

BLACK CARBON CONTENT AND DISTRIBUTION IN SURFACE SEDIMENTS FROM TEMPERATE-ZONE RESERVOIRS (POLAND)

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Abstract. This paper presents the results of first studies concerning the content of black carbon (BC) (which may be played a significant role in the accumulation of dangerous pollutants) in sediments of three reservoirs located in south-eastern Poland. The BC studies made use of the chemical-thermal oxidation method (CTO-375).

Key words: black carbon, total organic carbon, sediment, reservoir.

1. Introduction

One of the organic matter (OM) fractions is the so-called inert organic matter. Under criteria adopted, this is taken to include chemically and biologically inactive substances, including black carbon (BC). The BC fraction includes, for example, kerogen, charcoal, charred parts of plants, soot, graphite and ash [34]. Substances included in the composition of BC arise in the process of pyrolysis and/or incomplete combustion of biomass and fossil fuels [10] and may originate in both natural and anthropogenic sources. They are practically insoluble in water, have a high content of carbon in the molecules, rigid high-aromatic structures, a small number of polar substituents and a complicated spatial structure [16]. In terms of its molecular structure, BC differs significantly from other OM fractions, because it contains granular, crystalline organic components with a porous nanoparticulate structure [6].

BC is ubiquitous in the atmosphere, ice, soil and sediments, due to the widely dispersed nature of its production, as well as its stability and indifference in the environment, where it may play an important role in certain key processes. One of the main problems

associated with BC is its ability to absorb light energy and reduce the albedo of the surface of water, ice and snow, thereby accelerating the melting of glaciers [14]. BC is the main component of the atmospheric aerosol which absorbs light and affects the Earth's thermal radiation balance, in the sense that it may contribute to global warming [9]. Measurement of BC isotope composition proves a useful marker when it comes to the history of forest fires and biomass in a given area [5]. BC formed during combustion affects the global carbon cycle, because its residence time in the environment can be up to several million years [28]. Thus, BC can represent a significant sink of carbon, because it is a carbon transferred out of rapid biological-atmospheric cycling into long-term geological cycling [29].

BC is also considered an important carrier of certain kinds of pollution. Substances belonging to the BC fraction are characterised by complex spatial structure, large surface area and porosity, all of which allows for substantial sorption on to the surface, as well as penetration into the interior of the complex spatial structures of various dangerous contaminants, such as PAHs, PCBs and heavy metals [31, 3, 6, 7, 25, 21, 26].

The biological activity of the BC fraction is also a subject of study. People and animals in the immediate neighbourhood of local sources are exposed to BC aerosol by way of inhalation. The World Health Organisation (WHO) estimates that smoke from the combustion of solid fuels is one of the ten most important health-risk factors in the world, contributing to around 2 million deaths per year. When aerosol BC is inhaled, the particles migrate to the lungs. Because BC is a strong absorbent of toxic compounds, e.g. PAHs,

inhalation of BC leads to respiratory problems, and has been linked with lung cancer.

The work detailed here sought to analyse the spatial diversity of black carbon content (BC) in the surface layer of sediments in three reservoirs in south-eastern Poland. The relevance of this work lies in the way that this fraction of organic matter can affect the migration and fates of dangerous pollutants accumulated in the sediments.

2. Materials and methods

Study area

Solina, Rzeszów and Besko Reservoirs – all located in Poland’s Podkarpackie province-region – were selected for the research. Notwithstanding their similar geographical locations, the objects in question differ in terms of size, age, nature of catchment and trophic state. Characteristic parameters of the reservoirs are as presented in Fig. 1.

Solina Reservoir has a mountainous or hill catchment in which over 75 % is occupied by forest. Some of Poland’s most valuable natural areas are in its vicinity, and the entire area enjoys some form of protective status from the nature conservation point of view. Bieszczady National Park and Landscape Parks

are among the protected areas located here. The natural conditions and considerable morphological diversity of the area both affect the way land in the catchment is used, while also limiting agricultural activity. Holiday and tourist centres and village settlements with forest and agricultural tillage are located here, as are factories involved in light industry, and premises involved in the servicing of the permanent and seasonal populations making use of holiday and tourism facilities. The current state of wastewater management in the region has a major impact on the quality of tributary waters in the catchment of Solina Reservoir, which serves multi-purpose water management functions, though is used primarily to generate power and to provide flood control. The Reservoir also provides drinking water for surrounding towns and villages, including the 10,000-inhabitant Ustrzyki Dolne. Yet further functions of the Reservoir are in tourism and recreation.

