Review

Romanian Danube River Hydrocarbon Pollution in 2011–2021

Crina Radu 1,2, Valentina-Mariana Manoiu 3,*, Katarzyna Kubiak-Wojcicka 4, Emilia Avram 1, Andreea Beteringhe 1,5 and Alexandru-Ioan Craciun 1,6

1 Faculty of Geography, University of Bucharest, Bulevardul Nicolae Balcescu 1, 010041 Bucharest, Romania
2 National Meteorological Administration, Sos. Bucuresti-Ploiesti 97, Sector 1, 013686 Bucharest, Romania
3 Department of Meteorology and Hydrology, Faculty of Geography, University of Bucharest, Bulevardul Nicolae Balcescu 1, 010041 Bucharest, Romania
4 Department of Hydrology and Water Management, Faculty of Earth Sciences and Spatial Management, Nicolaus Copernicus University, Lwowska 1, 87-100 Torun, Poland
5 Active Interventions in the Atmosphere, Str. Aurel Vlaicu 88, Jud. Iasi, 707252 Iasi, Romania
6 IUCN Europe Regional Office, Bd Louis Schmidt 64, 1040 Brussels, Belgium

* Correspondence: valentina.manoiu@geo.unibuc.ro; Tel.: +40-744-691-750

Abstract: This review paper aims to analyze studies conducted over recent years (2011–2021) on hydrocarbon pollution in the Danube’s Romanian sector. This involves looking at three main issues: Space-related Romanian Danube hydrocarbon pollution; the nature of samples and the types of tests used for hydrocarbon authentication; hydrocarbon effects on bioindicators and fish cell lines. The papers extracted for this review were selected from three scientific article platforms: Web of Science, Science Direct, and Google Scholar, by using keywords, a specific search protocol and various selection filters. The main results of the present analysis are the following: the highest levels of hydrocarbon contamination in suspended particulate matter and sediments were found in the sector Iron Gates-Călărași (2013), and the main pollution sources were industry, navigation and wastewater discharges; sediment and biological samples accumulate higher concentrations of hydrocarbons than water samples, and are a good indicator for these pollutants’ presence; the most widely used bioindicators are aquatic worms, mollusks, crustaceans, the wild common bleak, and, in the laboratory, fish cells; various methods are used in order to confirm hydrocarbon presence and/or their effects on biota: fluorescence, comet assay technique, micronucleus test, complementary passive samplers, in vitro bioassays, fugacity-based calculation model, sensors, oil spill modeling.

Keywords: hydrocarbon; water pollution; lower Danube; Romania; JDS3

1. Introduction

Water pollution is one of the most important environmental problems faced by water managers [1–3]. The problem is particularly evident in large river basins that are international in nature [4–6]. Establishing a common environmental policy for the management and protection of water resources requires joint action by all countries across a river basin [7–10]. The implementation of the Water Framework Directive (WFD) (Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000) [11] in the Member States of the European Union was a significant step forward. The WFD established the framework for Community action in the field of water policy. The objectives of the WFD are to achieve a good ecological status of all EU bodies of water and to protect them from pollution. It also aims to promote sustainable water use and to implement Integrated River Basin Management (IRBM). The implementation of integrated river basin resource management is particularly important in the context of climate change. According to the Intergovernmental Panel on Climate Change (IPCC) report (2013) [12], global warming is being observed, while climate forecasts predict further warming, as well as changes in the distribution and amount of precipitation. Changes in both temperature and precipitation are the most important physical effects of climate change on river ecosystems. As a result
of climate change, extreme hydrological conditions such as flooding, and droughts are observed [13]. Long-term rain-free periods contribute to drought, and affect not only the amount, but also the quality of water resources. Dilution of pollutants in the form of point discharges, e.g., from wastewater treatment plants, and diffuse or unknown pollution sources from agricultural land are of key importance [10]. Low flows mean less dilution volume and hence higher concentrations of pollutant discharges [14,15]. In addition, an increase in water abstraction—water taken to meet the demands of human activities (drinking water supply, irrigation of agricultural areas, supplying various industries) contributes to the reduction of water resources enabling the normal functioning of river ecosystems [1]. Any change in temperature affects the river’s ability to purify itself by reducing the amount of oxygen that can be dissolved and used for biodegradation [16].

In addition, the change in climatic conditions affects the distribution and fauna and flora species composition [17,18]. In recent years, climate change and increasing human pressure have influenced the functioning of river ecosystems not only in small catchments, but also in large transboundary basins. The current registry covers 310 international river basins and reflects changes in political boundaries. Common to 150 countries and contested areas, these basins cover 47.1% of the Earth’s land surface and account for 52% of the world’s population [19]. Dealing effectively with river pollution is a sizable challenge in many countries, while pollution and other problems on transboundary rivers seem particularly difficult to solve [4]. The most serious obstacles to internationally integrated river management do not appear to be technical, but political [4]. An example of a river of international importance is the Danube [20,21], which is a cross-border river flowing through ten countries. Its basin covers 19 European states [22–27].

The present paper aims to assess the Romanian Danube River hydrocarbon water pollution over the last decade by thoroughly reviewing 14 relevant studies (out of 1091 initially available works) from the Web of Science (WoS), Science Direct and Google Scholar platforms. The primary purpose of our work is to synthetically analyze the body of data that consists of scientific papers previously published on said topic. The second goal of the work is to comprehensively investigate three main issues: space-related Romanian Danube hydrocarbon pollution (stretches, sections or sectors of Danube subjected to water quality analyses for hydrocarbon detection); the nature of samples and the types of tests, methods and simulations that are used in order to attest hydrocarbon presence and/or their effects on biota; hydrocarbon effects on the targeted aquatic organisms (bioindicators) and fish cell lines. It is also relevant to acknowledge that, given the importance of the Danube River, 14 scientific articles that provide open access data regarding Romanian Danube hydrocarbon water pollution add up to a limited database that should be enriched.

2. Materials and Methods

2.1. Study Area

The total length of the Danube is 2860 km and covers a basin area of 817,000 km². The source of the Danube is located in southwestern Germany, and it eventually flows into the Black Sea. The Danube Delta is located in Romania (Figure 1).

In Romania, the Danube has a length of 1075 km between Bazias village and Sulina town, and is divided into four sectors: Bazias-Iron Gates (Danube Gorge), Iron Gates-Călărași, Călărași-Brăila (Danube’s “ponds”), and Brăila-Black Sea. As the Romanian Danube is the last main sector before the river flows into the Black Sea, high levels of pollution are expected here, given the intense social and economic activities, such as industry and navigation [28]. Other authors showed however that the most polluted sectors are the Danube’s upper course [29,30] and middle section [30–32] because of the pressure of large urban centers such as Vienna, Bratislava, Budapest and Belgrade, situated on the river’s banks [32].
While pollutant concentrations are expected to be elevated given the Danube’s considerable size, the river’s high dilution capacity must also be taken into account [33]. This dilution capacity is associated with an average flow of 6500 m$^3$/s, due to which concentrations decrease, but their mixture may still affect the health and behavior of exposed organisms [34].

Water is polluted when its initial quality state changes through degradation of its physical, chemical, or biological parameters. While these changes can be natural, the worst are often anthropogenic, as it is the case of the Danube River [35–44]. Unfortunately, the effects of anthropogenic pollution are associated with climate change [45]. Since 1998, water management in the Danube River Basin (DRB) has been overseen by the International Commission for the Protection of the Danube River (ICPDR) [46]. ICPDR already predicts that there will be serious seasonal disturbances inside the Danube basin [47].

Hydrocarbons are compounds that contain carbon and hydrogen molecules. The most dangerous in terms of water quality and which are found in large quantities are crude petroleum, refined petroleum products and individual polycyclic aromatic hydrocarbons (PAHs) contained by petroleum [11,13,22]. Petroleum consists of crude oils and refined oil products. Hydrocarbons comprise more than 75% of most crude and refined oils. Hydrocarbons in petroleum are divided into four categories: aliphatics, aromatics, resins and asphaltenes [48]. PAHs are aromatic hydrocarbons with two or more fused carbon rings that have hydrogen, or an alkyl group attached to each carbon atom [49].

