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Priority Pollutants Effects on Aquatic Ecosystems Evaluated through Ecotoxicity, Impact, and Risk Assessments

Roxana Zait [1,2], Daniela Fighir [1], Brindusa Sluser [1,*], Oana Plavan [1] and Carmen Teodosiu [1,*]

1 Department of Environmental Engineering and Management, "Cristofor Simionescu" Faculty of Chemical Engineering and Environmental Protection, "Gheorghe Asachi" Technical University of Iasi, Bd. D. Mangeron 73, 700050 Iasi, Romania
2 Siret River Basin Water Administration, Str. N. Lascar Bogdan 15C, 600401 Bacau, Romania
* Correspondence: brobu@ch.tuiasi.ro (B.S.); cteo@ch.tuiasi.ro (C.T.)

Abstract: As water management is still a problem of international concern, scientists and practitioners are collaborating to develop new tools and methods to improve and help in the decision-making process. When addressing the priority pollutant monitoring and impact assessment, the ecotoxicity effects, carcinogenic and non-carcinogenic, should be considered together with the exposure factor and health hazards. The main goals of this study were to assess the ecological and health hazards and to apply integrated impact and risk assessment based on the ecotoxicity and exposure factors of each priority pollutant present in the aquatic ecosystem. This study used as a database the measured concentrations of 5 inorganic and 14 organic priority pollutants from the Siret river basin from NE Romania, from 18 river sections monitored in the period 2015–2020. The USEtox methodology and a new integrated index for environmental impact and risk assessment were developed and applied to evaluate the ecological and health hazards and environmental impacts and risks within the river basin. The total impact scores for heavy metals ranged from 2×10^3 to 2.25×10^9, and those for organic pollutants ranged from 2.72×10^{-1} to 2.95×10^6. The environmental risk in the case of inorganic priority pollutants ranged between 5.56 and 3136.35, and that in the case of organic pollutants was between 4.69 and 4059.17. The results revealed that there is a major to catastrophic environmental impact in almost all monitored river sections (10 out of 18), and the overall risk exposure was found to be at a significant to a major level. This study proved the harmful effects that the priority pollutants may have, even in very small concentrations, on non-target organisms and suggests that greater control over the pollution sources and mitigation of environmental impacts and risks should be applied.

Keywords: priority pollutants; river water quality; environmental impact; risk; and health hazards

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1. Introduction

One of the sustainable development goals (SDG6) is to "ensure availability and sustainable management of water and sanitation for all", with targets concerning water quality, water-use efficiency, and water resources management that should be achieved by 2030 [1,2]. The water quality targets can be achieved by reducing pollution from industrial, agricultural, and municipal sources and by minimizing the release of hazardous chemicals and the proportion of untreated wastewater while increasing wastewater recycling and safe reuse. The sustainable withdrawals and supply of freshwater and the integrated water resources management at all levels, including transboundary cooperation, are also envisaged [3]. Other parts of the SDG targets for 2030 are directly linked to the following objectives: (1) "to preserve and sustainably use the oceans, seas and marine resources for sustainable development" and (2) "to protect, restore and promote sustainable use of terrestrial ecosystems, sustainably manage forests, combat desertification, stop and reverse land degradation and biodiversity loss" [2]. In addition, access to water and education are human rights that should be ensured globally so that a good quality of life can be achieved [2,4].

In economic growth and industrial and agricultural development, good water quality and accessibility for all people should be considered, especially considering the effects of climate change [5]. Water abstraction and treatment (water supply) are the main aspects that may influence water quality for drinking and industrial use, especially considering the pollution of water resources due to anthropic activities [4]. Considering all these aspects, integrated and sustainable water resources management represents a global concern [6–8]. Due to urbanization and industrialization, surface water systems, especially rivers, are constantly threatened by the action of multiple sources of pollution, affecting aquatic biodiversity and compromising water safety and human health [9,10]. The surface water quality of a certain river catchment is usually evaluated by the regional water administrations, wastewater treatment plants, and environmental protection agencies [8], based on the monitoring of general water quality indicators and priority pollutants, as recommended by European regulations [7].

From this perspective, the Water Framework Directive (WFD) represents the main instrument at European Union (EU) level and the legal framework for decreasing surface water pollution [11,12]. The WFD focuses on river basin management, establishing a framework for protecting water through pollution prevention, aquatic ecosystem quality improvement, and sustainable use of water resources [13]. At the EU level, Directive 2013/39/EU amends Directives 2000/60/EC and 2008/105/EC and lists the priority substances in the field of water resources. At the national level, the Government Decision GD 570/2016 is the reference for the activities of monitoring and treatment of priority pollutants and their effects on surface water quality [14].

The presence of priority pollutants (organic or inorganic) in aquatic ecosystems is directly linked to industrial production processes, agriculture, and transport activities that do not meet environmental standards [15]. The discharge of effluents with a wide range of inorganic and organic compounds that belong to the priority (PP) and emerging pollutant (EP) classes such as pharmaceuticals and personal care products, pesticides, heavy metals, detergents, and flame retardants provide, even in very small concentrations, ecotoxicological and human health effects and bioaccumulation and degradation characteristics that may influence aquatic biota and the performance and costs of water and wastewater treatment plants [16–19]. Conventional water and wastewater treatment technologies are usually inefficient in removing priority and emerging pollutants, and therefore advanced processes such as membrane processes, advanced oxidation, adsorption on various sorbents, and combined processes should be considered [18,19].

Different acute and chronic health hazards are caused by these potentially toxic elements such as the priority organic and inorganic pollutants due to their bioaccumulation capacity, carcinogenicity, persistent nature, and toxicity [20,21]. The environmental pollution effects included chiral pollutants with serious long-term health effects [21,22]. The most important chiral pollutants are pesticides, poly-chloro-biphenyls, polyaromatic hydrocarbons, brominated flame retardants, drugs, and pharmaceuticals [22]. Thus, the toxicity due to the chirality works as a slow poison and must be considered in the analysis of chiral drugs and pharmaceuticals discharged into the environment [22,23]. The biological effects of such pollutants and the determination of harmful long-term health effects on the exposed living organisms are also key for an improved environmental risk assessment of chiral pollutants [23]. The accumulation of the priority pollutants, inorganic and organic, in the human body, even at low concentrations, can increase the risks of endocrine disruption; adverse reproductive effects; cancers; and diseases of the lungs, digestive tract, and skin, and it can affect the hematopoietic and immune systems and induce neurologic and reproductive toxicity [24]. Examples of such include Cd, Cr, Hg, and Pb, which may produce serious health effects [20,24,25]. Moreover, the international agenda for global sustainability has set as one of the main goals the reduction in chemical production, use, and discharges so that human health hazards are minimized as the United Nations organization requires [26–28]. To achieve this goal, new methodologies and guidelines to assess the negative effects, ecotoxicity, and exposure to chemicals need to be internationally updated [29–31]. In order to make successful decisions regarding the environmental negative effects on living organisms generated by the heavy metals and organic persistent chemicals, it is necessary to broadly apply the latest impact and risk assessment methods, integrated approaches, concepts, and models. The European Commission recommends, along with the health hazard assessment, risk assessment, and life cycle assessment (LCA), the application of the USEtox model. This model is endorsed by the Society for Environmental Toxicology and Chemistry (SETAC) under the United Nations Environmental Program (UNEP) for characterizing human and ecotoxicological impacts of chemicals and is suitable for performing ecosystem impact assessments [32].

On the other hand, the main purpose of an environmental impact assessment (EIA) is to contribute to the identification and evaluation of the significant environmental consequences of various economic and industrial activities [33–35]. Thus, due to its applicability, efficiency, and flexibility, the EIA became a tool for optimizing economic growth while preserving environmental quality and considering the principles of sustainable development [36,37]. The EIA is therefore a process in which the likely environmental impacts of new developments are quantified at an early stage [38]. In the case of EIA applied at the river basin level, the pressures on surface water quality, as well as the impacts on lakes or groundwater quality, should be identified and considered [39].

The purpose of this study was to assess the effects of priority pollutants on the aquatic ecosystems within the Siret river basin (northeastern part of Romania) by means of toxicity, impact, and risk assessments. The evaluated area was previously monitored using specific quality indicators by Zait et al. (2022) [8], and the water quality index was used to describe the water quality status. However, in the previous article, ecotoxicity, health hazards, impacts, and risks were not assessed, although these have an important role in the decisions concerning the selection of water treatment technology for drinking purposes.

This study focuses on the following objectives: (1) the identification of the type and number of pollution sources situated around the sampling/monitoring water stations within the river basin; (2) the identification of the land use around the surface water monitoring stations; (3) the ecotoxicity assessment and health hazard evaluation for 15 and 10 priority pollutants, respectively; (4) the integrated impact and risk assessment based on the ecotoxicity and exposure factor of each priority pollutant considered in our assessment, within the river basin (19 indicators). The monitoring data for the last 6 years, 2015 to 2020, were used for the following priority pollutants: 5 heavy metals (As, Cd, Hg, Ni, and Pb), 10 polyaromatic hydrocarbons (PAHs) (naphthalene, phenanthrene, anthracene, fluoranthene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(ghi)perylene, indeno(1,2,3-cd)pyrene), 3 pesticides (alpha-hexachlorocyclohexane (α-HCH), beta-hexachlorocyclohexane (β-HCH), gamma-hexachlorocyclohexane (γ-HCH)), and di(2-ethylhexyl)phthalate (DEHP).

