PAHs and persistent organic pollutants, such as polychlorinated biphenyls and dioxins (PCDD/Fs and dioxin like PCBs) in *M. galloprovincialis* sampled in the coastal waters of the Campania region during four years, in order to assess potential health risks for seafood consumers.

2. Materials and methods

2.1. Sampling

Samples of *M. galloprovincialis* were collected from 17 sea farms located along the coastal areas of the Campania region, from January 2016 to December 2019 (Fig. 1).

Sampling stations were distributed in three different sites, Domitio littoral, denoted as S1, Gulf of Pozzuoli, S2, and Gulf of Naples and its east coast, up to the cities of Torre Del Greco and Torre Annunziata, S3. These areas are characterized by a high population density and different pollution degree.

In order to evaluate chemical contamination levels, concentrations of heavy metals (Cd, Hg, Pb), PAHs, PCBs and PCDD/Fs were determined and the results compared with the maximum limits according to EU Regulations (EC, 2006).

To minimize variability and to obtain a representative sample, each mussel sample was obtained from about 40 individuals of similar lengths (range 4–7 cm) and weight (3–5 g). Soft tissues of mussels were removed from the shells and stored at −20 °C until analysis.

2.2. Heavy metals analysis

Mussel samples were thawed and homogenized. Then, 0.75 ± 0.01 g were weighted and after acid digestion, metals concentration were measured by an atomic absorption spectrophotometer, with a transverse heated graphite furnace (GF-AAS) with Zeeman effect for electrothermal atomization and equipped with an autosampler (mod. PinAAcle 900T, Perkin Elmer, Italy).

The system was coupled to an interfaced computer running WinLab software (Perkin Elmer, Italy). Hollow cathode lamps were used as line sources for both elements. The absorption wavelength was 228.8 nm and 283.3 nm for cadmium and lead, respectively. Argon was used as the inert gas, and internal flow was set at 250 ml/min.

Total mercury was determined by atomic absorption spectrometry (spectrophotometer mod. 5010, Perkin-Elmer, Italy) after reduction of mercury in volatile hydrides by sodium tetrahydroborate.

Each sample solution was analyzed in duplicate and the mean results reported as mg/kg wet weight (w.w.) of tissues analyzed.

The detection limits for the atomic absorption spectrophotometer were: 0.030 mg/kg, 0.005 mg/kg and 0.020 mg/kg for Hg, Cd and Pb, respectively.

For quality assurance, chemical blank determinations were regularly made, together with each batch of samples, to check the purity of reagents and for possible laboratory contamination. Data quality assurance also includes certified reference material (mussel tissue ERM®- CE278k).

2.3. PCDD/Fs and PCBs analysis

Determination of PCDD/Fs and PCBs congeners in mussel samples was carried out by an isotopic dilution method, according to Lambiase et al. (2017). The analytical procedure is based on samples extraction and clean-up. The two fractions used to analyze PCDD/Fs and PCBs were eluted from carbon column and alumina column respectively and evaporated to dryness under a nitrogen stream. After adding a recovery standard solution ($^{13}C_{12}$- labeled congeners), identification and quantification of PCDD/F and PCB congeners were performed by high-resolution gas chromatography coupled to high-resolution mass spectrometry (Thermo Scientific DFS).

2.4. PAHs analysis

Determination of PAHs in mussel samples was performed using a method following the performance criteria set out in Commission Regulation EU 836/2011 (Esposito et al., 2017). Briefly, 2 g of homogenized sample after saponification with 2 N ethanolic KOH were subjected to a liquid extraction with cyclohexane, then the extract was cleaned up through a SPE C18 cartridge. Chromatographic analyses of PAHs were performed by HPLC-FLD (mod. Alliance, Waters, Ireland). The limit of quantification (LOQ) of each PAHs was 0.2 μg/kg.

Analyses identified and quantified benzo[a]pyrene (BaP), benzo[a]anthracene (BaA), benzo[b]fluoranthene (BbF) and chrysene (CHR) selected as markers for the occurrence of the 16 EU priority PAHs in EU Regulations (EU, 2011a,b). Maximum allowable levels in food, for the sum of four PAHs have been fixed, with a separate maximum level for benzo[a]pyrene.