Sources of petroleum and PAHs in water are industry (e.g., power generation), cities and towns (municipal discharges), pipeline spills, oil fields and refineries, offshore oil platforms, natural oil seep, shipping accidents, or intentional oil discharges [23,50]. Petroleum has negative effects on aquatic organisms (toxic action and habitat modification) and PAHs are environmental organic contaminants [51] that have carcinogenic and mutagenic effects, and bioaccumulate in human and animal tissue [52,53]. Overall, petroleum and PAHs effects are impaired reproduction, altered DNA, altered endocrine function, reduced growth and development, tumors or lesions, blood disorders, death etc. [50,53].

Water pollution with hydrocarbons and other pollutants was a warning signal that led to the establishment of the Convention on Cooperation for the Protection and Sustainable Use of the Danube River (Danube River Protection Convention), which resulted in Joint Danube Surveys (JDSs)—expeditions conducted in 2001, 2007, 2013 and 2019. These
surveys are very important for the collection and distribution of reliable information on water quality, mainly addressed in the Action Plan of the EU Strategy for the Danube River Basin under Priority Area 4 “To restore & maintain the quality of waters”. JDS3 was undertaken from 13 August to 25 September 2013 and 68 sites were sampled along a 2851 km stretch of the Danube, of which 26 are located in Romania [54,55], as mentioned in the Results section. Six papers featured in this review include data on water quality and hydrocarbon concentrations sourced and processed from JDS3.

2.2. Methods

In this paper, we used the internet web search technique to collect the scientific basis, by consulting the Web of Science, Science Direct, and Google Scholar academic literature databases. The search targeted English-written scientific articles, regardless of the type of paper, either journal papers, conference papers, or book chapters published in the last 11 years 2011–2021 (both for the year of publication and for the data used) in which concrete results appear for the Romanian Danube. In addition, both open access and limited access papers were analyzed. The search syntax used was a unitary one for all databases “Romania Danube hydrocarbon water pollution”. A complex syntax was chosen, as it contains all the keywords necessary for the proposed review. The final search was dated 8 March 2022.

Following the search, as it appears in Table 1, four results were found in Web of Science, 87 in Science Direct, and 1853 in Google Scholar. All results were analyzed in order to select only papers that met the established criteria, except for Google Scholar. Of the 1853 results, only the first 1000 could be analyzed, because the platform does not allow the display of those above this predetermined threshold. Therefore, the total number of works analyzed was 1091 (Table 1, Figure 2).

Most selected works were found in the beginning of the database search process, and were the most relevant. In the end, 17 papers remained, and because three of them were found in two different databases, the selected works were reduced to 14—both scientific articles and conference papers (Figures 2 and 3).

As shown in Table 2, most of the final selected papers were published in 2016 and 2020.

Table 1. Selected papers from databases.

| Database                | Number of Results | Selected Papers |
|-------------------------|-------------------|-----------------|
| Google Academic         | 1000/1853         | 11              |
| Science Direct          | 87                | 4               |
| Web of Science          | 4                 | 2               |
| Total                   | 1091              | 17              |
| Recurrent papers        |                   | 3               |
| Final number of papers  |                   | 14              |

Table 2. The publication year of the final selected papers.

| Publication Year | Number of Papers |
|------------------|-----------------|
| 2011             | 1 [56]          |
| 2015             | 2 [30,57]       |
| 2016             | 3 [32,58,59]    |
| 2017             | 1 [60]          |
| 2018             | 1 [34]          |
| 2019             | 2 [61,62]       |
| 2020             | 2 [29,31]       |
| 2021             | 2 [25,63]       |
Initial evaluation of Web of Science (WoS), Science Direct (SD) and Google Scholar (GS) scientific works based on keywords: 1091 studies

Primary database out of the search on WoS, SD and GS (251 studies)

1st selection of works according to the open access, WoS, SD and GS categories and document types

2nd selection of works according to the language, (no) temporal option, relevant information and thematical fit, and recurrent papers

Final bank of research work for review and analysis: 14 scientific articles

**Figure 2.** The methodical review process scheme.

**Figure 3.** The overall selections of scientific papers from the total results of the three databases.

As shown in Table 2, most of the final selected papers were published in 2016 and 2020.

The complete texts of the 14 papers were retrieved and assessed in-depth, and the results are presented and discussed in the next chapter.
3. Results and Discussions

As shown in Figure 4, paper occurrence frequency has an ascending trend in the second half of the analyzed decade. 42.9% of papers (6/14) used JDS3 data for the analysis of hydrocarbon concentrations in Danube water. The rest of 57.1% (8/14) used other types of data, such as those obtained by the authors.

![Figure 4](image-url)  
**Figure 4.** The frequency of paper occurrence in the last decade, and data used.

In general, the authors used water, sediment, and/or biological sample analyses in their studies. While many of them use water and sediment samples to compare the results or even all three samples [30,32,58], biological samples are always used in correlation with water and sediment analyses. Figure 5 shows the main samples used in hydrocarbon pollution analyses. The most widely used are water samples (11), as expected [46], and, for sediment and biological samples, the numbers are ten and six, respectively. The last two showed that sediments absorb a higher number of pollutants than water, they are a good indicator of hydrocarbon concentrations [30], and their effects on aquatic organisms are a priority.

![Figure 5](image-url)  
**Figure 5.** The main samples used in hydrocarbon pollution analyses of Danube River [25,29,31,32,34,56,63].

Six papers analyzed hydrocarbon pollution along the entire lower Danube sector (Figure 6).

While five of them used JDS3 data of 26 Romanian sampling stations (Table 3, Figure 7), it is noteworthy that Literathy [30] compares data from JDS1, JDS2, and JDS3 to highlight...
the evolution of PAH and petroleum hydrocarbon concentrations in the three types of samples (water, sediment and organisms), and Atanacković et al. [29] compares the data of the last two JDS.

Figure 6. The sites and sections sampled and analyzed on the entire Romanian Danube [25,29–32,58].

Literathy [30] used the fluorescence analytical method for PAH and petroleum hydrocarbons contamination measurements in water, suspended particulate matter (SPM), bottom sediment samples and biota (mussels). The scientist focused on anthracene, benzene, fluoranthene, naphthalene, benzo[a]pyrene (petroleum hydrocarbons) and benzo[b]fluoran-
The results showed that the highest level of hydrocarbon contamination in suspended particles, for JDS3 (2013), on the Romanian Danube, was downstream of the confluence with Argeș River (at km 432). Values of approximately 280 mg/kg were recorded in the Iron Gates–Calărași sector (approximately between JDS45 and JDS60) (Figure 7). The lowest hydrocarbon amount in SPM was measured in the Delta sector (below 50 mg/kg). The highest level of hydrocarbon contamination in sediments was found in the aforementioned sector, Iron Gates–Calărași, at km 450—approximately 500 mg/kg, and the lowest amount was determined in the Danube Delta sector (40–50 mg/kg). In our opinion, the high hydrocarbon pollution levels in SPM and sediments are generated by industry, municipal discharges and navigation.