To the best of our knowledge, this is the first assessment that determines the influence of the inorganic and organic priority pollutants on the aquatic ecosystem at the river basin level and the human health hazards, while also considering the improved methodology for the integrated quantification of environmental impacts and risks. Thus, the novelty of the current work consists in new or improved methods applied in an integrative manner to quantify the environmental impacts and risks, ecotoxicity, and human health hazards. The characterization factors of the inorganic and organic priority pollutants were considered for ecotoxicity effects and human health hazards, while the exposure factors for each monitored pollutant were used in case of environmental impact and risk assessment.

2. Materials and Methods

2.1. Study Area: Sampling Points, Land Use, and Pollution Sources

Zait et al., in a previous work [8], presented the monitoring data for inorganic and organic priority pollutants and assessed the surface water quality status in the Siret river basin using the water quality index (WAWQI) method [8]. The findings of the previous study were that water quality is mostly unsuitable for drinking water supplies, being influenced by the quality of the main tributaries, as well as by the effluent of wastewater treatment plants. Therefore, this study presents the land use within the Siret river basin, identifying the main sources of pollution and their type and distribution in relation to the impacts, risks, and health hazards likely to occur due to exposure to the priority pollutants. The land use within the Siret river basin and the sampling points (monitoring station locations) are presented in Figure 1 and Figure S1 in the Supplementary Materials. According to reports from the National Institute of Statistics, the Siret river basin is the largest river basin in Romania and covers 42,890 km^2, 58.29% being forests, 22.70% being crops, 6.17% being urban and industrial areas, and 0.67% being surface waters [13]. Therefore, there are various industrial activities that significantly contribute to surface water pollution, as well as agricultural activities and wastewater treatment plant effluents. The main industrial activities and types of pollution sources in the studied area are presented in Table S1 in the Supplementary Materials. It can be observed that there are different industrial companies developed within the river basin limits, such as chemical industry companies, food industry companies, paper industry companies, farms, zootechnical activities, and wastewater treatment plants. Most of these activities are under EU regulations in terms of integrated pollution prevention and control measures and water management [40].

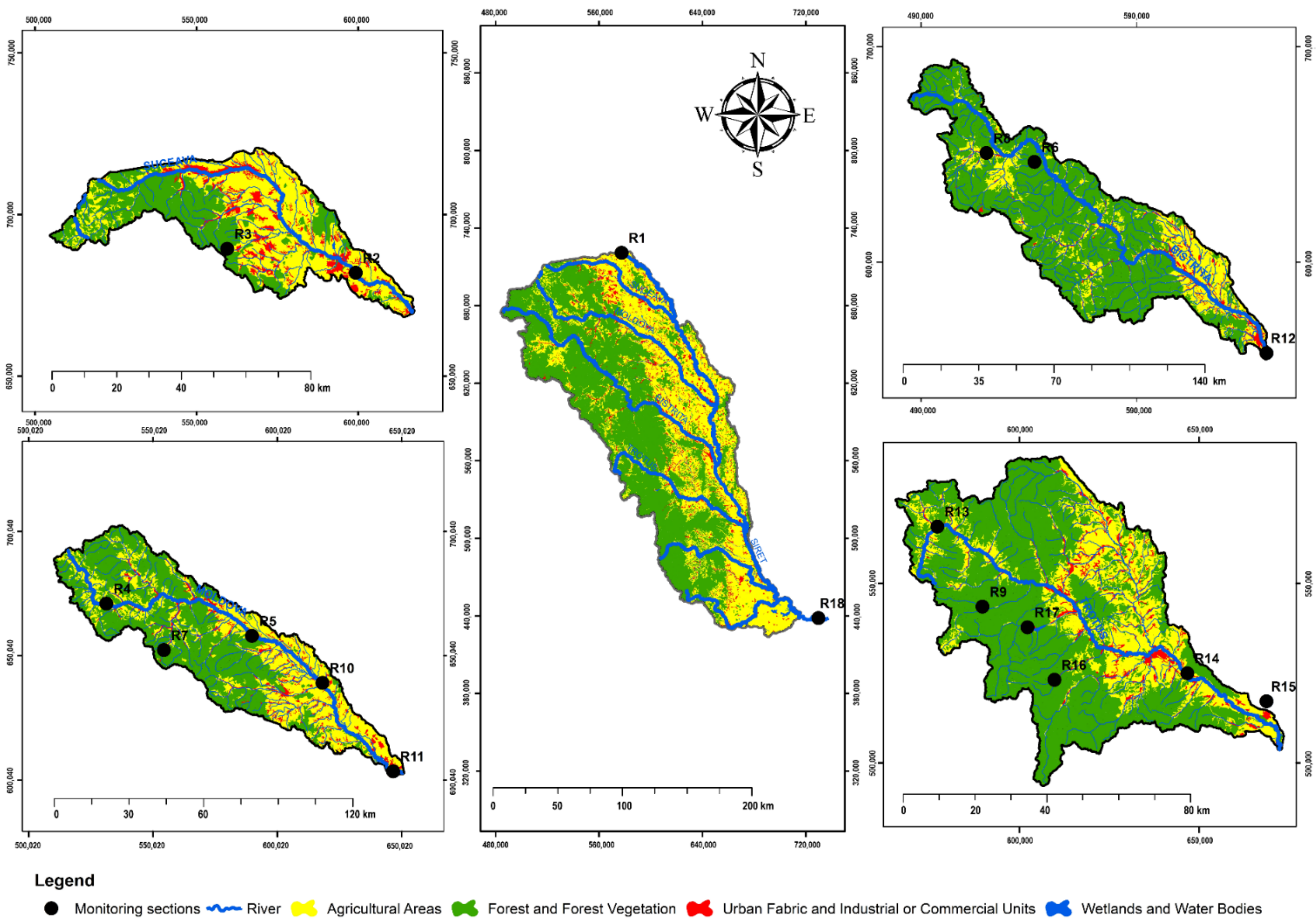


Figure 1. Siret river basin land use and water quality monitoring stations.

2.2. Experimental Section

The assessment of the impact and risk induced by priority pollutants (As, Cd, Hg, Ni, Pb, PAH, α-HCH, β-HCH, γ-HCH, and DEHP) on the surface water quality, within the Siret river basin, considered 18 sampling sections (monitoring stations) as presented in Figure 1. The sampling was run 2/4/8/12 times annually, during the period 2015–2020, and the integrated approach considered the annual average of the measured concentrations of the inorganic and organic priority pollutants. The variation in the river's annual average flow (multiannual values) was considered for the environmental impact assessment (Table 1). Simultaneously, the land use for 5 km around the sampling stations (78.5 km^2) was also considered, as presented in Table 2.

Table 1. The average values of the river's multiannual flow in the monitored sections.

River Section (See Figure 1)	Average Annual Flow, (m^3/s)/Year					
	2015	2016	2017	2018	2019	2020
R1	5.32	5.86	7.14	13.10	10.80	10.40
R2	8.27	12.30	11.2	24.00	19.60	14.70
R3	0.09	0.13	0.12	0.13	0.11	0.06
R4	2.27	3.36	2.48	4.59	3.85	2.96
R5	10.60	18.20	14.00	28.20	20.80	15.80
R6	0.52	0.48	0.76	1.03	0.88	0.37
R7	0.75	1.13	1.14	1.40	1.25	1.10
R8	5.46	7.82	6.46	8.25	7.34	6.02
R9	0.91	1.19	0.88	0.20	1.17	0.79
R10	15.00	22.20	19.50	42.90	31.90	22.30
R11	17.10	24.90	20.30	43.10	32.30	19.10
R12	3.85	4.38	3.30	6.24	5.15	1.89
R13	0.56	0.91	0.65	0.97	0.84	0.85
R14	20.60	42.00	33.10	15.60	27.40	16.40
R15	20.76	23.90	30.35	15.60	27.40	16.40
R16	0.93	1.45	1.22	1.26	0.94	0.88
R17	2.91	4.42	2.98	4.04	3.73	3.00
R18	176.00	181.00	129.00	226.00	207.00	168.00

Table 2. Land use (%) around the sampling station (5 km around).

Section	Priority Pollutants Measured	Urban Fabric and Industrial or Commercial Units (%)	Agricultural Areas (%)	Forest and Forest Vegetation (%)	Wetlands and Water Bodies (%)
R1	As, Cd, Hg, Ni, Pb, PAH, DEHP	11.32	86.24	0.23	2.21
R2	As, Cd, Hg, Ni, Pb, PAH, DEHP	20.55	75.60	1.57	2.28
R3	As, Cd, Hg, Ni, Pb, PAH, DEHP	6.42	16.54	77.04	-
R4	As, Cd, Hg, Ni, Pb, PAH	10.91	37.15	51.94	-
R5	As, Cd, Hg, Ni, Pb, PAH	6.47	52.49	36.46	4.58
R6	As, Cd, Hg, Ni, Pb, PAH	0.09	2.86	96.80	0.25
R7	As, Cd, Hg, Ni, Pb, PAH	5.20	6.42	88.38	-
R8	As, Cd, Hg, Ni, Pb, PAH	-	51.63	48.37	-
R9	As, Cd, Hg, Ni, Pb, PAH	-	1.60	98.40	-
R10	As, Cd, Hg, Ni, Pb, PAH	12.82	74.96	5.30	6.92
R11	As, Cd, Hg, Ni, Pb, PAH	24.86	68.23	2.95	3.96
R12	As, Cd, Hg, Ni, Pb, PAH, DEHP	16.75	57.82	7.95	17.48
R13	As, Cd, Hg, Ni, Pb, PAH, DEHP	11.08	34.46	54.46	-
R14	As, Cd, Hg, Ni, Pb, PAH, DEHP	9.63	45.28	42.45	2.64
R15	As, Cd, Hg, Ni, Pb, PAH, α-HCH, β-HCH, γ-HCH, DEHP	12.37	69.75	11.33	6.55
R16	As, Cd, Hg, Ni, Pb, PAH, α-HCH, β-HCH, γ-HCH, DEHP	1.22	0.47	98.31	-
R17	As, Cd, Hg, Ni, Pb, PAH	1.07	1.20	94.08	3.65
R18	As, Cd, Hg, Ni, Pb, PAH	22.48	59.45	12.56	5.51

2.3. Ecotoxicity and Health Hazard Assessment—USEtox Methodology

The health risk assessment (HRA) is widely used to quantify the risk of human exposure to certain pollutants. Generally, HRA offers important information that can contribute to the decision-making process by giving a quantitative estimation of risk [20].