2.5. Statistical data analysis

All data were statistically evaluated using GraphPad Prism 8.3.1 statistic software. The Mann–Whitney test (GraphPad Prism 8.3.1) for independent samples, was performed to test significant differences between the two samples. A *p*-value less than 0.05 was considered statistically significant ($p<0.05$) and when the measured value was below the limit of quantification (LOQ) of the test method, we used the LOQ/2 value, except for PAHs for which lower bound concentrations were calculated on the assumption that all values of the four substances below the limit of quantification are zero (Reg EU 835/2011). The results are reported also as descriptive statistics through the box and whisker plots that display the data distribution through their quartiles.

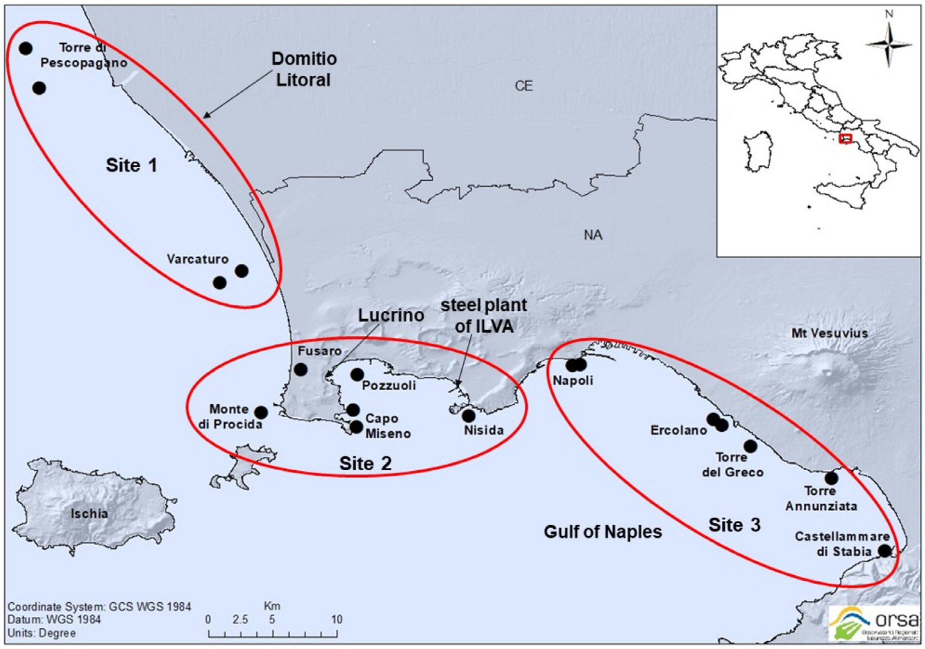

Fig. 1. Map of sampling sites of mussels in the Gulf of Naples (Campania region, Italy) from January 2016 to December 2019.

3. Results and discussion

3.1. Heavy metals

Table 1 shows the ranges, the means and standard deviations of heavy metal concentrations in 100 mussel samples collected in the three different sites, 19 samples in S1, 46 in S2 and 35 in S3.

All mean values were well below maximum levels set by Commission Regulation (EC) No 1881/2006 for Cd (1.0 mg/kg ww), Hg (0.5 mg/kg ww) and Pb (1.5 mg/kg ww) in bivalve mollusks and none of the samples analyzed exceeded the maximum allowable limit.

The Mann–Whitney test showed non-statistically significant differences of Hg levels in the three areas ($p>0.05$). On the contrary, the dataset highlighted a significant difference for Pb in S1 compared to S2 ($p<0.05$) and for Cd in S1 in relation to S2 and S3 ($p<0.05$).