Table 3. Romanian Danube JDS3 (26) sampling stations (according to Liška et al. [54]).

| Station Code | Location Name                                  | Country Code |
|--------------|-----------------------------------------------|--------------|
| JDS43        | BanatskaPalanka / Bazias                      | RS/RO        |
| JDS44        | Iron Gate reservoir (Golubac / Koronin)       | RS/RO        |
| JDS45        | Iron Gate reservoir (Tikija / Orșova)         | RS/RO        |
| JDS46        | Vrbica/Simijan                                | RS/RO        |
| JDS47        | Upstream Timok (Rudujevac / Gruia)            | RS/RO        |
| JDS48        | Timok (rkm 0.2)                               | RS/RO        |
| JDS49        | Pristal / NowoSel Harbor                       | RO/BG        |
| JDS50        | Downstream Kozlodouy                           | BG/RO        |
| JDS51a       | Upstream Olt                                  | RO/BG        |
| JDS51b       | /Olt km 0.4                                   | RO           |
| JDS52        | Downstream Olt                                | RO/BG        |
| JDS53        | Downstream Zimnicea/Svistov                  | RO/BG        |
| JDS55        | Downstream Jantra                             | RO/BG        |
| JDS57        | Downstream Ruse / Giurgiu                     | BG/RO        |
| JDS58        | Argeș                                         | RO           |
| JDS59        | Downstream Argeș/Oltenia                     | RO/BG        |
| JDS60        | Chiciu / Silistra                             | RO/BG        |
| JDS61        | Giurgeni                                      | RO           |
| JDS62        | Braila                                        | RO           |
| JDS63        | Siret (rkm 1.0)                               | RO           |
| JDS63a       | Upstream Prut                                | RO           |
| JDS64        | /Prut 1.0                                     | RO/MD        |
| JDS65        | Reni                                          | RO/UA        |
| JDS66/JDS2-93a | Viłkova—Chilia arm   | RO/UA        |
| JDS67        | Sulina-Sulina arm                             | RO           |
| JDS68        | St. Gheorghe-St. Gheorghe arm                 | RO           |

Figure 7. Romanian Danube JDS3 sampling sites (modeled on Liška et al. [54]).

Kolarević et al. [32], during August and September 2013, also used mussel species (Unio sp. and Sinanodonta woodiana) to detect the level of genotoxic pollution, caused especially by benzo[a]pyrene, by measuring the level of DNA damage in the haemocytes of these aquatic organisms. The Comet assay technique was used for DNA damage evaluation. Unio sp. was more sensitive to Danube River pollution compared to S. woodiana. Results
showed that section VIII (Western Pontic Section—rkm 942–375.5) and section IX (Eastern Wallachian Section—rkm 375.5–100) (Figure 6) were characterized by a relatively low degree of DNA damage to mussel species, with insignificant variations between sites. A higher level of DNA damage was detected in specimens downstream of the Zmínicea/Svistov ports (JDS53) (Figure 7), given that ports could be potential sources of genotoxic pollutants, mainly PAHs. A higher level of DNA damage was also found in specimens of *Unio sp.* in the Danube Delta (JDS66 and 67) (Figure 7), considered to be a consequence of pollutant accumulation in delta sediment. We underline that the level of DNA damage in mussels was in correlation with the concentrations of hazardous priority substances (PAH) in SPM and sediment.

Genotoxicity as a marker for hydrocarbon pollution exposure in wild common bleak (*Alburnus alburnus*) was the focal point of the investigation conducted by Deutschmann et al. [58] in August and September 2013. In this regard, two surveys were used: micronucleus frequency and comet tail intensity of fragmented DNA material in erythrocytes. Micronucleus test (MN) and Comet assay (CM) are sensitive and rapid tests for DNA damage characterization. In addition, concentrations of petroleum hydrocarbons (among other various groups of pollutants) were measured in water, SPM and sediment.

The highest MN values were identified at JDS60 (Chiciu/Silistra—50 km downstream of Argeș River) (Figures 6 and 7) and JDS57 (downstream of Ruse/Giurgiu) (Figures 6 and 7), which we consider can be correlated to untreated effluents in the catchment area of the metropolitan region Bucharest/Ruse, while for CM, the highest genotoxicity potential was determined at JDS47 (upstream of Timok, Rudujevac/Gruia) (Figures 6 and 7) because of the highest anthracene concentration in SPM; the site is also located downstream of cities Drobeta-Turnu Severin (Romania) and Kladovo (Serbia), and their industrial activities and wastewater discharges contribute to the high genotoxic potentiality of those Danube sites.

The CM and MN in erythrocytes of wild fish are suitable long-scale monitoring tools for the assessment of the genotoxic potential of rivers, as the collection of fish blood for investigations was conducted without sacrificing the organisms. We emphasize that DNA damage in fish erythrocytes was mainly caused by untreated wastewaters (also containing hydrocarbons) of industrial centers or densely populated regions.

Belháčová-Minaříková et al. [31] analyzed the following PAHs: naphthalene, anthracene, fluoranthene, and benzo[a]pyrene, in sediment samples, during JDS3, in 2013. The highest values were detected at site JDS46 (Vrbica/Simijan–Serbia/Romania) (Figures 6 and 7), then a decline was seen, and PAHs concentrations slightly increased toward the Danube Delta, particularly for fluoranthene and benzo[a]pyrene, more present in crustaceans and mollusks. We see that navigation, industry and municipal wastewater discharges are recurrently the main pollution sources for the aforementioned PAHs, affecting the biodiversity of Danube stretches.

Atanacković et al. [29] analyzed the impacts of different pollutants, including two PAHs (benzo[b]fluoranthene and benzo[k]fluoranthene), on 51 taxa of aquatic worm (Oligochaeta) communities along the Danube River, for JDS3. As Oligochaetes filter mud, they are suitable bioindicators. According to these scientists, physico-chemical and chemical determinants in water and sediment (organic pollution, including PAHs, and nutrient load) are not the main factors that affect Oligochaeta communities and influence their distribution. Hydromorphological degradation is in fact the major one. To conclude, Oligochaeta could be used as indicator for hydromorphological degradation in large rivers.

Chitescu et al. [25] synthesized in their review paper (which highlighted the fact that only 10 WoS and Google Scholar studies investigated PAHs from 2010 to 2021) the fact that PAHs concentrations in Lower Danube water and sediment samples were high in the first years of the last decade (2011–2013) comparing to the values measured in the upper Danube, Danube middle stretch and other European river basins, thus indicating an organic pollution boost (following an increase in wastewater discharge, we consider), especially in the Danube Delta.
To conclude, at present, for the entire lower Danube sector, the main hydrocarbon pollution sources consist of industry, untreated municipal and industrial wastewater discharges and navigation (including ports). The methods used in order to validate hydrocarbon presence and/or their effects on biota were fluorescence, Comet assay technique (CM) and Micronucleus test (MN). The indicators used were mussels, wild common bleak and aquatic worms (Oligochaeta). The highest hydrocarbon amount in SPM was found downstream of the confluence with Argeș River, and in sediments—in the Iron Gates-Călărași sector (2013); a high DNA damage level was identified in mussels downstream of the Zimnicea/Svistov ports, and in fish erythrocytes—50 km downstream of Argeș River and downstream of Ruse/Giurgiu (2013).

Regarding the methodologies adopted to detect and measure environmental hydrocarbon pollution, in order to eliminate any doubt, we must stress the fact that each method generates particular, distinct information, and only the outcomes acquired using a singular, specific method can be considered for a comparative evaluation. For example, Literathy [30] applied the fluorescence method for PAH determination in water, SPM, sediments and mussels (biota) (including the Romanian Danube), and results presented similar trends, with concentrations below European Union Environmental Quality Standard values. In fact, the fluorescence approach was used in the first three JDSs. It provided information on hydrocarbon pollution of water, SPM and sediments, and it proved to be a useful and cost-effective instrument for a comparable analysis.

The CM technique (also known as single cell gel electrophoresis), on the other hand, is also a sensitive tool, but at the same time is simple, versatile and rapid, for DNA damage evaluation in eukaryotic and prokaryotic cells. Kolarević et al. [32] used this technique in mussels (also for the Romanian Danube), as they are important indicators of water, SPM and sediment pollution. Deutschmann et al. [58] applied both the CM and MN methods on erythrocytes of common bleak (*Alburnus alburnus*) in order to prove the efficiency of these two techniques for genotoxicity evaluation. MN is applied to determine the chromosomal damage as a mutagen exposure consequence. CM and MN genotoxicity results could be correlated to the hydrocarbon pollution in Romanian Danube water, SPM and sediments. We find that scientists may choose any of said methods for hydrocarbon determination depending on the analysis target, the type of analysis they want to perform, the objective of their study, and the hypothesis they wish to test.

Four papers analyzed hydrocarbon pollution in certain sectors of the Romanian Danube (Figure 8). Except for Novák et al. [34], the other authors used data collected outside of JDS3.