The risk assessment can be divided into human health risk assessment and ecological risk assessment, depending on the protection focus. Various assessments of water quality have been developed by studies correlated to human health and risks posed by potentially toxic elements (PTEs) in aquatic environments [25].

The USEtox methodology has also been applied and described in numerous studies [32,41–45], but not for the case presented herein.

The methodology describes that the USEtox model is usually designed to supply the characterization factors (CFs) for human health impact [32], and it is associated with the emissions of a given chemical [44]. The impacts are calculated by multiplying the rate of the chemical released into the environment (i.e., surface water) by its toxicological characterization factors [46]. It can be expressed as presented in Equations (1) and (2) [44,47]:

$$CF = FF \cdot XF \cdot EF \tag{1}$$

where:

CF is a characterization factor for human health impact (number of cases/kg emitted);

FF, the fate factor, is the residence time, in days (d), of a chemical in a particular environment (kg·d/kg emitted);

XF, the exposure factor, is the "rate at which a pollutant is able to transfer from a receiving compartment into the human population through a series of exposure pathways" (kg intake/d·kg);

EF, the human effect factor, shows "the change in the lifetime disease probability, due to the change in lifetime intake of a pollutant" (number of cases/kg intake).

$$CF_{eco} = FF \cdot XF_{eco} \cdot EF_{eco} \tag{2}$$

where:

CF_{eco} is a characterization factor for aquatic ecotoxicity impacts ($PAF \cdot m^3 \cdot d/kg_{emmited}$);

FF, the fate factor, is exactly the same as for human health impact characterization (kg·d/kg emitted);

XF_{eco}, the freshwater ecosystem exposure factor, equals the fraction of a chemical dissolved in water and is given in the freshwater ecosystem exposure factor matrix (kg chemical dissolved/d·kg);

EF_{eco} is the ecotoxicological effect factor ($PAF \cdot m^3 \cdot d/kg$).

The CFs were calculated by using the USEtox 2.01 model, downloaded from the USEtox website [47]. This template is developed as a Microsoft Excel spreadsheet. In this version, USEtox covers three impact categories, namely human cancer toxicity, human non-cancer toxicity, and freshwater aquatic ecotoxicity. For each of these impact categories, USEtox follows the whole impact pathway from a chemical emission to the final impact on humans and ecosystems.

Specific information for the chemicals considered for this study was collected and stored in the "Substance data" sheet. Databases with specific chemical properties are available for organic and inorganic substances in Microsoft Excel format: "USEtox_substance _data_organics.xlsx" and "USEtox_substance_data_inorganics.xlsx" [48]. In this study, according to the USEtox database availability, 10 chemicals were characterized for human health hazard evaluation, namely As, Cd, Hg, Ni, Pb, naphthalene, anthracene, fluoranthene, γ-HCH, and DEHP; for ecotoxicity evaluation, 5 more priority pollutants were considered (phenanthrene, benzo(a)anthracene, benzo(a)pyrene, α-HCH, and β-HCH). Two USEtox databases were used, one for inorganic substances and one for organic substances. Through its matrix format, USEtox allows the identification of the main routes of exposure (e.g., inhalation, water ingestion, various food intakes), as well as the relative significance of potential carcinogenic and non-carcinogenic effects in the overall score [30]. CFs for human toxicity are estimated for carcinogenic and non-carcinogenic effects and consider emissions on different scales.

To make USEtox CFs compatible with the needs of LCA, the units for human toxicity are expressed as cumulative cases of either cancer or non-cancer health outcomes per kg of contaminant emission (No. of cases/$kg_{emmited}$), and the units for freshwater aquatic ecotoxicity impacts are expressed as the potentially affected fraction (PAF) of aquatic species integrated over the exposed water volume (m^3), time (day), and per kg emitted ($PAF \cdot m^3 \cdot d/kg_{emmited}$) [29–31].

The impact score (IS) was calculated by using a weighted summation of the releases of chemicals, considering the characterization factors ($CF_{x,i}$) (Equations (3) and (4)) [44]:

$$IS = \sum_{i} \sum_{x} CFx, i \cdot Mx, i \tag{3}$$

where:

IS is the impact score for, e.g., human toxicity (number of disease cases at midpoint level or disability-adjusted life years (DALY)) (number of the disease cases);

CFx,i is the characterization factor of substance x emitted to compartment i (number of disease cases/kg);

Mx,i is the emitted mass of substance x to compartment i (kg).

$$ISeco = \sum_{i} \sum_{x} CFecox, i \cdot Mx, i \tag{4}$$

where:

IS_{eco} is the impact score for ecotoxicity expressed at the midpoint level as potentially affected fraction (PAF) of freshwater species integrated over exposed volume and time ($PAF \cdot m^3 \cdot d$);

$CF_{eco}x,i$ is the characterization factor for the potential toxicity impacts of substance x released to compartment i for ecotoxicity impacts ($PAF \cdot m^3 \cdot d/kg$ emitted);

Mx,i is the emitted mass of substance x to compartment i (kg).

In the case of health hazard assessment for chemicals' presence in freshwater, the USEtox model calculates the characterization factors for carcinogenic and non-carcinogenic impacts, and the total impacts as carcinogenic and non-carcinogenic aggregated, assuming equal weighting. The unit of the characterization factor for freshwater ecosystem toxicity is the potentially affected fraction of species (PAF) at the midpoint level and the potentially disappeared fraction of species (PDF) at the endpoint level integrated over the freshwater volume (m^3) and the duration of 1 day (d) per kg emission, $PAF.m^3.d/kg$ (midpoint level) and $PDF \cdot m^3 \cdot d/kg$ (endpoint level) [30–32]. The unit of USEtox characterization factors for human toxicity are the number of disease cases at the midpoint level and the number of the DALY at the endpoint level per kg emission, cases/kg (midpoint level) and DALY/kg (endpoint level). Thereby, USEtox characterization factors are summarized as comparative toxic units (CTUs) at the midpoint level and comparative damage units (CDUs) at the endpoint level to stress the comparative nature of the characterization factors [30–32,45,47].

2.4. Integrated Impact and Risk Assessment

An effective approach strategy for addressing environmental concerns and identifying local, regional, and national priorities is to identify and assess the occurring environmental impacts and risks. The decision-making process integrates the risk assessment (RA) into the environmental impact assessment (EIA) due to both RA and EIA sharing the same final goal [33,35]. From the first stage of impact prediction and evaluation, through the implementation, to the post-closure stage, environmental impact and associated risk assessment (EIRA) supports a suitable decision-making process [35,49]. The integrated impact and risk assessment was previously applied in many studies for various situations such as mining activities, economic activities, chemical industry activities, and surface water resources [35,39,49–51].

The first step in the integrated approach is to assign importance on a scale from 0.1 to 1 (the most important), in terms of how valuable the environmental component is and how dangerous the measured pollutant is for the aquatic ecosystem. In this case, the assigned importance was considered the maximum, based on the priority pollutant toxicity. In addition, when quantifying the impact and risk generated by the priority pollutant presence within aquatic ecosystems, the alert threshold level according to the environmental standards was considered.

As compared to other impact assessment studies, the current study developed a new formula to integrate the impact and risk with the ecotoxicity factor, F_{eco}, which was calculated by using the USEtox 2.01 model, previously described, based on 19 priority pollutants (Table 3). The EIRA methodology considers environmental risks (ERs) as a function of the magnitude of environmental impacts (EIs) and their probability of occurrence and the factor of exposure to the priority pollutants as well (Equation (5)) [35]:

$$ER = EI \cdot p \cdot F_{eco} \tag{5}$$

where:

ER—environmental risk (dimensionless);

EI—environmental impact (dimensionless);

p—probability of impact occurrence (dimensionless);

F_{eco}—the freshwater ecosystem exposure factor for a substance in water, equal to the fraction of dissolved substance (Table 4) (dimensionless).

Table 3. Exposure factor (F_{eco}) values, according to USEtox [46,47].

Monitored Priority Pollutants	F_{eco}
As	0.89
Cd	0.66
Hg	0.19
Ni	0.71
Pb	0.01
Naphthalene	1.00
Phenanthrene	0.96
Anthracene	0.96
Fluoranthene	0.88
Benzo(a)anthracene	0.64
Benzo(b)fluoranthene	0.72
Benzo(k)fluoranthene	0.72
Benzo(a)pyrene	0.36
Benzo(ghi)perylene	0.32
Indeno(1,2,3-cd)pyrene	0.35
alpha-Hexachlorocyclohexane (α-HCH)	1.00
beta-Hexachlorocyclohexane (β-HCH)	1.00
gamma-Hexachlorocyclohexane (γ-HCH)	1.00
Di(2-ethylhexyl)phthalate (DEHP)	0.07

For USEtox, the determination of the freshwater ecotoxicological effect factor is based on the EC50 level (50% concentration on a vital feature of life history) [52].

F_{eco} values were calculated using the USEtox model for the inorganic and organic priority pollutants and are presented in Table 4.