Higher lead concentrations along the coastal area of Pozzuoli, S2, near to Lucrino (0.324 mg/kg), Bacoli (0.263 mg/kg) and Nisida (0.238 mg/kg), could be related not only to submarine hydrothermal springs and volcanic activity, but also to anthropogenic pressures caused by urban agglomerations, bathhouses and many shipyards. Furthermore, marine lead pollution has been attributed to the presence of a brownfield site, now undergoing remediation, in the Bagnoli area on the coast of the Gulf of Pozzuoli (Albanese et al., 2010; Trinchella et al., 2013; Trifuoggi et al., 2017). Lead concentrations were lower than amounts detected for sea farms along Campania coast during years 2007–2009 and along the West coast of Mediterranean Sea. Instead, these levels were in the range reported by other Authors (Arienzo et al., 2019) for the Gulf of Pozzuoli.

Comparisons with other studies revealed that the cadmium mean concentrations found in the three sites are in line with values obtained in the same areas during the previous years (Serpe et al., 2010; Fiorito et al., 2019), with slight increases in the maximum values, especially during the winter seasons. However, levels are lower than those measured along the French and western coasts of the Mediterranean Sea during 2004–2006 (Andral and Tomasino, 2007; Andral et al., 2011).

Table 1
Range, mean, standard deviation of Cd, Pb and Hg (mg/kg wet weight) in mussel tissues from three sampling sites from January 2016 to December 2019.

Site	N.[a]	Cd	Law limits[b]
1	19	0.02 - 0.13 (0.05 ± 0.03)	
2	46	0.01 - 0.29 (0.07 ± 0.04)	1.0
3	35	0.01 - 0.26 (0.07 ± 0.04)	
		Pb	Law limits[b]
1	19	0.03 - 0.40 (0.15 ± 0.10)	1.5
2	46	0.01 - 0.56 (0.26 ± 0.15)	
3	35	0.01 - 0.76 (0.25 ± 0.18)	
		Hg	Law limits[b]
1	19	0.03 - 0.10 (0.07 ± 0.05)	0.5
2	46	0.03 - 0.12 (0.06 ± 0.04)	
3	35	0.02 - 0.11 (0.07 ± 0.04)	

[a]Number of samples.
[b]Commission Regulation (EC) No 1881/2006 and subsequent amendments.

Finally, mercury concentrations were very low and almost constant in the three sites; their values were similar to those detected along Mediterranean coast in the last few years. It is important to underline that mercury is one of the most toxic elements in the environment and exposure to it can also occur through the consumption of seafood containing methyl-mercury in their tissues (Kuplulu et al., 2018).

On the basis of these data, the human health risk as a result of mollusk consumption was calculated by determining the Estimated Daily Intake (EDI), as follows: $EDI = C \times DC/BW$. Where C is the mean concentration of heavy metals in the sample (mg/kg); DC is the daily mussel consumption (g/*capita*/day) as reported by Food and Agriculture Organization Corporate Statistical Database and BW represents mean body weight (70 kg for an adult) (Squadrone et al., 2016).

Consumption of mussels reported for European Market Observatory for Fisheries and Aquaculture Products (EUMOFA) is approximately 1.27 kg/capita/year corresponding to 3.48 g/capita/day (EUMOFA, 2016; Chiesa et al., 2018). Therefore, EDI values for adult consumers calculated on the basis of mussels consumption and considering the highest mean concentration for

Table 2
Range, mean, standard deviation of PCDD/PCDF, DL-PCB and sum (pg WHO-TEQ/g fresh weight) in mussel tissues from three sampling sites.

Site	N.[a]	PCDDs/PCDFs	DL-PCBs	SUM
1	12	0.03-0.40 (0.12 ± 0.13)	0.14-0.87 (0.40 ± 0.24)	0.22-1.04 (0.53 ± 0.29)
2	45	0.04-1.40 (0.23 ± 0.23)	0.19-2.30 (0.97 ± 0.53)	0.24-2.62 (1.20 ± 0.58)
3	14	0.06-1.85 (0.38 ± 0.46)	0.38-3.72 (1.90 ± 0.89)	0.44-4.55 (2.28 ± 1.07)

[a]Number of samples.

each metal were 0.003 μg/kg/day for Cd, 0.013 μg/kg/day for Pb and 0.003 μg/kg/day for Hg. The obtained EDI values were compared with the respective provisional tolerable daily intake (PTDI) recommended by the FAO/WHO Expert Committee on Food Additives and set at 1 μg/kg bw/day, 3.6 μg/kg bw/day and 0.57 μg/kg bw/day for Cd, Pb and Hg, respectively (WHO, 2011, 2016; Zhelyazkov et al., 2018).