In contrast, the catchment of Rzeszów Reservoir is afforested to only a limited extent, while 68 % of its entire area is agricultural. There are built-up areas in this Reservoir’s immediate vicinity, though the water again has flood-control and recreational functions, while also serving to increase the water level for the water intake of the city of Rzeszów.

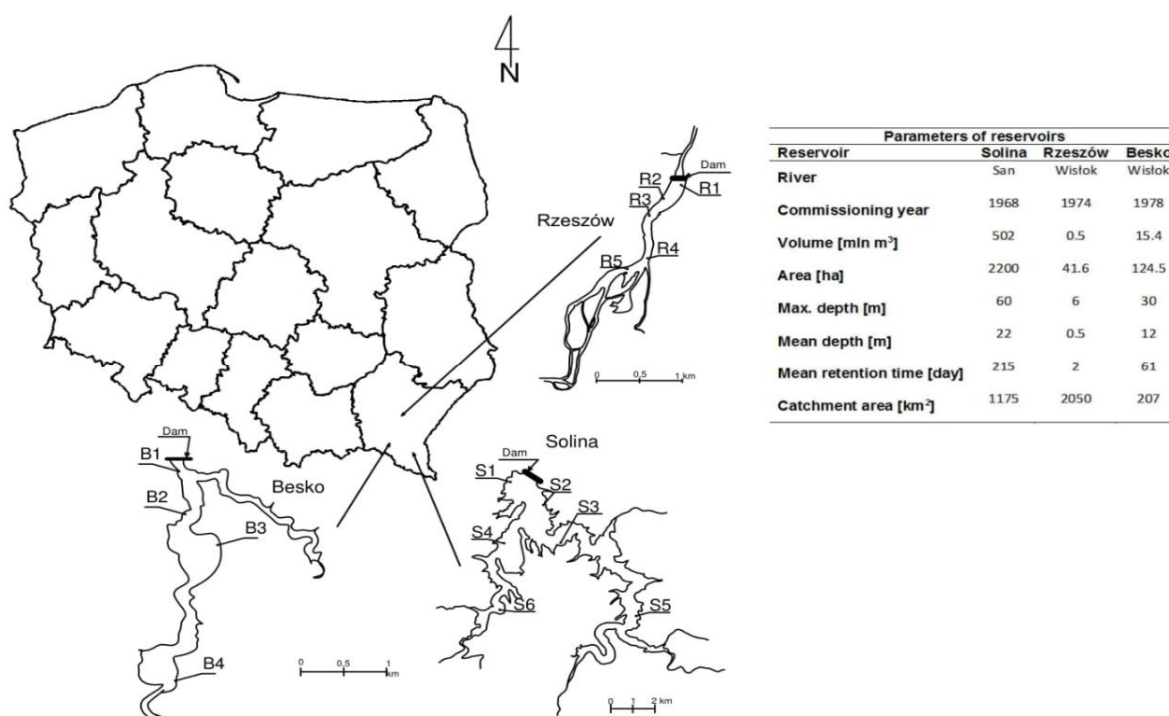


Fig. 1. Locations of the studied reservoirs and sampling stations

Besko Reservoir on the River Wisłok is another multi-purpose facility whose primary functions are

nevertheless in water storage and flood control. The catchment area has many villages located within it, and

is of an agricultural nature, albeit with forestry management also well-developed. There is no industry present, however, though agricultural enterprises do exist here. Construction serving recreational functions is a rarity in this area.

Sediment sampling and analysis

Sediment was collected using a gravity sampler (KC Kajak), once only at 6 research stations in Solina Reservoir (S1, S2, S3, S4, S5 and S6), 5 in Rzeszów Reservoir (R1, R2, R3, R4 and R5), and 4 in Besko Reservoir (B1, B2, B3 and B4). The locations of the Reservoirs studied and the sampling stations are as presented in Fig. 1.

From each sediment core, a top layer 5 cm thick was removed and dried to constant mass, before being milled and made subject to determinations of contents of organic matter (OM), total organic carbon (TOC), total nitrogen (TN) and black carbon (BC).

OM content was analysed by the loss on ignition method involving a temperature of 550 °C for 4h [30].