The probability units were calculated with Equation (6). The probabilities of impact occurrences were calculated for each indicator as a frequency of discharge events that exceed the attention threshold of 70% of MAC [35,39]. The attention threshold is regularly imposed by national legislation [14] to create awareness of possible pollution events that might occur.

$$P = \frac{n}{m} \tag{6}$$

where:

n—number of attention thresholds (ATs) reached over the data series (number of "pollution events");

m—total number of measurements of the data series.

The global environmental impacts and associated risks caused by each economic activity were considered and were calculated using the following equation (Equation (7)) [35]:

$$I = \frac{\sum_{i=1}^{k}(EI_i)}{k} \tag{7}$$

where:

EI—total environmental impact on surface water,

EIi—environmental impact on surface water, considering quality indicator i;

i—quality indicator;

k—number of quality indicators considered in the evaluation process.

Table 4. The characterization factors for human toxicity and ecotoxicity effects.

Pollutants	Human Toxicity Effect Factors (Cases/kg_{intake}) Cancer	Human Toxicity Effect Factors (Cases/kg_{intake}) Non-Cancer	Ecotoxicity Effect Factors (PAF·m^3/kg)
As	543.48	38.83	211.80
Cd	10.00	6.21	30,209.63
Hg	69.44	43.49	6477.02
Ni	113.64	0.0254	2394.68
Pb	166.67	8.63	69,208.90
Naphthalene	n/a	0.0129	356.38
Phenanthrene	n/a	n/a	1647.39
Anthracene	n/a	0.0005	29,077.25
Fluoranthene	n/a	0.0036	1647.38
Benzo(a)anthracene	n/a	n/a	100,000
Benzo(a)pyrene	n/a	n/a	4309.39
α-HCH	n/a	n/a	370.43
β-HCH	n/a	n/a	596.25
γ-HCH (lindane)	n/a	0.77	6499.54
Di(2-ethylhexyl)-phthalate (DEHP)	n/a	0.04	249.63

Note: n/a–not applicable.

The quality of the assessed environmental component is quantified in this study as the ratio between the attention threshold of 70% of MAC (AT) of a certain quality indicator, according to the current legislation mentioned above, and the measured concentration (MC) at a certain moment (Equation (8)) [35]:

$$Q = \frac{AT}{MC} \tag{8}$$

The environmental impact is a function of two parameters, namely the magnitude, which depends directly on the concentration of the environmental pollutant, and the importance of the environmental component (Equation (9)) [35]:

$$EI = \frac{IU}{Qi} \tag{9}$$

where:

EI_i—environmental impact considering the quality indicator I (dimensionless);

IU—the importance units assigned to each environmental factor (dimensionless);

Q_i—quality of the environmental factor considering the quality indicator "i" (dimensionless).

Thus, the environmental impact (EI) is given by Equation (10) [35]:

$$EI = \frac{MC}{AT} \cdot IU \quad (10)$$

where:

EI_i—environmental impact considering the quality indicator I (dimensionless);
MC—measured concentration of monitored indicator (μg/L);
AT—alert threshold (μg/L);
IU—the importance units of each environmental factor (dimensionless).

3. Results and Discussion

3.1. Ecotoxicity and Health Assessment Results

3.1.1. Human Health Impact Score

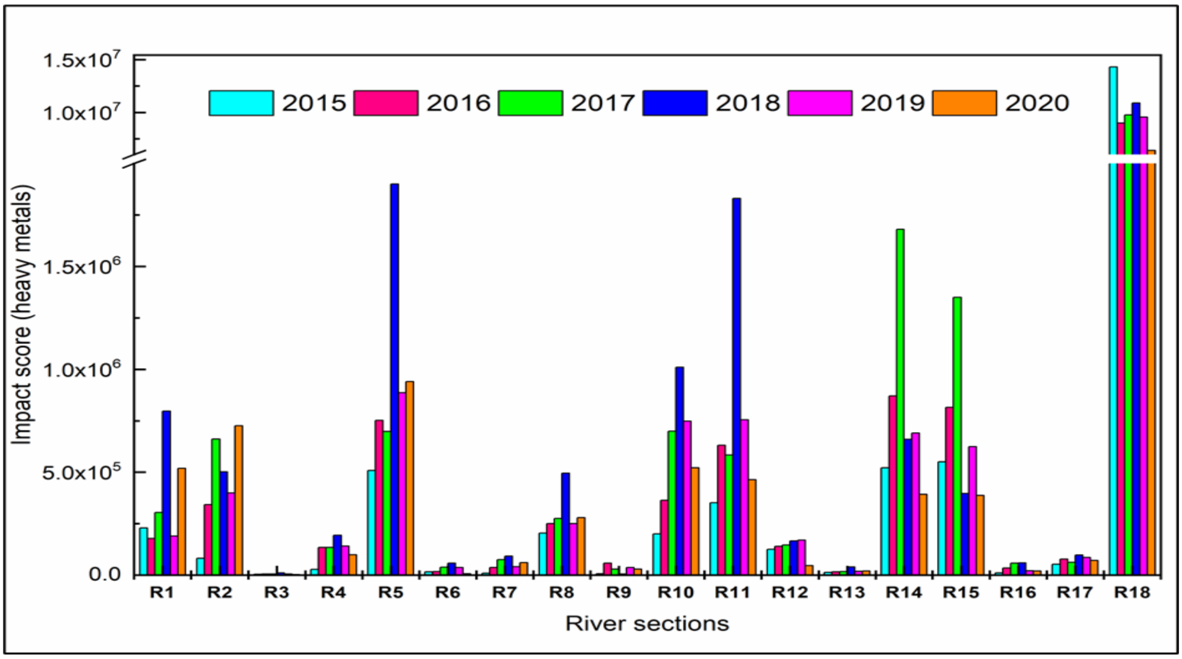
The pollutants considered in this study are persistent and toxic and may cause different acute and chronic health hazards, even at very low concentrations. They are classified as toxic and carcinogenic compounds and produce adverse health effects. The contamination of aquatic systems with these pollutants represents an important environmental problem of public health concern, and therefore it is crucial to assess their impact on human health and the environment. The characterization factors generated by USEtox for the selected pollutants (five heavy metals and five organic pollutants) are presented in Table 4. In this work, carcinogenic and non-carcinogenic health risks caused by oral ingestion were explored for drinking water because the river basin water serves as a water supply. The highest human toxicity effect factors were generated for As, Cd, and Pb, which led to high impact scores.

The results were structured into two parts considering carcinogenic and non-carcinogenic effects for inorganic and organic pollutants, respectively. The carcinogenic impact scores calculated for the human health damages from heavy metals are presented in Figure 2.

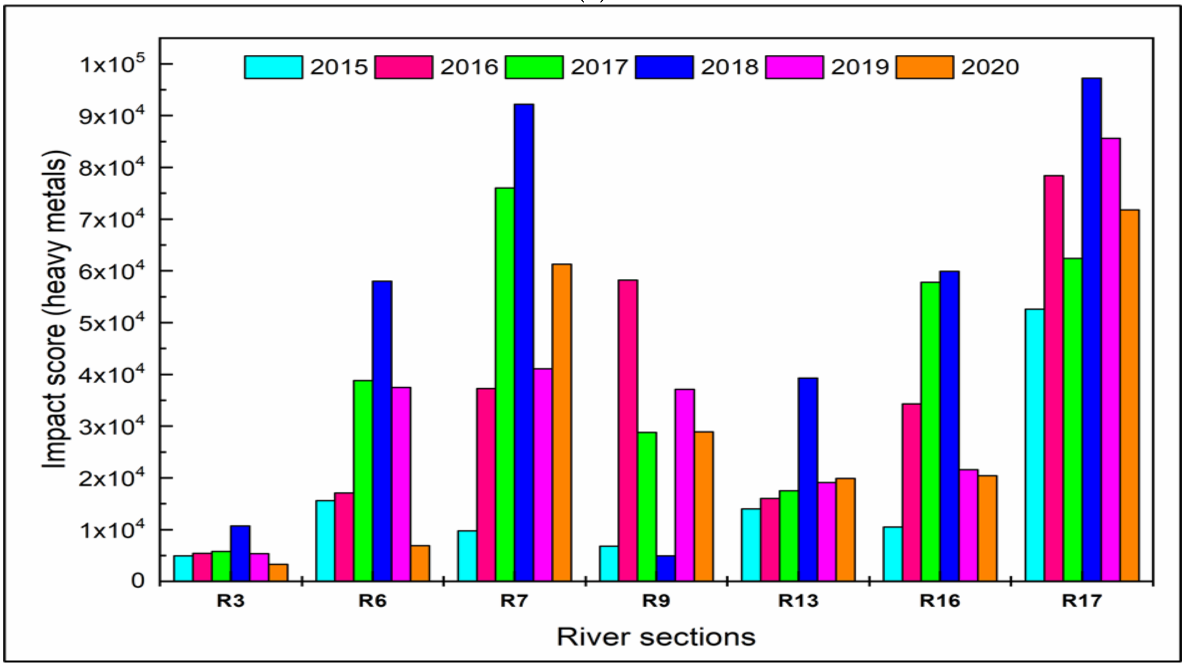
It can be observed that in the R18 river section, the impact score for the entire study period presents the highest values, followed by R11, R5, R14, and R15 for carcinogenic effects due to the presence of heavy metals. These values can be explained not only by the higher pollutant concentrations, but also by the average river flows, which can be correlated with the pollution sources (industrial activities) according to Table S1. The impact scores for carcinogenic effect ranged from 3.29×10^3 to 1.43×10^7. The obtained values demonstrated a significant carcinogenic health risk of the river water consumption and use, especially in the R18 river section. The highest impact scores were obtained in 2015 in R18, and the impact increased in the period 2015–2017, followed by a decrease until 2020. The same trend can be observed in Figure 3, and it should be mentioned that the impact scores were 100 times smaller in the case of non-carcinogenic effects.