In order to characterize the human health risk associated with contaminated seafood intake, the Target hazard Quotient (THQ) should be calculated for each metal, as follow:

$$THQ = \frac{10^{-3}x\,(EfxED_{tot}xDCxC)}{(RfDoxBW_{a}xAT)}$$

where EF represents exposure frequency (365 days/year), ED_{tot} is the exposure duration (70 years), BW is the average body weight (kg), AT is the period over which exposure is averaged (for carcinogenic substance 365 days/year x 70 years) and RfDo oral reference dose (mg/kg/day). As regard this latter, USEPA established a value of 1×10^{-3} for Cd, 2×10^{-3} for Pb and 3×10^{-4} mg/kg/day for Hg (USEPA, 2016).

Because different toxics can cause similar adverse health effects, combining target hazard quotients (THQ) from different metals is often suitable to assess cumulative effects. For this purpose, the hazard index (HI) was calculated adding THQ values of each analyzed element ($HI = \Sigma_i\ THQ_i$); a HI value of 1 or lower means pollutants are unlikely to cause non-cancer adverse health effects over a lifetime of exposure (EPA, 2018).

In our study, the calculated value of HI was 0.023 which is much lower than 1. It can be concluded that heavy metal concentrations of mussels farmed along the Campania coasts are not a concern and the consumption of this seafood does not represent a risk for consumer health.

3.2. PCDD/Fs and PCBs

A total number of 71 samples of farmed mussels were analyzed for 17 polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) and for 12 congeners of *dioxin-like* PCBs which exhibit toxicological profile similar to PCDD/Fs. Hence, minimum, maximum and standard deviation for PCDD/Fs, DL-PCBs and their sum were reported in Table 2. In order to facilitate risk assessment and regulatory compliance and to add up the toxicity of the different congeners, the toxic equivalency factors (TEFs) approach was used. Experimental results, regarding the individual dioxin and dioxin-like PCB congeners, were given in terms TCDD toxic equivalent (TEQ) (Reg EU 1259/2011; Wang et al., 2016).

The sum of PCDD/Fs and DL-PCB ranged from 0.22 to 1.04 pg WHO-TEQ/g wet weight in mussels sampled in S1, from 0.24 to 2.62 pg WHO-TEQ/g ww for samples coming from S2 and between 0.44 and 4.55 pg WHO-TEQ/g ww in S3.

With regards to the maximum limits established by Commission Regulation (EU) No 1259/2011 fixed at 3,5 pg/g of toxic equivalent (WHO-TEQ) for the PCDD/Fs and 6,5 pg/g for PCDD/FS plus DL-PCB in fish and fishery products, all mean values were compliant and below legal permissible limits.

The spatial distribution is provided in the Box–Whisker plots of median concentrations (Fig. 2), that show a gradual buildup in both PCDD/F and PCB levels in mussels, from S1 to S3. This trend were also underlined by Mann–Whitney test, which confirmed significant differences among the three sites of interest ($p<0.05$).

Due to the lack of data available on PCDD/Fs and DL-PCBs in mussels reared along the Mediterranean coasts in the recent years, particularly in the Southern Tyrrhenian Sea, analytical values were compared with those obtained in other areas of Mediterranean Sea and of Adriatic and Ionian Seas.

Current levels of mussel contamination by PCDDs/Fs in S1 were similar to concentrations measured in the North Adriatic Sea as well as the French coast, near to Marseille and Thau Lagoon. Instead, the PCDD/F levels in S2 and S3 were comparable with the western Mediterranean Sea data recorded between 2004 and 2006 (Bayarri et al., 2001; Caixach et al., 2007; Munschy et al., 2008; Di Leo et al., 2014).