Prior to the analysis for OC and TN, carbonates were removed from samples by 72 h contact with vapor from 30 % HCl in desiccators [39]. Analysis was preceded by repeat drying of the sediment sample at 60 °C to constant mass. TOC and TN contents were measured subsequently applying a CN analyser (CN Flash EA 1112, ThermoQuest). For this purpose about 10 mg sediment subsamples were weighed to tin capsules. After folding the capsule the samples were placed in the autosampler. The tin capsules enclosing the samples were fallen into the oxidation reactor chamber where excess oxygen was introduced before. In the reactor filled copper oxide and silvered/cobaltic oxide, at about 900 °C the material was mineralised into simple compounds such as CO₂, H₂O and NO_x. The product gas mixture next was flowed through a silica tube packed with copper granules. In this zone held at about 600 °C remaining oxygen was bound and nitric/nitrous oxides were reduced. High purity helium (quality 6.0) was used as carrier gas. Finally the gas mixture was passed to a gas chromatographic system, where N₂ and CO₂ were detected with thermal conductivity detector. Blank (empty tin capsule) and standard samples of known elemental composition (acetanilide) were used in quality control.

The content of black carbon (BC) was determined using the chemical-thermal oxidation method (CTO-375) [14]. To this end, dried sediments were first made subject to removal of carbonates (in the same way as with TOC), prior to the combustion of samples in a muffle furnace supplied with a constant air flow (of about 200 ml/min), for 18 hours at a temperature of

375 °C. Residual carbon was then made subject to the BC determination using a CN analyser (CN Flash EA 1112, ThermoQuest).

Statistical analysis

Obtained results were described in a basic manner by reference to minimum, maximum and average values. R^2 values were also determined for linear relationships. The MS Excel 2013 program was used in calculations.

3. Results and discussion

The results of the study are as presented in Table 1.

The 3 Reservoirs were found to differ in terms of the OM content accumulated in sediments, across the 1.23-9.22 % range (Table 1). The highest OM content (6.83 % on average) characterised sediments of Rzeszów Reservoir, while the lowest related to Solina Reservoir sediments (with their average of 2.28 %). In both Rzeszów and Besko Reservoirs, values for OM content were progressively higher in sites located closer and closer to the dams.

Table 1

Contents of organic matter (OM), total organic carbon (TOC) and total nitrogen (TN), molar ratio of total organic carbon/total nitrogen (TOC/TN), content of black carbon (BC) and ratio of black carbon/total organic carbon (BC/TOC) reported for sediments in the Reservoirs studied

Station	OM	TOC	TN	Molar TOC/ TN	BC	BC/T OC
	(%)	(%)	(%)		(%)	(%)
S1	2.80	0.80	0.046	20	0.059	7.4
S2	1.23	0.37	0.036	12	0.018	4.8
S3	4.86	0.96	0.080	14	0.098	10.3
S4	1.40	0.33	0.016	24	0.002	0.7
S5	1.66	0.31	0.033	11	0.001	0.3
S6	1.73	0.52	0.022	27	0.011	2.1
R1	9.22	2.58	0.499	6	0.381	14.8
R2	6.40	1.56	0.279	7	0.220	14.1
R3	6.14	1.45	0.223	8	0.213	14.7
R4	4.83	1.35	0.234	7	0.124	9.2
R5	7.55	2.20	0.433	6	0.219	10.0
B1	5.93	1.81	0.120	18	0.236	13.0
B2	4.26	1.22	0.071	20	0.557	45.8
B3	4.07	1.11	0.053	24	0.135	12.1
B4	3.88	1.09	0.059	22	0.310	28.4

Reported contents of organic carbon (TOC) in sediment surface layers were in the 0.31-2.58 % range (Table 1). In all cases, trends for TOC resembled those for OM, albeit with the former on average accounting for 25, 27 and 29 % of the organic matter in sediments

from Solina, Rzeszów and Besko Reservoirs, respectively.

Significant correlations between OM and TOC were thus obtained for all the reservoirs (Fig. 2). Pearson correlation coefficients (r) significant at $p < 0.05$ were 0.9259, 0.9612 and 0.9981 respectively, in the cases of Solina, Rzeszów and Besko Reservoirs.