Stanescu et al. [57] used water and sediment samples collected in 2014 (four campaigns in February, April, June and October), from Bazias to Călărași (Figure 8), in order to analyze the ecological status. The hydrocarbons analyzed were oil products and PAHs. For these pollutants, the ecological status turned out to be good to very good because of a significant decrease in anthropogenic pressure, induced by a decline in industrial activities, and also due to an effective water self-purification process.

29 PAH compounds were sampled and subsequently analyzed (applying gas chromatography and mass spectrometry) by Novák et al. [34] using two types of complementary passive samplers: silicone rubber sheets (SR) for non-polar chemicals, and adsorption SDB-RPS Empore disks (ED) for non-polar, as well as for more hydrophilic compounds. Sample extracts were then characterized by a battery of in vitro bioassays covering endocrine disruption, xenobiotic metabolism, and adaptative stress responses. We highlight that SRs are more efficient in absorbing hydrocarbons, but, unfortunately, except for estrogenity and anti-androgenity (related to the untreated wastewater discharged into the Romanian Danube), only a small part of the biological effects could be demonstrated by the analyzed chemicals, and this is also influenced by a data shortage on biological consequences.

The transfer of volatile organic compounds (VOCs) and volatile aromatic hydrocarbons (BTEXs) from Olt River to the Danube (Figure 8) was also studied with a fugacity-based calculation model, using data collected in spring 2018, by Iordache et al. [62].
Figure 8. The sites and sections sampled and analyzed in certain stretches of the Romanian Danube [34,57,61,62].

The VOCs assessed were 1,2 dichloroethane and perchlorethylene, and the BTEXs analyzed were benzene, toluene, ethylbenzene, o-xylene, m-xylene, and p-xylene. The model relates the compound concentration from sediment, water or air to biota concentration, based on experimental data, and can be an effective tool for preventive measures in contaminated areas. The modeling revealed the tendency of investigated compounds to transfer from water to air.

For the Câlărași-Brăila stretch (km 375–km 175) (Figure 8) of the Lower Danube River, Radu et al. [61] aimed to assess the surface sediment quality in terms of hazardous substances content, including PAHs. Data was collected from ten monitoring sites, between September 2011 and August 2017. This study was carried out by using a set of statistical methods for surface sediment quality assessment, such as the Principal Component Analysis (PCA) and Cluster Analysis (CA). Based on the study’s results, we certify that PAH values did not exceed normal limits.

As a conclusion of these last four papers, none of the analyzed Romanian Danube sectors presented PAH high values and this is the consequence of industrial drop off.

Figure 9 illustrates the locations of the sites and sections sampled and analyzed in specific sectors of the Danube Delta.
likely cause of this high concentration of PAHs is wastewater discharge without adequate treatment.

Figure 9. The sites and sections sampled and analyzed in specific sectors of Danube Delta. [56, 59, 63].

Pascu et al. [56] investigated the physical and chemical quality of surface water and sediments in seven locations of the St. Gheorghe branch of the Danube Delta (Figure 9), in the period 2009–2011; this last year is important for the present review. Among other contaminants, concentrations of PAHs (fluoranthene, benzo[b,k]fluoranthene, benzo[a,g,h,i]pyrene, benzo[a]anthracene, indeno(1,2,3-cd)pyrene, naphthalene, phenanthrene, anthracene, chrysene, pyrene) were analyzed in water and sediment samples, using fluorescence detection. The presence of PAHs over official limits [64] was detected in sampling section S6—Murighiol (Figure 9), in the vicinity of the ship supply pontoon, in February 2011: 21.3 mg/kg, while the maximum admissible value was 1 mg/kg. The most likely cause of this high concentration of PAHs is wastewater discharge without adequate treatment.

Pérez-Albaladejo et al. [59] assessed the environmental quality of sediments collected from the Black Sea coast. We have to mention that the study areas were located around the mouth of the Danube River (Figure 9), close to the main harbors, wastewater treatment plant (WWTPs) effluents, tourist resorts and city-influenced areas. Scientists used a battery of bioassays based on the fish hepatoma cell line (PLHC-1 cells), zebrafish-Pxr(pregnane X receptor)-transfected COS-7 cells (fibroblast-like cell lines), and sea bass ovarian subcellular fractions, in order to determine cytotoxicity. Organic extracts of sediments influenced by the Danube River and collected near harbors and urban discharges showed significant cytotoxicity.

The highest cytotoxicity was detected in sediment samples collected from harbors (Constanta and Mangalia) and from the Danube mouth. Constanta harbor—80% average loss of cell viability—is strongly affected by heavy boat traffic, and urban and industrial discharges; Mangalia harbor, a smaller port used mainly as shipyard, is impacted by a mixture of both boat traffic and WWTP effluents. The areas influenced by the Danube mouth are associated with boat traffic, and urban, industrial and agricultural discharges from several European cities.
Canning et al. [63], using a sensor installed on a houseboat, assessed and analyzed methane (CH$_4$) surface water concentrations in the Danube Delta (Figure 9), on three expeditions held in three seasons in 2017 (spring, summer and autumn). CH$_4$ is formed in anaerobic milieus, particularly in sediments. Overall, the delta was a source of CH$_4$ throughout all seasons, with concentrations ranging between 0.113–15.6 µmol/L. The dataset was split into three different subsystems: lakes, rivers and channels, with channels showing the highest variability. Overlapping CH$_4$ concentrations were found in each subsystem, with large inflows coming from reed beds and channels into lakes. Seasonal variability and water flow direction also influenced the overall dynamics in each region. We assert that the spatial variability in and around lakes, reed bed edges and within channels should be the main areas of focus in terms of CH$_4$ release.

These last three studies stated that high PAHs concentrations in Murighiol’s water and sediment samples, and Constanta and Mangalia harbor sediment samples are mainly caused by inadequately treated WWTP effluents, harbor activity and boat traffic. Additionally, the Danube Delta is a source of CH$_4$ during all seasons.

In addition to all aforementioned papers, a study presenting the aftermath of a naval accident modeling with an impact on hydrocarbon pollution was also considered for this review [60]. Oil spill modeling showed the impact of hydrocarbon spillage (heavy fuel oil, diesel oil) on different Danube Delta species (amphibians, annelids, crustaceans, fishes, insects, mammals, mollusks, and birds). The model proved that, after the impact, 0.5 t evaporates in two hours, and almost 50 t in 48 h (from 1500 t). The most heavily affected species are insects, as 1% die in the first 48 h. The compact spill got to cover up to 0.3 km$^2$ in 48 h. Considering all the results presented in the study, a potential intervention area can be identified for ensuring good action timing and an appropriate outcome.

To support our review conclusions, we wanted to use remote sensing imagery, but unfortunately, the Landsat 8 satellite does not have a high enough spatial resolution to capture the spatial dynamics of hydrocarbons in the Danube area. Moreover, the temporal resolution (two weeks) makes it difficult to capture spills. After 2015, the year Sentinel-2 started transmitting imagery, bibliography sources do not cite the dates and exact locations of spills. We therefore deem it difficult or even unfeasible to use satellite imagery to identify hydrocarbon sources in the analyzed area and period.

Regarding the hydrocarbon concentrations measured in the Romanian Danube and their effects on bioindicators, the following main results must be highlighted (Table 4).

Even though our study did not aim to present solutions for reducing hydrocarbon pollution in the Romanian Danube, given how urgent it is to remove some of the most dangerous pollutants, we will briefly mention several likely effective ones. We believe that the current global economic, geo-political and military context continues to stimulate the use of fossil energy sources/fossil fuels, to the detriment of green, environmentally friendly, clean, and sustainable energy. On Web of Science, the most well-known website for valuable scientific papers, in the last 22 years only 11 open scientific articles have been published on possible ways to reduce hydrocarbon pollution in aquatic environments.