Regarding the risk of exposure through the consumption of mussels, it should be taken into account that the most important route for human exposure to dioxins and dioxin-like PCBs is food consumption, contributing 90% of total exposure (Liem et al., 2000; Shen et al., 2017). The estimated dietary intake (EDI) was obtained combining seafood consumption data as previously reported, with the highest mean TEQ of the sum of PCDD/Fs plus dioxin-like PCBs in mussels, in relation to body weight. When a congener concentration was under the limit of detection (LOD), the value was assumed to be respective LOD (upper bound approach).

The estimated daily intake (0.11 pg/kg/day) was below PTDI (2.0 pg TEQ/kg bw/week) set by EFSA expert Panel on Contaminants in the Food Chain in 2018 (EFSA CONTAM Panel, 2018). Therefore, human exposure to PCDD/Fs and DL-PCB was not of concern and mussels farmed in the studied areas should be considered safe for human consumption.

In addition to 12 DL-PCBs congener analysis, 42 samples of mussels were analyzed for the six "indicator" non dioxin-like PCBs (NDL-PCBs) recommended by European Union. These compounds show biological activity and toxicological properties different from DL-PCBs. In fact, while DL-PCBs exert toxic effects on the endocrine system, binding to aryl hydrocarbon receptor (Ah), the NDL-PCBs could produce neurotoxic effects (Storelli et al., 2014).

The results of PCB determination are reported in Table 3, and they are expressed as sum of PCB congeners 28, 52, 101, 138, 153 and 180. PCB concentrations ranged from 1.24 to 1.98 ng/g ww in S1, from 1.20 to 8.42 ng/g ww in S2 and from 0.30 to 37.3 ng/g ww in S3. Statistical analysis showed significant differences between S1 and S2 ($p<0.05$) as well as between S1 and S3 ($p<0.05$); moreover, mean values follow an upward trend moving from the north to the south along the coast of the Gulf of Naples, similar to trend of DL-PCB (Fig. 2).

The higher levels of contamination measured near Pozzuoli and Naples could reflect the urban and shipping activities and could be linked to their proximity with harbors(Firth et al., 2019).

Therefore, by comparison to previous monitoring studies, a decrease in contamination levels was observed during the last few years. Mean values in our study were much lower than those observed in the Gulf of Naples and Pozzuoli Bay (Naso et al., 2005; Tornero and Ribera d'Alcalà, 2014), as well as levels measured

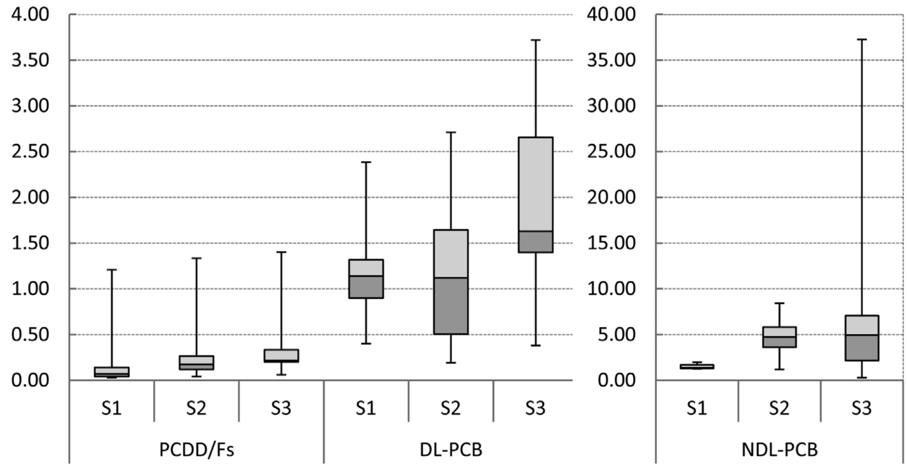

Fig. 2. Box–Whisker plot of median concentrations of PCDD/Fs, DL-PCBs (pg TEQ/g) and NDL-PCBs (ng/g) in mussels tissues from three sampling sites in the Gulf of Naples (Italy).

Table 3
Range, mean, standard deviation of NDL-PCBs (ng/g wet weight) in mussel from three sampling sites.