In Figure 4, where the effects of organic pollutants are presented, it can be seen that the highest impact score was recorded in R15, followed by R11, R14, and R18. The values obtained ranged from 2.77×10^{-5} to 44.68, and the values were very small (10,000 times smaller) in comparison with the values obtained for heavy metals. It can be observed that the impact scores decreased in 2020, apart from those registered for the R1 river section. It is necessary to understand which pollutants determine the highest impact score, and therefore, a deep analysis was performed. Three heavy metals (As, Ni, and Pb) are responsible for these high impact scores, and the percentages for each pollutant are presented in Table 5.

As can be seen in Table 5, three heavy metals (As, Ni, and Pb) present evidence of carcinogenic and non-carcinogenic effects, and two heavy metals have no evidence of carcinogenic and non-carcinogenic effects (Cd and Hg). In a recent study, Fatoki and Badmus [53] have presented the toxicity effect of arsenic on the environment and human health. Arsenic occurs as both arsenite and arsenate in drinking water. A strong correlation has been established between arsenic ingestion and the pathogenesis of disorders such as cancer, neurological effects, organ toxicity, cardiovascular disease, diabetes, and immunotoxicity [53]. In the case of Pb, the main effect on the human body is impairment of the nervous system and blood disorders that can lead to encephalopathy and edema in the cerebellum, as well as cognitive behavioral disturbances [54]. Chronic exposure to Ni can produce allergy and dermatitis, cancer, and neuronal central system damage [55]. Table 6 presents the contribution to the total impact of the non-carcinogenic effects only. In this case, significant contributions can be observed for DEHP and γ-HCH.



(**a**)



(**b**)

Figure 2. Carcinogenic impact score for heavy metals: (**a**) R1 - R18 sections on the original scale; (**b**) a deeper view of selected sections (R3, R6, R7, R9, R13, R16, R17).

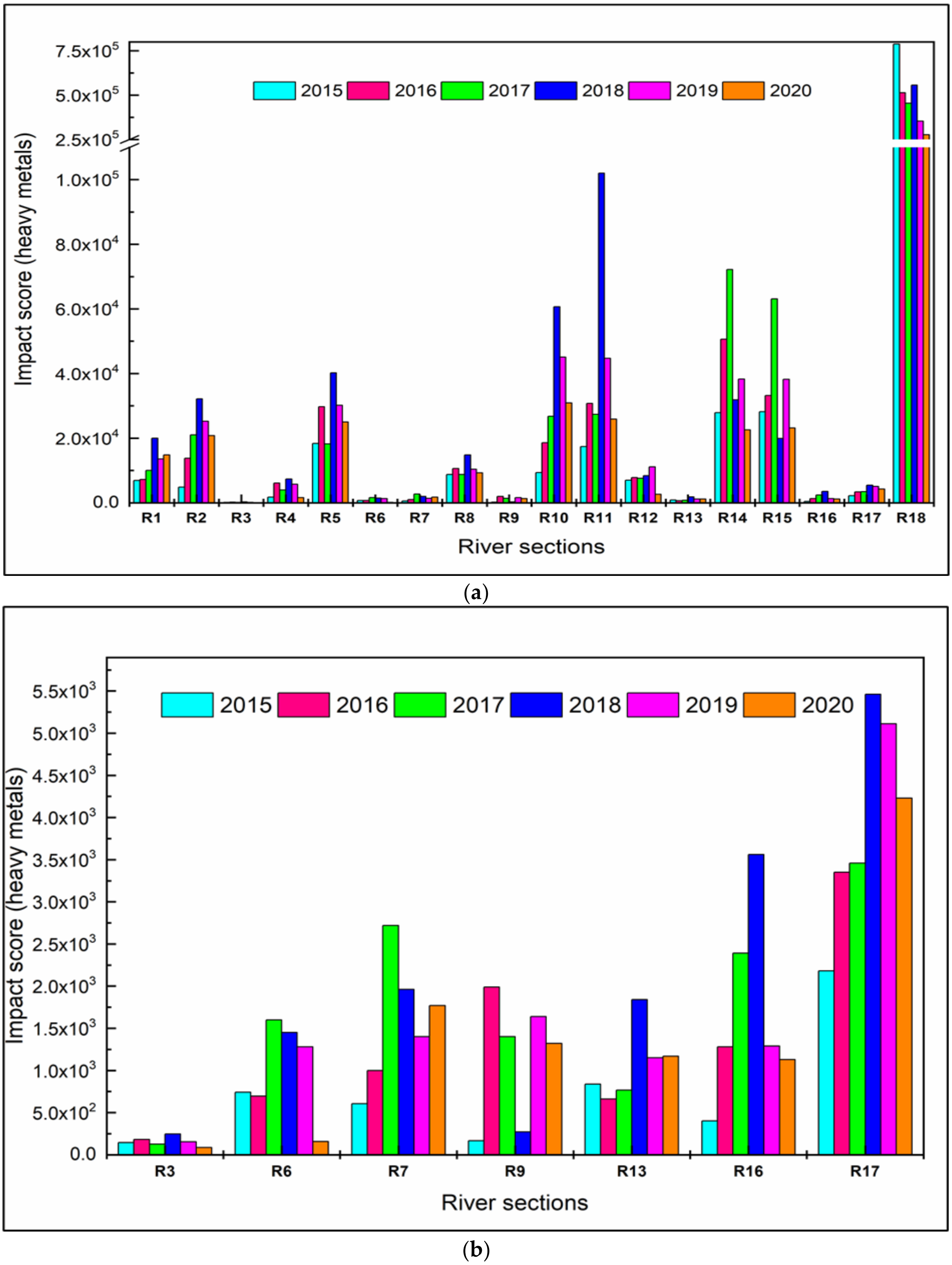
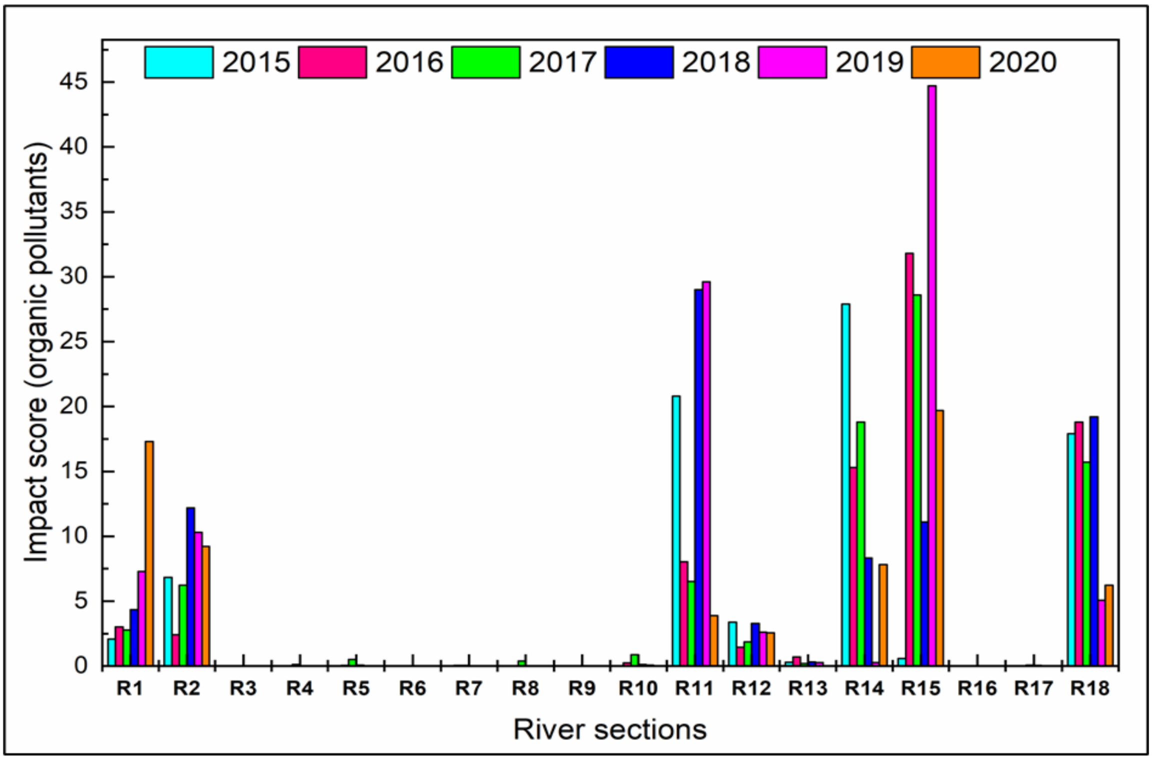


Figure 3. Non-carcinogenic impact score for heavy metals (2015–2020): (**a**) R1-R18 sections on the original scale; (**b**) a deeper view of selected sections (R3, R6, R7, R9, R13, R16, R17).