In 2001, Adewuyi [65] analyzed in a review paper the sonochemistry as an application for the destruction of organic pollutants (including different categories of hydrocarbons) in aqueous environments. These pollutants can be transformed by ultrasonic irradiation (or the combination of ultrasound and other advanced oxidation techniques) to inorganic ions, CO$_2$, and short-chain organic substances as final products.

Eight years later, Mahr and Chase [66] discussed in their paper a particularly useful and efficient oil spill detection and alarm sensor system for aqueous media, similar to a “smoke warning”, but tailored to oil spills, based on UV/fluorometry, and with a micron-level sensitivity.
Table 4. The evolution of hydrocarbon concentration levels in the Romanian Danube, causes and effects on bioindicators.

| Hydrocarbon Concentration Level | Study/Analysis Year | Sample Type Analyzed for Hydrocarbons | Romanian Danube Sector/Site | Main Hydrocarbon Pollution Sources | Effects on Bioindicators (if Highlighted) |
|---------------------------------|----------------------|---------------------------------------|----------------------------|-----------------------------------|------------------------------------------|
| PAHs over limits                | 2011                 | Water and sediments                   | Section 6-Murighiol (St. Gheorghe branch of the Danube Delta) (Figure 9) | Wastewater discharge without adequate treatment | -                                        |
|                                 |                      |                                       | Iron Gates-Călărași (approximately between JD645 and JD660) (Figure 7) | Industry, navigation and wastewater discharges | -                                        |
| Highest level                   | 2013                 | SPM and sediments                     | Downstream of Zimnicea/Svislov ports (JD535) (Figure 7) | Port and industrial activities | High DNA damage in mussel species *Unio sp.* and *Sinanodonta woodiana* |
|                                 | 2013                 | SPM and sediments                     | Upstream Timok-JD547 (Rudujevac/Gruia) and downstream of Drobeta-Turnu Severin and Kladovo cities (Figures 6 and 7) | Industrial activities and effluent discharges | Highest hydrocarbon genotoxicity potential on the wild common bleak |
| Highest anthracene concentration| 2013                 | SPM                                   | JDS57-downstream of Ruse/Giurgiu and upstream of Timok-JD547 (Rudujevac/Gruia) (Figures 6 and 7) | Untreated effluents | Highest hydrocarbon genotoxicity potential on the wild common bleak |
|                                 | 2011–2013            | Water, SPM and sediments              | Lower Danube and Danube Delta | Untreated or insufficient treated wastewater discharge | -                                        |
| Good to very good ecological status| 2014                 | Water and sediments                   | Baziaș to Călărași (approximately between JD543 and JD560) (Figures 7 and 8) | (low human pressure) | -                                        |
|                                 | 2015                 | Sediments                             | The Black Sea coast, around the mouth of the Danube River (Figure 9) | Harbor activities (Constanta and Mangalia) and urban, agricultural and industrial discharges | Significant cytotoxicity on fish cell lines |
| Normal limits according to the law (but higher than those of upper and middle Danube) | 2011–2017            | Sediments                             | Călărași-Brăila stretch (km 375-km 175, approximately between JD560 and JD62) (Figures 7 and 8) | Industry, navigation and wastewater discharges | -                                        |

In 2011, Olivella et al. [67] stressed in their article the effectiveness of *Quercus cerris* bark cork for hydrocarbon sorption in PAH-contaminated waters, and, one year later, Hao et al. [68] investigated cerium-loaded activated carbon (Ce-AC) as a catalyst for PAH ozonation, degradation and removal from water, and proved its efficiency. Trčković et al. [69], in 2016, presented another solution for the remediation of PAH contaminated sediments, namely carbon nanotubes. In 2019, Li et al. [70] proved that nanofiltration membranes with narrow pore size can remove water PAHs, and Maletić et al. [71] analyzed the remediation and limitation techniques for sediment PAHs, such as bioaugmentation, biostimulation, phytoremediation, electrokinetic remediation, surfactant addition and different sorbent amendments featuring activated carbon and biochar.

In 2021, Abdibattayeva et al. [72] designed a helio device, based on solar energy, and used it for a new treatment method to extract oil products from wastewater or solid waste. The following year, Ghouas et al. [73] succeeded to remove VOC from aqueous solutions using the cloud point extraction and a biodegradable nonionic surfactant. During the same year, 2022, Shahi Khalaf Ansar et al. [74] investigated in their review article the removal of PAHs using algae. The PAHs’ elimination depended on the algae species and the hy-
hydrocarbon type. The most efficient alga, *Selenastrum capricornutum*, degraded PAH by 78%, over a 7-day procedure, and the least efficient alga, *Chlorella vulgaris*, reduced PAH by 48%, over a similar treatment period. Finally, Kothiyal et al. [75] reviewed in their study recent PAHs removal approaches for different environments, such as biodegradation (with an efficiency of 73% to 92%, very useful for water sources, and using biomolecules such as biosurfactants, humic acids, proteins and enzymes; nanoparticles, such as silica nanoparticles; bacteria and fungi; algae; photodegradation (more efficient for the atmosphere and soil PAH pollution); chemical degradation (using oxidants such as ozone); phytodegradation (involving different types of grasses and leguminous plants and being efficient for soil PAH pollution); degradation by adsorption (practical for water PAH pollution and using different adsorbents); and wastewater filtration (forward osmosis, ultrafiltration, nanofiltration, reverse osmosis). The information above was presented in order to emphasize the advancements made over recent years in the different PAH degradation methods and techniques. Some of them can be used as part of future efforts to reduce hydrocarbon pollution in the Romanian Danube.

4. Conclusions

Given the severity of the issue, the number of studies on Danube hydrocarbon pollution in the lower sector (Romanian Danube) is still too low.

In the scientific studies conducted in the last decade, hydrocarbons are highlighted and analyzed in water, sediment and/or biological samples. Sediment and biological samples accumulate higher concentrations of hydrocarbons, and are a good indicator for these pollutants’ presence. The most widely used bioindicators are aquatic worms (Oligochaeta), mollusks (such as mussels), crustaceans, the wild common bleak, and in the laboratory—fish cells, e.g., zebrafish PXR-transfected COS-7 cells and sea bass ovarian subcellular fractions.

Different tests, methods and modeling are used in order to indicate hydrocarbon presence and/or their effects on biota: fluorescence analytical method, comet assay technique, micronucleus test, complementary passive samplers (SR and ED), in vitro bioassays, fugacity-based calculation model, statistical methods (e.g., PCA and CA), sensors, oil spill modeling.

From the results presented in the previous chapter, it is noticeable that, after 2013, Lower Danube sample hydrocarbon concentrations decreased to normal limits (except for sediment samples collected from the Black Sea coast, around the mouth of the Danube, where many pollutants accumulate), but were higher than those reported for the upper and middle Danube stretches.

Many hydrocarbons are defined as priority pollutants, as they are persistent, bioaccumulative and toxic for aquatic life. The Water Framework Directive set out the strategy to reduce the chemical pollution of European waters, including the Danube, the continent’s most important river, in order to protect water life.

As a final point, more investigations are needed to elucidate the correlations between hydrocarbon concentrations in Danube water, SPM and sediments, and their genotoxicity and cytotoxicity. The present review provides a synthetic illustration of the current level of examination, and can be a basis for future studies.

**Author Contributions:** Conceptualization, C.R., V.-M.M., K.K.-W., E.A., A.B. and A.-I.C.; methodology, C.R., V.-M.M., K.K.-W., E.A., A.B. and A.-I.C.; software, C.R., V.-M.M. and A.-I.C.; validation, C.R., V.-M.M., K.K.-W. and A.-I.C.; formal analysis, C.R., V.-M.M., K.K.-W. and A.-I.C.; investigation, C.R., V.-M.M., K.K.-W., E.A., A.B. and A.-I.C.; resources, C.R., V.-M.M., K.K.-W., E.A., A.B. and A.-I.C.; data curation, C.R., V.-M.M., K.K.-W., E.A., A.B. and A.-I.C.; writing—original draft preparation, C.R., V.-M.M., K.K.-W., E.A., A.B. and A.-I.C.; writing—review and editing, C.R., V.-M.M. and A.-I.C. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research received no external funding.