Site	N.[a]	NDL-PCBs
1	3	1.24 - 1.98 (1.53 ± 0.39)
2	17	1.20 - 8.42 (4.89 ± 2.00)
3	22	0.30 - 37.3 (6.73 ± 7.92)

[a]Number of samples.

from other Mediterranean and Adriatic regions in the same years (Perugini et al., 2004; Andral and Tomasino, 2007; Giandomenico et al., 2013).

The sum of six NDL-PCB in mussel samples was found to be lower than limit established by Commission Regulation (EU) No 1259/2011, set at 75 ng/g ww.

The EDI for the highest ΣPCB-6 was 0.33 ng kg^{-1}day^{-1}, much lower than the value of 10 ng kg^{-1}day^{-1}, the ADI recommended by the National Institute for Public Health and the Environment (RIVM) (Baars et al., 2001) and used to carried out risk characterization (Arnich et al., 2009; Giandomenico et al., 2016; Renieri et al., 2019).

With regard to the outcome of the study, farmed mussels from the three sites do not pose a health risk for consumers.

3.3. PAHs

Determination of PAH concentrations was conducted on 216 samples: 16 from S1, 164 from S2 and 36 from S3. Values of BaP and PAH4 (sum of BaA, BaP, BbF and CHR) are reported in Table 4 as range, mean and standard deviation. Since in our previous study (Esposito et al., 2017), high levels of PAH were found in S2, this site was further divided in three different locations: S2a, S2b and S2c. S2a (n = 40 samples) corresponds to the area of Capo Miseno (Bacoli), S2b (n = 79 samples) identifies the marine area of Lucrino in the center of the Gulf of Pozzuoli, and S2c (n = 45 samples) corresponds to the islet of Nisida, near the industrial area of Bagnoli well known for the presence of the disused steel plant (ILVA). Moreover, the Gulf of Pozzuoli is also characterized by an intensive commercial fishing supporting an active industrial sector, by military activities and pleasure boating, as well as an intense coastal urbanization, all considered potential sources of PAHs (Arienzo et al., 2017).

The PAH concentrations in S2b were markedly higher than levels measured in all other sites, as shown in Fig. 3. In particular, out of 80 samples analyzed, 12 (15%) resulted with a BaP content higher than the maximum limit of 5.0 µg/kg and 11 (14%) with a value of the sum of four PAHs greater than the maximum allowabled limit.

Moreover, the results confirmed a decrease in PAH concentrations during the spring–summer season and an increase in the winter season, as already reported (Esposito et al., 2017). In this regard, different physiological conditions in the mussel populations, related to different stages in their lifecycle, have been suggested as probable causes of seasonal variability (Arienzo et al., 2019). In addition, the trend of concentrations was also attributed to hydrodynamic processes of remobilization, suspension and transport of polluted sediments and to seasonal differences of biotic and abiotic factors regulating PAHs metabolic mechanisms and their bioaccumulation (Mercogliano et al., 2016).

PAHs leached by rains and percolating through contaminated soils are transported to water bodies (Arienzo et al., 2017). Being hydrophobic compounds, PAHs in seawaters aggregate with other suspended material and are deposited in sediments. Nevertheless, they have a certain water solubility, which affects their marine dispersion, spatial distribution and bioaccumulation (Ferrara et al., 2020).

Therefore, the relationship between PAHs levels observed in the mussels of the present study and levels measured in sediments of the area of the Gulf of Pozzuoli must also be considered (Albanese et al., 2010; Arienzo et al., 2017; Qu et al., 2018).

Previous works highlighted a common pyrogenic origin of PAHs and a relevant contamination of sediments that starting from the disused plant of Bagnoli, extends over the whole Gulf of Pozzuoli, due to sediment transport and water circulation. This mobility added to local anthropogenic activities could explain the highest levels of PAHs found in S2b.

Another source of contamination could be attributed to the plastic debris increasingly accumulating in aquatic environments. A study concerning the content of persistent organic compounds in plastic debris taken along coastal and oceanic areas showed that microplastics and nanoplastics constitute a carrier of PAHs. Because of degradation phenomena, due to marine current and atmospheric transport, they release low molecular weight PAHs (BaP) that could accumulate in marine organisms (Bouhroum et al., 2019).