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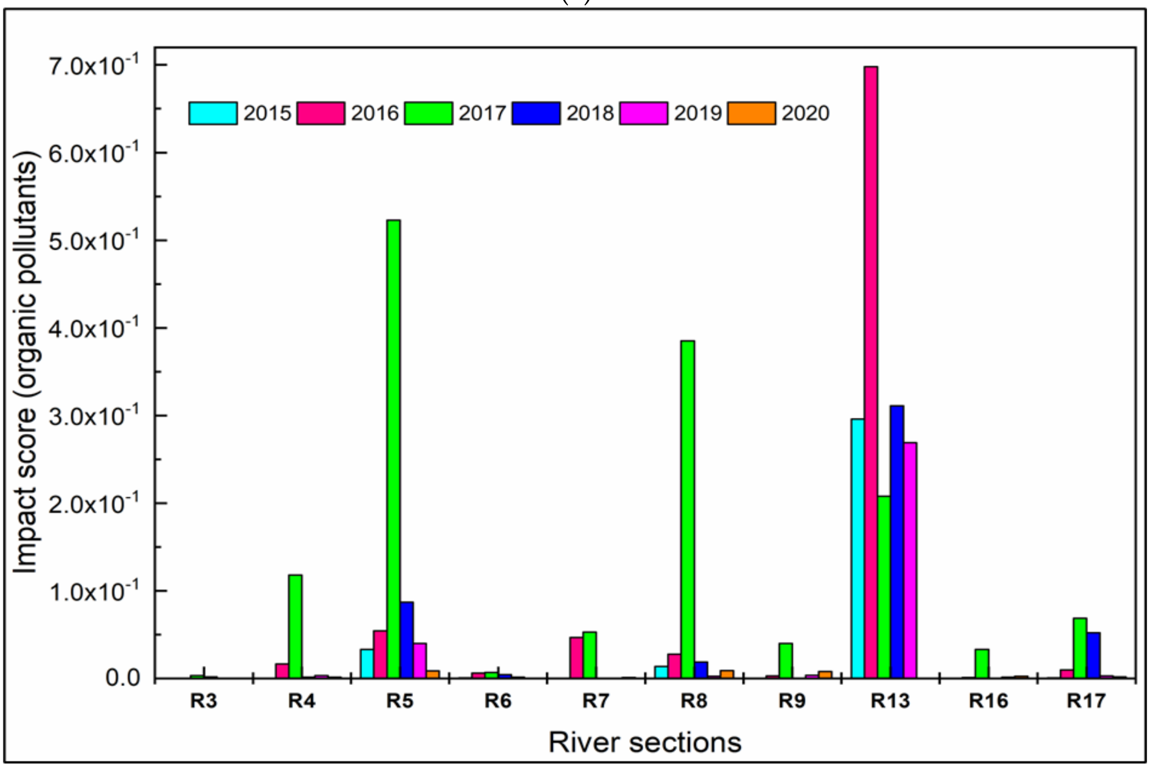


(**b**)

Figure 4. Non-carcinogenic impact score for organic pollutants (2015–2020): (**a**) R1-R18 sections on the original scale; (**b**) a deeper view of elected sections (R3, R6, R7, R9, R13, R16, R17).

Table 5. The contribution of heavy metals to the total impact scores.

	2015		2016		2017		2018		2019		2020		Total	
Pollutant	c *	n-c **	c *	n-c **	c *	n-c **	c *	n-c **	c *	n-c **	c *	n-c **	c *	n-c **
As	56.57	75.54	53.88	72.28	37.94	61.94	41.55	62.62	47.40	79.09	46.50	78.74	47.05	70.91
Cd	0.09	1.01	0.16	1.84	0.04	0.58	0.08	1.08	0.06	0.92	0.12	1.75	0.09	1.16
Hg	0.07	0.79	0.11	1.26	0.10	1.38	0.06	0.79	0.05	0.70	0.03	0.51	0.07	0.92
Ni	19.94	0.08	20.62	0.09	31.53	0.16	25.90	0.12	36.70	0.19	38.03	0.20	28.17	0.13
Pb	23.33	22.57	25.24	24.53	30.39	35.94	32.41	35.39	15.79	19.10	15.32	18.80	24.62	26.88

Note: * c—carcinogenic effect; ** n-c—non-carcinogenic effect.

Table 6. The contribution of organic pollutants to the total impact scores.

Pollutant	2015	2016	2017	2018	2019	2020	Total
Naphthalene	0.37	2.57	9.79	1.19	0.32	0.39	2.39
Fluoranthene	0.01	0.03	0.01	0.03	0.00	0.01	0.02
Anthracene	0.02	0.12	0.02	0.02	0.01	0.02	0.04
DEHP	69.56	64.66	47.15	66.21	79.03	70.26	66.70
γ-HCH	30.04	32.62	43.02	32.54	20.64	29.32	30.86

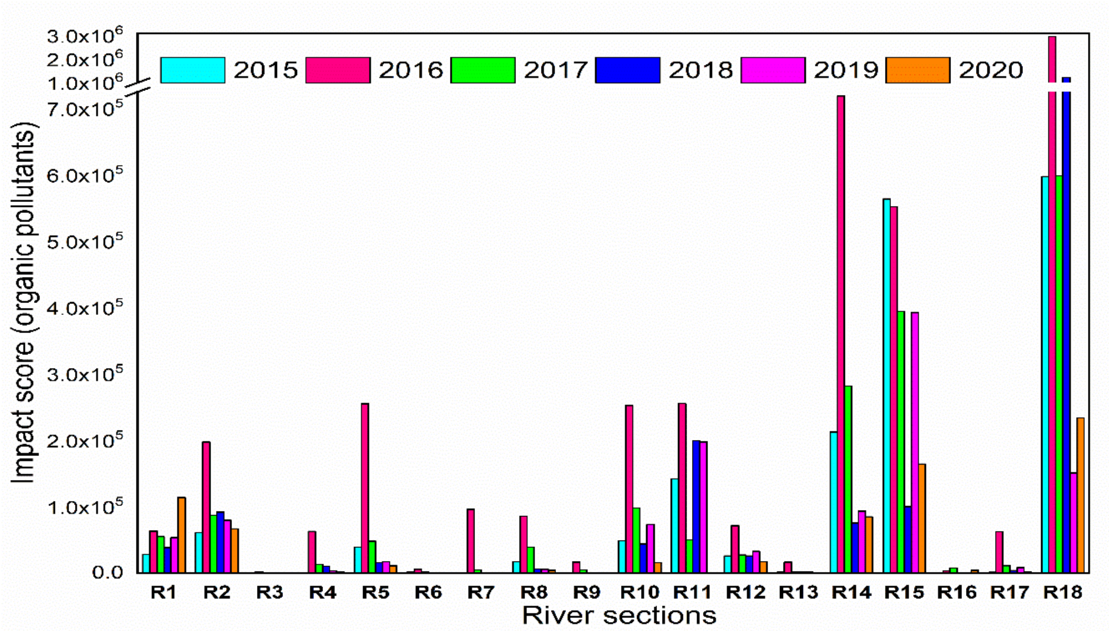
3.1.2. Ecotoxicity Impact Score

The ecotoxicity effect factors for freshwater ecosystems generated by USEtox are presented in Table 4, and the impact scores can be seen in Figure 5. The results are expressed as the total impact scores resulting from summing the impact scores obtained for heavy metals or organic pollutants in every single river section for each year. Similar to the previous results obtained for human toxicity, it can be observed (Figures 4 and 5) that the impact score presents the highest values in the R18 river section, followed by R14, R15, R5, R10, and R11. These high impact scores can be correlated with the pollution sources, especially with industrial activities. The obtained values demonstrate a significant ecotoxicity risk for river water consumption and use, especially in the R18 river section. After 2018, an improvement in water quality can be observed, especially in R18. In the case of R14 and R15, high impact scores were obtained in 2017, followed by a continuous decrease in the impact score.

In a deeper analysis (Tables 7 and 8), it was observed that in every year of the entire period of study, the highest contribution to the total impact score was given by lead (Pb), which also had the highest ecotoxicity effect factor (69,208.90 PAF·m^3/kg), causing detrimental effects to the aquatic organisms. The total impact scores for heavy metals ranged from 2×10^3 to 2.25×10^9, and those for organic pollutants ranged from 2.72×10^{-1} to 2.95×10^6. Ni and Cd are the next contributors to the total impact score with a contribution ranging between 0.91% and 4.73% for Cd and between 3.89% and 10.65% in the case of Ni. Overall, the ecological impact and health hazards are mostly due to the inorganic priority pollutants (heavy metals) as compared to the organic micropollutant results.

Table 7. Total impact scores obtained in the period 2015–2020 and the contribution of heavy metals.

Pollutant	**2015**		**2016**		**2017**		**2018**		**2019**		**2020**		**Total**	
	∑ ISeco	**%**	**∑ ISeco**	**%**	**∑ ISeco**	**%**	**∑ ISeco**	**%**	**∑ ISeco**	**%**	**∑ ISeco**	**%**	**∑ ISeco**	**%**
As	3.81×10^6	0.21	2.86×10^6	0.19	2.45×10^6	0.11	3.10×10^6	0.11	2.71×10^6	0.24	1.86×10^6	0.23	1.68×10^6	0.16
Cd	4.55×10^7	2.54	6.50×10^7	4.25	2.04×10^7	0.91	4.76×10^7	1.74	2.81×10^7	2.54	3.81×10^7	4.73	2.45×10^8	2.40
Hg	1.09×10^6	0.06	5.86×10^6	0.38	1.49×10^6	0.07	1.08×10^6	0.04	6.55×10^5	0.06	3.49×10^5	0.04	1.05×10^7	0.10
Ni	7.25×10^7	4.04	5.94×10^7	3.89	1.10×10^8	4.95	1.11×10^8	4.06	1.13×10^8	10.24	8.60×10^7	10.65	5.52×10^8	5.42
Pb	1.67×10^9	93.15	1.39×10^9	91.29	2.09×10^9	93.96	2.56×10^9	94.04	9.63×10^8	86.91	6.81×10^8	84.35	9.37×10^9	91.91
Total	1.79×10^9	100	1.53×10^9	100	2.23×10^9	100	2.73×10^9	100	1.11×10^9	100	8.07×10^8	100	1.02×10^{10}	100