**Institutional Review Board Statement:** Not applicable.
Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The authors want to thank their colleague Marius Budileanu (Faculty of Geography, University of Bucharest) for his technical support regarding the Landsat 8 and Sentinel-2 imagery analyses.

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

References
1. Negm, A.M.; Zelenakova, M.; Kubiak-Wójcicka, K. Water Resources Quality and Management in Baltic Sea Countries; Springer Water: Cham, Switzerland, 2020; ISBN 978-3-030-39700-5.
2. Backhaus, T.; Brack, W.; Brink, P.V.D.; Deutschmann, B.; Hollert, H.; Posthuma, L.; Segner, H.; Seiler, T.-B.; Teodorovic, I.; Focks, A. Assessing the ecological impact of chemical pollution on aquatic ecosystems requires the systematic exploration and evaluation of four lines of evidence. *Environ. Sci. Eur.* 2019, 31, 1–9. [CrossRef]
3. Zelerajkova, M.; Kubiak-Wójcicka, K.; Weiss, R.; Weiss, E.; Elhamid, H.F.A. Environmental risk assessment focused on water quality in the Laborec River watershed. *Ecohydrol. Hydrobiol.* 2021, 21, 641–654. [CrossRef]
4. Bernauer, T. Explaining success and failure in international river management. *Aquat. Sci.* 2002, 64, 1–19. [CrossRef]
5. Rajosoa, A.S.; Abdelbaki, C.; Mourad, K.A. Water assessment in transboundary river basins: The case of the Medjerda River Basin. *Sustain. Water Resour. Manag.* 2021, 7, 1–13. [CrossRef]
6. Breaban, I.G.; Breaban, A.I. Causes and Effects of Water Pollution in Romania. In *Water Resources Management in Romania*; Springer Water: Cham, Switzerland, 2019; pp. 57–131.
7. Hein, T.; Funk, A.; Pletterbauer, F.; Graf, W.; Zsuffa, I.; Haidvogl, G.; Schinegger, R.; Weigelhofer, G. Management challenges related to long-term ecological impacts, complex stressor interactions, and different assessment approaches in the Danube River Basin. *River Res. Appl.* 2018, 35, 500–509. [CrossRef]
8. Kovalenko, S.; Bykovets, N.; Kolmykova, O. The “Lower Danube” Euroregion: Development Emergence of the Environmental Management Cluster Forms. *Balt. J. Econ. Stud.* 2021, 7, 140–149. [CrossRef]
9. Simionov, I.-A.; Cristea, D.S.; Petrea, S.-M.; Mogodan, A.; Jijie, R.; Ciornea, E.; Nicoara, M.; Rahoveanu, M.M.T.; Cristea, V. Predictive Innovative Methods for Aquatic Heavy Metals Pollution Based on Bioindicators in Support of Blue Economy in the Danube River Basin. *Sustainability* 2021, 13, 8936. [CrossRef]
10. Halder, J.; Vystavna, Y.; Wassenaar, L.I. Nitrate sources and mixing in the Danube watershed: Implications for transboundary river basin monitoring and management. *Sci. Rep.* 2022, 12, 1–9. [CrossRef]
11. European Commission (2000) Directive 2000/60/EC of the European Parliament and of the Council Establishing a Framework for Community Action in the Field of Water Policy. 2000. Available online: https://eur-lex.europa.eu/resource.html?uri=cellar:5c835afb-2ec6-4577-bdf8-756d3d694eeb.0004.02/DOC_1&format=PDF (accessed on 18 September 2022).
12. The Intergovernmental Panel on Climate Change (IPCC). Climate change 2013: The physical science basis. In *Contribution of Working Group I to the 5th Assessment Report of the Intergovernmental Panel on Climate Change*; Cambridge University Press: Cambridge, UK; New York, NY, USA, 2013.
13. Kubiak-Wójcicka, K. Variability of Air Temperature, Precipitation and Outflows in the Vistula Basin (Poland). *Resources* 2020, 9, 103. [CrossRef]
14. Whitehead, P.G.; Wilby, R.L.; Battarbee, R.W.; Kernan, M.; Wade, A.J. A review of the potential impacts of climate change on surface water quality. *Hydrol. Sci. J.* 2009, 54, 101–123. [CrossRef]
15. Kubiak-Wójcicka, K.; Zeleńajkova, M.; Blištan, P.; Simonová, D.; Pilarska, A. Influence of climate change on low flow conditions. case study: Laborec river, eastern Slovakia. *Ecol. Hydrolog. Hydrobiol.* 2021, 21, 570–583. [CrossRef]
16. Rajesh, M.; Rehana, S. Impact of climate change on river water temperature and dissolved oxygen: Indian riverine thermal regimes. *Sci. Rep.* 2022, 12, 1–12. [CrossRef] [PubMed]
17. Bänáduc, D.; Joy, M.; Olosutean, H.; Afanasyev, S.; Curtean-Bănăduc, A. Natural and anthropogenic driving forces as key elements in the Lower Danube Basin–South-Eastern Carpathians–North-Western Black Sea coast area lakes: A broken stepping stones for fish in a climatic change scenario? *Environ. Sci. Eur.* 2020, 32, 1–14. [CrossRef]
18. Siddha, S.; Sahu, P. Traditional and existing methods of urban water supply and their loopholes. In *Current Directions in Water Scarcity Research* 2022; Srivastav, A.L., Madhav, S., Bhardwaj, A.K., Eugenia Valsami-Jones, E., Eds.; Elsevier: Amsterdam, The Netherlands, 2022; Volume 6, pp. 245–271. [CrossRef]
19. McCracken, M.; Wolf, A.T. Updating the Register of International River Basins of the world. *Int. J. Water Resour. Dev.* 2018, 35, 732–782. [CrossRef]
20. Trábert, Z.; Duleba, M.; Bíró, T.; Dobosy, P.; Földi, A.; Hidas, A.; Kiss, K.T.; Óvári, M.; Takács, A.; Várberó, G.; et al. Effect of Land Use on the Benthic Diatom Community of the Danube River in the Region of Budapest. *Water* 2020, 12, 479. [CrossRef]
21. Salvai, A.; Grăbic, J.; Josimov-Dundjerski, J.; Zemunac, R.; Antonic, N.; Savic, R.; Blagojevic, B. Trend Analysis of Water Quality Parameters in the Middle Part of the Danube Flow in Serbia. *Ecol. Chem. Eng. S* 2022, 29, 51–63. [CrossRef]
22. Calmuc, M.; Calmuc, V.; Arseni, M.; Topa, C.; Timofti, M.; Georgescu, L.; Iticescu, C. A Comparative Approach to a Series of Physico-Chemical Quality Indices Used in Assessing Water Quality in the Lower Danube. *Water* **2020**, *12*, 3239. [CrossRef]

23. Crnković, D.; Sekulić, Z.; Antonović, D.; Marinković, A.; Popović, S.; Nikolić, J.; Drmanić, S. Origins of Polycyclic Aromatic Hydrocarbons in Sediments from the Danube and Sava Rivers and Their Tributaries in Serbia. *Pol. J. Environ. Stud.* **2020**, *29*, 2101–2110. [CrossRef]

24. Calmuc, V.A.; Calmuc, M.; Arseni, M.; Topa, C.M.; Timofti, M.; Burada, A.; Iticescu, C.; Georgescu, L.P. Assessment of Heavy Metal Pollution Levels in Sediments and of Ecological Risk by Quality Indices, Applying a Case Study: The Lower Danube River, Romania. *Water* **2021**, *13*, 1801. [CrossRef]

25. Chițescu, C.L.; Ene, A.; Geana, E.-I.; Vasile, A.M.; Ciucure, C.T. Emerging and Persistent Pollutants in the Aquatic Ecosystems of the Lower Danube Basin and North West Black Sea Region—A Review. *Appl. Sci.* **2021**, *11*, 9721. [CrossRef]

26. Daus, M.E.; Daus, Y.V. Estimating environmental risk assessment for drinking and fisheries use (on the example of the Danube river—the city Vilkovo). *J. Geol. Geogr. Geoecol.* **2021**, *30*, 23–33. [CrossRef]

27. Frîncu, R.-M. Long-Term Trends in Water Quality Indices in the Lower Danube and Tributaries in Romania (1996–2017). *Int. J. Environ. Res. Public Health* **2021**, *18*, 1665. [CrossRef]

28. Vosniakos, G.; Petre, J.; Pascu, L.; Vasil, G.; Iancu, V.; Staniloae, D.; Nicolau, M.; Cruceru, L.; Golumbeanu, M. Aquatic eco-system quality assessment of the Danube Delta in the periods April-October 2007 and 2008. *Frontiers Environ. Bull.* **2010**, *19*, 20–29.