4. Conclusions

The results of our study show that the marine area of the Gulf of Naples dedicated to *M. galloprovincialis* farming, guarantees

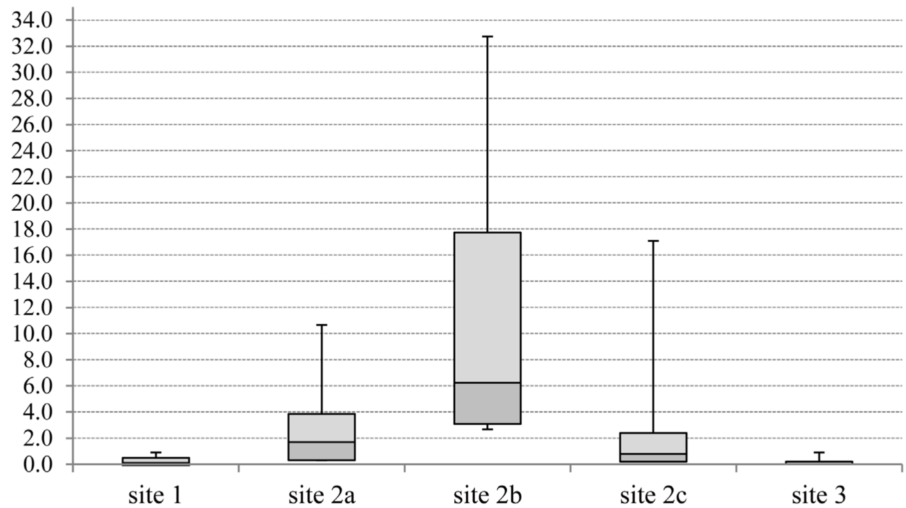

Fig. 3. Box–Whisker plot of median concentrations of BaP (μg/kg) in mussels from sampling sites in the Gulf of Naples (Italy).

Table 4
Range, mean ± standard deviation of values of Bap and PAH4 sum (mug/kg wet weight) in mussel tissues from three sampling sites in the Gulf of Naples (Italy).

Site	N.[a]	Bap	P-value	PAH4 sum
1	16	<0.2 - 0.8 (0.2 ± 0.3)		0 - 4.2 (0.1 ± 0.2)
2a	40	<0.2 - 7.5 (0.9 ± 1.3)	S2a *vs* S2b ($P < 0.05$)	0 - 23.5 (0.9 ± 1.6)
2b	79	<0.2 - 17.8 (3.4 ± 3.7)	S2b *vs* S2c ($P < 0.05$)	0 - 68.4 (2.9 ± 4.3)
2c	45	<0.2 - 15.4 (0.8 ± 2.3)	S2c *vs* S2a ($P < 0.05$)	0 - 56.4 (2.4 ± 8.4)
3	36	<0.2 - 0.9 (0.3 ± 0.6)		0 - 5.6 (0.1 ± 0.2)

[a]Number of samples.

the food safety of this product. The mussels from this area were resulted slightly polluted by toxic metals and organic pollutants.

In the four years of the investigation, levels of lead, mercury, cadmium, PCBs and dioxins did not exceeded the maximum limits for contaminants as established by European legislation. The only exception were the PAH concentrations which, in relation to a well-defined area of the Gulf of Pozzuoli, during the winter season reach high levels, in some cases above the maximum limits. For this reason, the area is subject to monthly monitoring in order to identify non-compliant mussel farms.

Further investigations may be needed in the near future to determine the presence of other emerging pollutants that may affect the quality and safety of seafood.

CRediT authorship contribution statement

Mauro Esposito: Conceptualization, Validation, Methodology, Writing - review & editing. **Silvia Canzanella:** Writing - original draft, Investigation. **Sara Lambiase:** Investigation, Visualization. **Alfredo Scaramuzzo:** Investigation, Visualization. **Raffaella La Nucara:** Investigation, Visualization. **Teresa Bruno:** Investigation, Visualization. **Giuseppe Picazio:** Investigation, Visualization. **Germana Colarusso:** Visualization, Supervision. **Roberta Brunetti:** Visualization, Data curation. **Pasquale Gallo:** Validation, Supervision.

Declaration of competing interest

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

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