As noted already, the Reservoir sediments only had relatively low contents of organic matter, and of total organic carbon in consequence. However, the values obtained are in fact rather typical for Polish reservoirs. For example, sediments in Dobczyce Reservoir had OM contents in the range 1.76–3.13 %, as well as TOC values of 1.02–1.82 % [38]. In turn, in Rożnów Reservoir, OM content accounted for between 1 and 3 % of the dry mass of sediments [15], while in Sulejów Reservoir the content constituted 9.9–21.8 %, while values for TOC were in the 5.2–10.3 % range [4]. Further reported values for OM in sediments were, in Czorsztyn Reservoir – from 2.3 to 11.9 % [17], in Solina Reservoir, values varying in the ranges 8.72–10.11 % and 1.94–2.92 % respectively, and in Myczkowce Reservoir 10.78–11.94 % and 3.95–4.08 %, respectively [22]. Finally, in Włocławek Reservoir the average organic matter content in sediments was 11.5 % [36]. For comparison, the noted content of organic matter in the surface layer of sediments of the tropical, oligotrophic Balbina Reservoir in Brazil was 29–30 %, while total organic carbon there was at 12 % [1].

The content of total nitrogen (TN) ranged from 0.016 to 0.499 % (Table 1). As in the case of OM and TOC, the highest content for this indicator was recorded in sediments deposited in Rzeszów Reservoir (average 0.334 %), and the lowest in those of Solina Reservoir (average 0.039 %). All observed trends for content in the case of this indicator were analogous to those described above, and the TN content does correlate significantly with that of TOC (Fig. 2). Pearson correlation coefficients (r) at the significance level $p < 0.05$ were of 0.8412, 0.9922 and 0.9925 for Solina, Rzeszów and Besko Reservoirs, respectively. Carbon and nitrogen are the basic components of organic matter, and the relationships shown between these components indicate that organic nitrogen was the main component of total nitrogen [37, 12].

Differentiation of TOC and TN content in the sediments translated into differentiation of the molar ratio of TOC to TN (TOC/TN). Different values for this indicator were observed between both Reservoirs and their stations. The largest differences between stations were observed in sediments from Solina Reservoir, the TOC/CN ratio there varying across the range 11–27, with the average value being 18. In Rzeszów Reservoir, values for the TOC/TN ratio characterising the

superficial layers of sediment varied within the narrow range from 6 to 8, the average value being 7. Sediments collected from the bottom of Besko Reservoir were in turn characterised by values for the TOC/TN ratio in the 18–24 range, with an average value of 21 (Table 1).

The TOC/TN ratio is one of the indicators used most commonly in identifying the origin of organic matter deposited in the sediments of aquatic ecosystems. The assumption is that a TOC/TN value above 12 indicates matter of allochthonous origin, while a value below 8 denotes autochthonous organic matter [27, 18, 23]. The TOC/TN values characterising the sediments in Rzeszów Reservoir were thus indicative of an autochthonous origin of OM, while the OM deposited in Besko and Solina reservoirs would seem to be of terrigenous origin.

The black carbon content (BC) in the studied sediments ranged from 0.001 to 0.557 % (Table 1). The lowest values for BC (in the 0.001–0.098 % range, average 0.032 %) were those noted in sediments of Solina Reservoir. In contrast, higher values in the range 0.124–0.381 % (average 0.231 %) characterised sediments in Rzeszów Reservoir, while the highest values of all (between 0.135 and 0.557 %) were noted for sediments of Besko Reservoir, where the average BC content was of 0.310 %.

Sediments of Solina Reservoir manifested marked station-to-station variation BC content. Values significantly higher were noted for the sediments deposited in the lower and central parts of the Reservoir (at stations S1, S3 and S3), as compared with sediments from the upper part (at stations S4, S5 and S6). The share of TOC accounted for by BC was in the range 4.84–10.25 % at stations S1, S2 and S3, while only much lower figures in the range 0.32–2.11 % were reported for stations S4, S5 and S6 (Table 1). Analysis of the TOC/TN values showed that the organic matter deposited in the sediments of Solina Reservoir is terrigenous, while the significant correlation noted between BC and TOC ($r = 0.9626$, $p < 0.05$) (Fig. 2) indicated the same sources of origin [20, 19]. There are no industrial plants in the catchment of Solina Reservoir, so potential pollution must originate in territorial and point sources. In fact, the distribution observed for BC in the sediments of Solina Reservoir sustains the conclusion that the main rivers supplying the Reservoir are not its main source of BC in sediments. Indeed, the relatively high BC contents at stations in the central and lower parts of the Reservoir reflect soil erosion or BC atmospheric transport following the combustion of fossil fuels and biomass in villages close by, not least given the location of resorts there. The higher BC content may also be associated with the movement of small recreational boats, as there are many marinas in the region.