29. Atanacković, A.; Sporka, F.; Marković, V.; Slobodnik, J.; Zorić, K.; Csányi, B.; Paunović, M. Aquatic Worm Assemblages along the Danube: A Homogenization Warning. *Water* **2020**, *12*, 2612. [CrossRef]

30. Literathy, P. PAH and Petroleum Hydrocarbon Contamination in Water, Suspended Particulate Matter, Sediments, and Biota in the Danube. *Handb. Environ. Chem.* **2015**, *39*, 217–233. [CrossRef]

31. Bělčáková-Minaříková, M.; Smedes, F.; Rusina, T.P.; Vraan, B. Application of equilibrium passive sampling to profile pore water and accessible concentrations of hydrophobic organic contaminants in Danube sediments. *Environ. Pollut.* **2020**, *267*, 115470. [CrossRef]

32. Kolarević, S.; Kračun-Kolarević, M.; Kostić, J.; Slobodnik, J.; Liška, I.; Gačić, Z.; Paunović, M.; Knežević-Vukučević, J.; Vukučević-Gačić, B. Assessment of the genotoxic potential along the Danube River by application of the comet assay on haemocytes of freshwater mussels: The Joint Danube Survey 3. *Sci. Total Environ.* **2016**, *540*, 377–385. [CrossRef] [PubMed]

33. Keller, V.; Williams, R.; Lofthouse, C.; Johnson, A. Worldwide estimation of river concentrations of any chemical originating from sewage-treatment plants using dilution factors. *Environ. Toxicol. Chem.* **2013**, *33*, 447–452. [CrossRef] [PubMed]

34. Novák, J.; Vraan, B.; Rusina, T.; Okonski, K.; Grabic, R.; Neale, P.A.; Escher, B.I.; Macová, M.; Ait-Aissa, S.; Creusot, N.; et al. Effect-based monitoring of the Danube River using mobile passive sampling. *Sci. Total Environ.* **2018**, *636*, 1608–1619. [CrossRef] [PubMed]

35. Brboric, M.; Vraan, B.; Radonic, J.; Vojinovic-Miloradov, M.; Turk-Sekulic, M. Spatial distribution of PAHs in riverbed sediments of the Danube River in Serbia: Anthropogenic and natural sources. *J. Serb. Chem. Soc.* **2019**, *84*, 1439–1453. [CrossRef]

36. Bloesch, J. The International Association for Danube Research (IAD)—portrait of a transboundary scientific NGO. *Environ. Sci. Policy* **2020**, *64*, 141–154. [CrossRef]

37. Kittinger, C.; Baumert, R.; Folli, B.; Lipp, M.; Liebmann, A.; Kirschner, A.; Farnleitner, A.H.; Grisold, A.J.; Zarfel, G.E. Preliminary Toxicological Evaluation of the River Danube Using in Vitro Bioassays. *Water* **2015**, *7*, 1959–1968. [CrossRef]

38. Chapman, D.V.; Bradley, C.; Gettel, G.M.; Hatvani, I.G.; Hein, T.; Kovács, J.; Liska, I.; Oliver, D.M.; Tanos, P.; Trázy, B.; et al. Developments in water quality monitoring and management in large river catchments using the Danube River as an example. *Environ. Sci. Policy* **2016**, *64*, 141–154. [CrossRef]

39. Habersack, H.; Hein, T.; Stanica, A.; Liska, I.; Mair, R.; Jäger, E.; Hauer, C.; Bradley, C. Challenges of river basin management: Current status of, and prospects for, the River Danube from a river engineering perspective. *Sci. Total Environ.* **2016**, *543*, 828–845. [CrossRef] [PubMed]

40. Tyler, A.; Hunter, P.D.; Spyrrakos, E.; Groom, S.; Constantinescu, A.M.; Kitchen, J. Developments in Earth observation for the assessment and monitoring of inland, transitional and shelf-sea waters. *Sci. Total Environ.* **2016**, *572*, 1307–1321. [CrossRef] [PubMed]

41. Kirschner, A.K.T.; Reischer, G.H.; Jakwerth, S.; Toth, E.; Sommer, R.; Mach, R.L.; Linke, R.; Eiler, A.; et al. Multiparametric monitoring of microbial faecal pollution reveals the dominance of human contamination along the whole Danube River. *Water Res.* **2017**, *124*, 543–555. [CrossRef] [PubMed]

42. Malagò, A.; Bouroufi, F.; Vigiak, O.; Grizzetti, B.; Pastori, M. Modelling water and nutrient fluxes in the Danube River Basin with SWAT. *Sci. Total Environ.* **2017**, *593*, 603–604, 196–218. [CrossRef]

43. Apetrei, C.; Iticescu, C.; Georgescu, L.P. Multisensory System Used for the Analysis of the Water in the Lower Area of River Danube. *Nanomaterials* **2019**, *9*, 891. [CrossRef] [PubMed]

44. Cozzi, S.; Ibáñez, C.; Lazar, L.; Raimbault, P.; Giani, M. Flow Regime and Nutrient-Loading Trends from the Largest South European Watersheds: Implications for the Productivity of Mediterranean and Black Sea’s Coastal Areas. *Water* **2018**, *11*, 1. [CrossRef]

45. Clarvis, M.H.; Fatichi, S.; Allan, A.; Fuhrer, J.; Stoffel, M.; Romerio, F.; Gaudard, L.; Burlando, P.; Beniston, M.; Xoplaki, E.; et al. Governing and managing water resources under changing hydro-climatic contexts: The case of the upper Rhone basin. *Environ. Sci. Policy* **2014**, *43*, 56–67. [CrossRef]
46. Mănoiu, V.-M.; Crăciun, A.-I. Danube river water quality trends: A qualitative review based on the open access web of science database. *Ecolhydrol. Hydrobiol.* 2021, 21, 613–628. [CrossRef]

47. International Commission for the Protection of the Danube River (ICPDR). 2018. ICPDR Climate Change Adaptation Strategy, Vienna: 84p. Available online: https://www.icpdr.org/main/activities-projects/climate-change-adaptation (accessed on 25 March 2022).

48. Atlas, R.M.; Bartha, R. Fate and effects of polluting petroleum in the marine environment. In *Residue Reviews*; Springer: New York, NY, USA, 1973; pp. 49–85.

49. Abdel-Shafy, H.I.; Mansour, M.S.M. A review on polycyclic aromatic hydrocarbons: Source, environmental impact, effect on human health and remediation. *Egypt. J. Pet.* 2016, 25, 107–123. [CrossRef]

50. Albers, P.H. Petroleum and Individual Polycyclic Aromatic Hydrocarbons. In *Handbook of Ecotoxicology*; Hoffman, D.J., Rattner, B.A., Burton, G.A., Cairns, J., Eds.; CRC Press: Boca Raton, FL, USA, 2003; pp. 341–371. [CrossRef]

51. Wernicke, T.; Abel, S.; Escher, B.I.; Koschorreck, J.; Rüdel, H.; Jahnke, A. Equilibrium sampling of suspended particulate matter as a universal proxy for fish and mussel monitoring. *Ecotoxicol. Environ. Saf.* 2022, 232, 113285. [CrossRef] [PubMed]

52. Ajiboye, O.O.; Yakubu, A.F.; Adams, T. A Review of Polycyclic Aromatic Hydrocarbons and Heavy Metal Contamination of Fish from Fish Farms. *J. Appl. Sci. Environ. Manag.* 2011, 15, 235–238. [CrossRef]

53. Visca, A.; Caracciolo, A.B.; Grenni, P.; Rolando, L.; Mariani, L.; Rauseo, J.; Spataro, F.; Monostory, K.; Sperlagh, B.; Patrolecco, L. Legacy and Emerging Pollutants in an Urban River Stretch and Effects on the Bacterioplankton Community. *Water* 2021, 13, 3402. [CrossRef]

54. Liška, I.; Wagner, F.; Sengl, M.; Deutsch, K.; Slobodnik, J. *Joint Danube Survey 3—A Comprehensive Analysis of Danube Water Quality. Final Report*; ICPDR—International Commission for the Protection of the Danube River: Vienna, Austria, 2015; 369p. Available online: http://www.danubesurvey.org/jds3/results (accessed on 5 August 2022).