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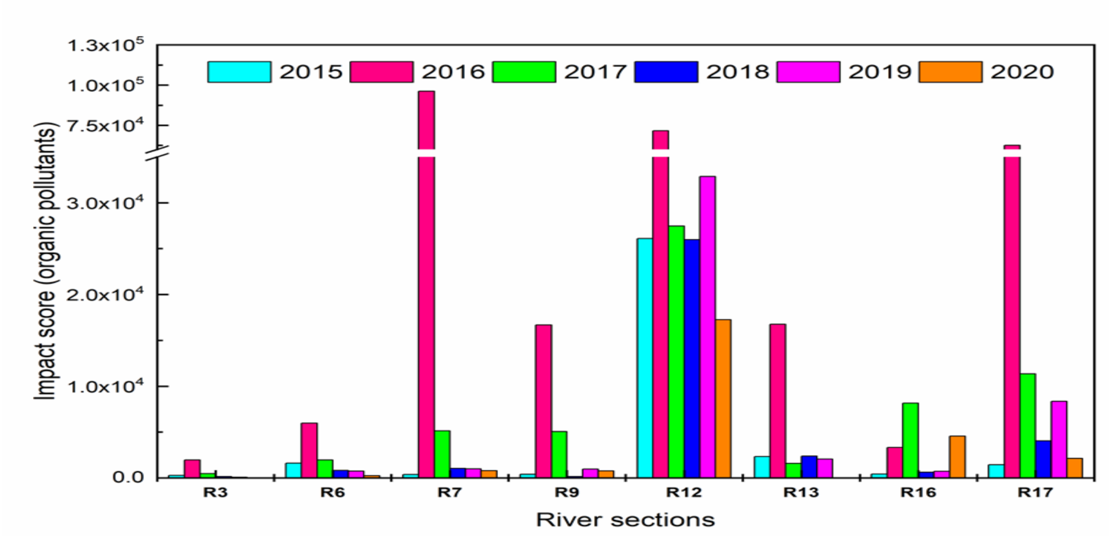


(**b**)

Figure 5. Ecotoxicity impact scores generated by organic pollutants (2015–2020): (**a**) R1-R18 sections on the original scale; (**b**) a deeper view of selected sections (R3, R6, R7, R9, R13, R16, R17).

Table 8. Total impact scores obtained in the period 2015–2020 and the contribution of organic pollutants.

Pollutant	2015		2016		2017		2018		2019		2020		Total	
	∑ ISeco	%	∑ ISeco	%	∑ ISeco	%	∑ ISeco	%	∑ ISeco	%	∑ ISeco	%	∑ ISeco	%
Naphthalene	8.15×10^3	0.47	5.91×10^4	1.04	2.24×10^5	13.00	3.77×10^4	2.06	9.68×10^3	0.87	7.21×10^3	1.00	3.46×10^5	2.70
Phenanthrene	8.52×10^4	4.88	2.07×10^5	3.65	2.53×10^5	14.67	2.51×10^5	13.67	6.27×10^3	0.56	3.97×10^4	5.48	8.42×10^5	6.57
Fluoranthene	8.30×10^4	4.75	2.24×10^5	3.96	1.08×10^5	6.25	3.16×10^5	17.20	4.00×10^4	3.59	3.55×10^4	4.90	8.06×10^5	6.29
Anthracene	1.18×10^5	6.78	8.98×10^5	15.84	1.27×10^5	7.36	1.87×10^5	10.18	1.40×10^5	12.60	1.35×10^5	18.57	1.61×10^6	12.53
Benzo(a)anthracene	3.40×10^5	19.47	3.49×10^6	61.57	4.18×10^5	24.22	3.88×10^5	21.14	1.45×10^5	13.01	2.66×10^4	3.67	4.81×10^6	37.52
Benzo(a)pyrene	1.50×10^4	0.86	2.25×10^5	3.97	1.81×10^4	1.05	1.67×10^4	0.91	1.19×10^4	1.07	1.93×10^4	2.67	3.06×10^5	2.39
DEHP	7.70×10^5	44.11	3.20×10^5	5.64	2.40×10^5	13.88	3.61×10^5	19.70	5.41×10^5	48.52	2.64×10^5	36.48	2.50×10^6	19.48
α-HCH	1.85×10^4	1.06	7.60×10^3	0.13	2.06×10^4	1.19	2.93×10^4	1.60	1.67×10^4	1.50	1.92×10^4	2.66	1.12×10^5	0.87
β-HCH	1.61×10^4	0.92	1.23×10^4	0.22	1.68×10^4	0.97	1.12×10^4	0.61	1.16×10^4	1.04	1.28×10^4	1.76	8.07×10^4	0.63
γ-HCH	2.92×10^5	16.71	2.26×10^5	3.98	3.01×10^5	17.41	2.37×10^5	12.93	1.92×10^5	17.25	1.65×10^5	22.81	1.41×10^6	11.02
Total	1.75×10^6	100	5.67×10^6	100	1.73×10^6	13.00	1.83×10^6	100	1.11×10^6	100	7.24×10^5	100	1.28×10^7	100

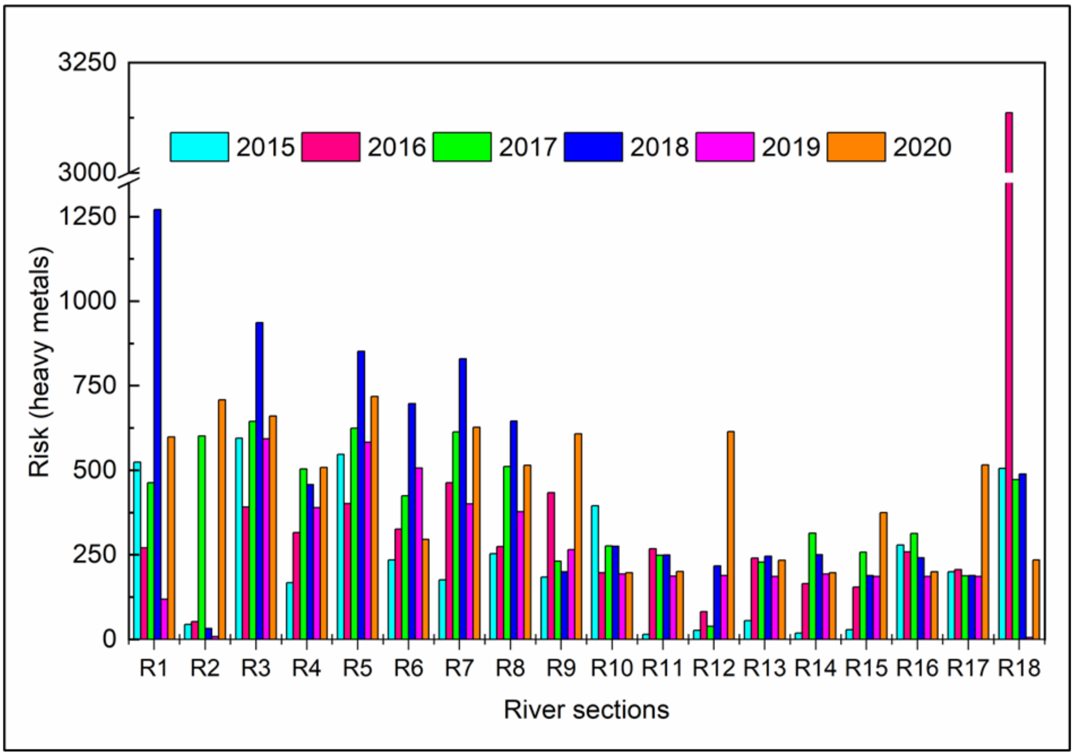
3.2. Integrated Impact and Risk Assessment Results

The integrated approach considered the quantification of risks as a direct correlation with the level of impact on environmental quality, in this case, surface water quality (river sections); the probability of impact occurrence; and the exposure factor for each priority pollutant considered in this study. Thus, the results reflect the impact on surface water quality based on the measured concentrations for the whole period, in relation to the alert threshold as stated by environmental standards.

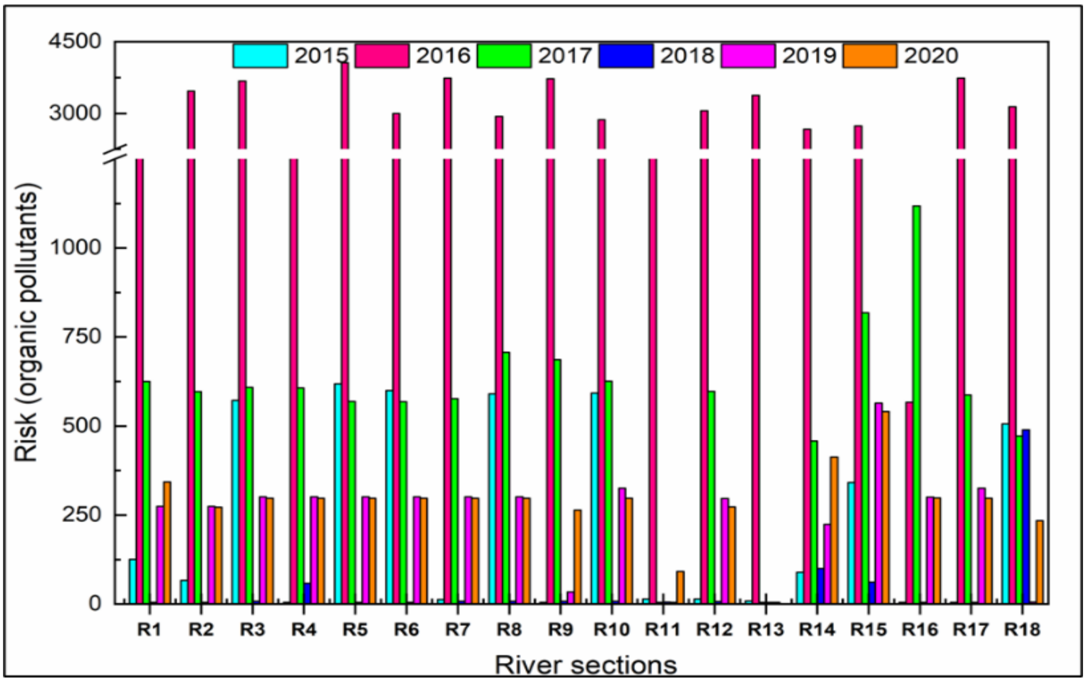
It can be observed (Figure 6) that river section 1, located on the border with Ukraine, is at major risk, while river section 18 (R18), situated in the southeastern part of the river basin, at the entrance of the Danube river, is less polluted but still at high risk. The impact and risk are mainly caused by the presence of heavy metals in the aquatic ecosystems in comparison to the monitored organic priority pollutants. In the case of heavy metals, major or catastrophic impacts and risks were recorded, while in the case of organic pollutants, there is a constant level of pollution with significant or major impacts and risks. In most cases, Cd, Ni, As, Hg, benzo(a)anthracene, and benzo(a)pyrene are the main contributors to major and catastrophic environmental impacts and risks at the river basin scale. It is worth mentioning that, for the last three monitored years (2018, 2019, and 2020), the impact trend is descending for both inorganic and organic pollutants. The most dangerous situation was recorded in 2016, when the highest precipitation volume and floods were reported and pollutant transportation and migration occurred.