55. International Commission for the Protection of the Danube River (ICPDR). JDS3 Results. JDS3 Overview Map (Sampling Sites of the JDS3). 2015. Available online: http://www.danubesurvey.org/jds3/jds3-files/nodes/documents/jds3_overview_map.pdf (accessed on 18 September 2022).

56. Pascu, L.F.; Cruceanu, L.; Vasilie, G.; Iancu, V.; Dinu, C.; Niculescu, M.; Nicolau, M. Investigation of physical and chemical quality of surface water and sediment in seven locations from Danube Delta, in the period 2009–2011. In *International Symposium “The Environment and the Industry” SIMI*; National Research and Development Institute for Industrial Ecology, INCD-ECOIND: Bucharest, Romania, 2011; Volume II, pp. 254–262. ISSN 2457-8371.

57. Stânescu, E.; Vosniakos, F.; Scraedanu, D.; Paun, I.; Lucaciu, I.; Stânescu, B.; Cruceanu, L.; Vasilie, G.; Iancu, V.; Niculescu, M. Assessment of the abiotic components of the Danube River and main tributaries from Southern part of Romania. *J. Environ. Prot. Ecol.* 2015, 16, 1371–3179.

58. Deutschmann, B.; Kolarevic, S.; Brack, W.; Kaisarevic, S.; Kostic, J.; Kracun-Kolarevic, M.; Liska, I.; Paunovic, M.; Seiler, T.-B.; Shao, Y.; et al. Longitudinal profile of the genotoxic potential of the River Danube on erythrocytes of wild common bleak (Alburnus alburnus) assessed using the comet and micronucleus assay. *Sci. Total Environ.* 2016, 573, 1441–1449. [CrossRef] [PubMed]

59. Pérez-Albaladejo, E.; Rizzi, J.; Fernandes, D.; Lille-Langøy, R.; Karlsen, O.A.; Goksøyr, A.; Oros, A.; Spagnoli, F.; Porte, C. Assessment of the environmental quality of coastal sediments by using a combination of in vitro bioassays. *Mar. Pollut. Bull.* 2016, 108, 53–61. [CrossRef]

60. Nicolae, F.; Ristea, M.; Cotorcea, A.; Perkovic, M. Modelling the naval transport associated hydrocarbon pollution risks in the Danube Delta biosphere reserve. *J. Environ. Prot. Ecol.* 2017, 18, 30–39.

61. Radu, M.; Ionescu, P.; Ivanov, A.A.; Deak, G.; Marcu, E.; Ciobotaru, I.-E.; Diacu, E.; Pipirigeanu, M. Assessment of Contamination with Hazardous Substances in Surface Sediments in the Lower Danube River. *Technium* 2019, 1, 37–44. [CrossRef]

62. Iordache, A.; Iordache, M.; Sandru, C.; Voica, C.; Stegarus, D.; Zgarovarega, R.; Ionete, R.E.; (Ticu), S.C.; Miriciu, M.G. A Fugacity Based Model for the Assessment of Pollutant Dynamic Evolution of VOCS and BTEX in the Olt River Basin (Romania). *Rev. Chim.* 2019, 70, 3456–3463. [CrossRef]

63. Cancrin, A.; Wehrl, B.; Körtzinger, A. Methane in the Danube Delta: The importance of spatial pattern and die cycles for atmospheric emission estimates. *Biogeoosciences* 2021, 18, 3961–3979. [CrossRef]

64. Order 161/2006 of the Romanian Ministry of the Environment and Water Management (Ordinul nr. 161/16.02.2006, Privind Clasificarea Calității apelor de Suprafața), Transposed from Water Framework Directive 2000/60/EC. Monitorul Oficial al României 2006, I/511. Available online: https://legislatie.just.ro/Public/FormaPrintabila/00000G3B3OTFI5P2O4U34BVCBDTBBRTT (accessed on 30 March 2022).

65. Adewuyi, Y.G. Sonochrome: Environmental Science and Engineering Applications. *Ind. Eng. Chem. Res.* 2001, 40, 4681–4715. [CrossRef]

66. Mahrt, R., Jr.; Chase, C.R. Oil Spill Detection Technology for Early Warning Spill Prevention. *IEEE OCEANS* 2009, 1–8. [CrossRef]

67. Olivella, M.A.; Jové, P.; Şen, A.; Pereira, H.; Villaescusa, I.; Fiol, N. Sorption performance of Quercus cerris cork with polycyclic aromatic hydrocarbons and toxicity testing. *BioResources* 2011, 6, 3363–3375.

68. Hao, L.; Huiying, D.; Jun, S. Activated carbon and cerium supported on activated carbon applied to the catalytic ozonation of polycyclic aromatic hydrocarbons. *J. Mol. Catal. A Chem.* 2012, 363–364, 101–107. [CrossRef]
69. Tričković, J.; Isakovski, M.K.; Watson, M.; Maletić, S.; Rončević, S.; Dalmacija, B.; Kónya, Z.; Kukovecz, A. Sorption Behaviour of Trichlorobenzenes and Polycyclic Aromatic Hydrocarbons in the Absence or Presence of Carbon Nanotubes in the Aquatic Environment. *Water Air Soil Pollut.* 2016, 227, 374. [CrossRef]

70. Li, S.; Luo, J.; Hang, X.; Zhao, S.; Wan, Y. Removal of polycyclic aromatic hydrocarbons by nanofiltration membranes: Rejection and fouling mechanisms. *J. Membr. Sci.* 2019, 582, 264–273. [CrossRef]

71. Maletić, S.P.; Beljin, J.M.; Rončević, S.D.; Grgić, M.G.; Dalmacija, B.D. State of the art and future challenges for polycyclic aromatic hydrocarbons is sediments: Sources, fate, bioavailability and remediation techniques. *J. Hazard. Mater.* 2018, 365, 467–482. [CrossRef]

72. Abdibattayeva, M.; Bissenov, K.; Zhubandykova, Z.; Orynbassar, R.; Tastanova, L.; Almatova, B. Purification of Oil-Containing Waste Using Solar Energy. *Environ. Clim. Technol.* 2021, 25, 161–175. [CrossRef]

73. Ghouas, H.; Haddou, B.; Canselier, J.P.; Gourdon, C. Wastewater Pollution Prevention for volatile organic compounds (Benzene, Toluene, Ethylbenzene, and Xylene) using cloud point extraction and regeneration of surfactant by evaporation. *Euro-Mediterr. J. Environ. Integr.* 2022, 7, 1–12. [CrossRef]

74. Ansar, B.S.K.; Kavusi, E.; Dehghanian, Z.; Pandey, J.; Lajayer, B.A.; Price, G.W.; Astatkie, T. Removal of organic and inorganic contaminants from the air, soil, and water by algae. *Environ. Sci. Pollut. Res.* 2022, 1–29. [CrossRef]

75. Kothiyal, N.C.; Abhay; Khan, L.; Kumar, V.; Saruchi; Singh, S. Recent advances in emission, analysis and remediation technique of carcinogenic polycyclic aromatic hydrocarbons: A review. *Glob. NEST J.* 2022, 24, 177–194.