3.3. Critical Analysis

River basins are the main surface water sources for drinking and industrial usage. Thus, achieving the sustainable development goals regarding water quality and management by 2030 is still a challenge. It is recommended to have more applications of wastewater recycling and reuse in the future, and therefore the chemicals present in the wastewater systems prompt scientists to develop solutions for the prevention and mitigation of acute and chronic health effects [56,57]. On the other hand, this study proved that human exposure is directly correlated to the quality of the water supply systems, and potential synergism due to co-exposure to other environmental contaminants must also be considered and integrated into new models and tools that can predict future negative effects [56]. For instance, heavy metal pollution of agricultural soils is of public concern due to the high potential toxicity and mobility, and mining activities contribute to pollutant accumulation [57,58]. However, the priority pollutants were shown to influence the water quality, harm the ecosystems, and pose risks to human health. This study analyzed the effects of inorganic and organic priority pollutants present in aquatic ecosystems and proved that there are major to catastrophic impacts on water quality and ecosystems (Figure 7a) in all monitoring stations for all 18 river sections, and yet the situation is at a likely significant or significant level in case of risk exposure (Figure 7b). Of great concern is the northern part of the Siret river basin (R3, R5, R7), where major risk values were recorded due to the presence of Cd and Ni in high concentrations and correlated to their toxicity and exposure factor. It can also be observed that within this area, around the R5 monitoring station, 29 industrial pollution sources are concentrated, while R3 and R7 are directly exposed to the agricultural sources. Therefore, the agricultural activities intensively developed in this area were shown to negatively influence the surface water quality. Moreover, R3 is proposed for drinking water abstraction and treatment. Thus, the results warn of possible harmful effects that chemicals may have on aquatic ecosystems and suggest a thorough analysis of the pollution source type, environmental impacts, and risk mitigation. It would be of interest for future work to study how these pollutants are accumulated in vegetables and microfauna and investigate the mitigation from the pollution source through the trophic chain.

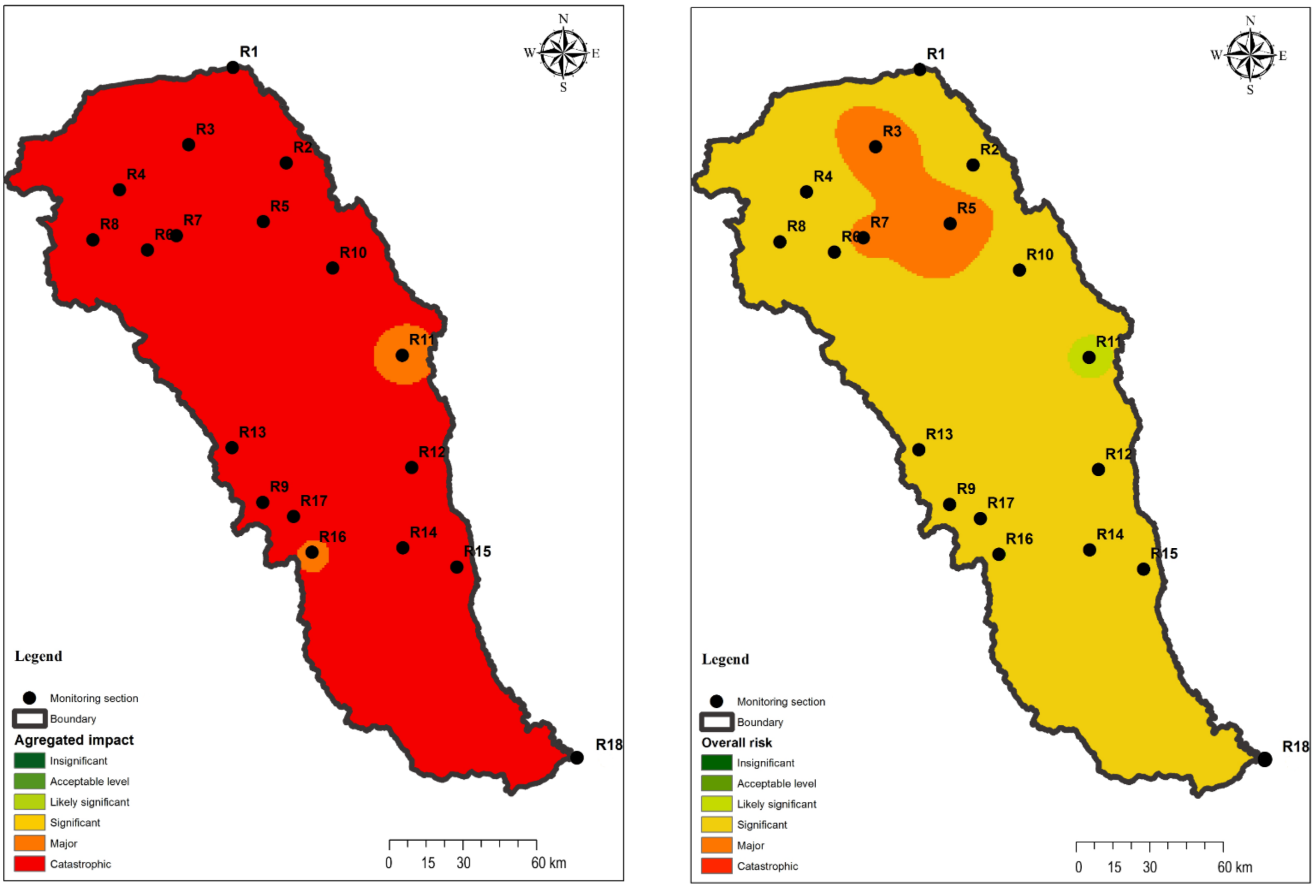


(a)



(b)

Figure 6. Environmental risks, period 2015–2022: (**a**) risk of heavy metals; (**b**) risk of organic pollutants.



(**a**) (**b**)

Figure 7. (**a**) Global environmental impact distribution; (**b**) global risk exposure distribution.

4. Conclusions

As water use is still a great concern at the international level, the main purpose of this work was to investigate whether the presence of inorganic and organic priority pollutants in aquatic ecosystems can harm the human health and aquatic environment of the NE region of Romania, in the Siret river basin. This study used the monitoring data for the last 6 years (2015 to 2020) from the Siret river basin. The USEtox methodology was applied together with the improved integrated method to assess the health hazards and ecological effects of inorganic and organic priority pollutants, carcinogenic and non-carcinogenic. The risk of exposure to the priority pollutants from the water systems was quantified as a direct function of the measured concentration of a certain priority pollutant, its alert threshold, and its exposure factor. The results proved that even at river basin scale there is a major to catastrophic impact on water quality, and the overall risk exposure is mainly at likely significant risk level. The main contributors to this severe situation are Cd, Ni, As, Hg, benzo(a)anthracene, and benzo(a)pyrene, especially considering the fact that the river section (R3) is used for drinking water supply. Therefore, continuous monitoring of the industrial and agricultural pollution sources is recommended, together with better decisions that should be made regarding the water management in this area. The evaluation methods developed for this study can be replicated to assess the ecological and health hazards at river basin level by environmental protection agencies or water administration authorities. Future work should be focused on the mitigation and bioaccumulation of these pollutants at the regional level.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w14203237/s1, Figure S1: Location of the pollution sources and water quality monitoring stations; Table S1: Industrial pollution source types within Siret river basin; Table S2: Annual average of measured concentrations for the inorganic and organic priority pollutants, in the monitored river sections (R1–R18), for the period 2015–2020.

Author Contributions: Conceptualization, B.S., D.F. and C.T.; methodology, R.Z, D.F. and B.S.; software, R.Z. and D.F.; validation, D.F., B.S. and C.T.; formal analysis, R.Z. and D.F.; investigation, R.Z. and D.F.; resources, R.Z. and O.P.; data curation, D.F.; writing—original draft preparation, R.Z, O.P. and D.F.; writing—review and editing, B.S. and C.T.; visualization, B.S. and D.F.; supervision, B.S. and C.T.; project administration, C.T. All authors have read and agreed to the published version of the manuscript.

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