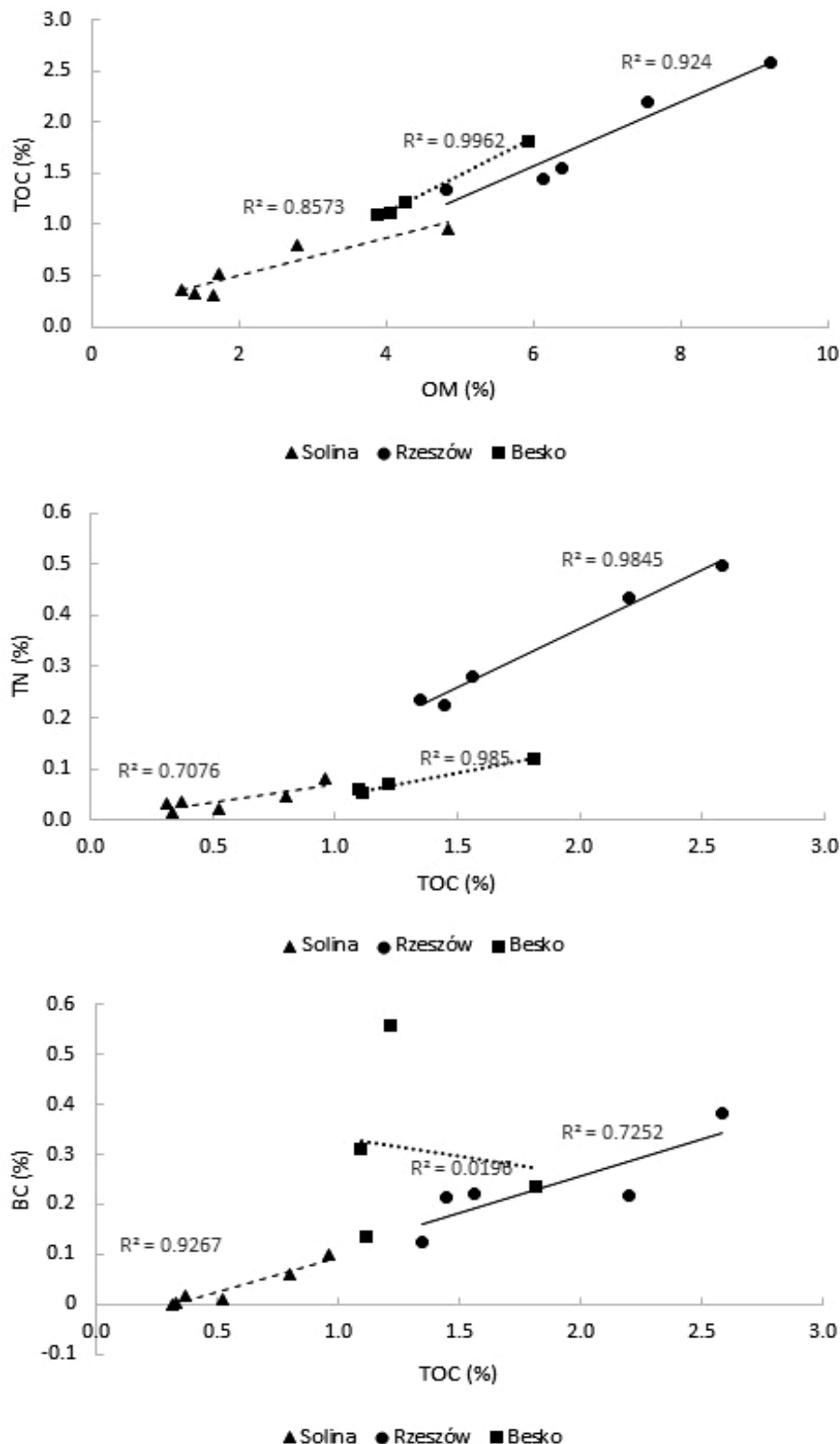


Fig. 2. Relationships between the selected parameters in the sediments studied

Sediments of Rzeszów Reservoir have BC contents about ten (or even 100) times higher than those in Solina Reservoir, with the BC fraction in TOC in the 9.22–14.76 % range (Table 1). Rzeszów Reservoir is a small,

degraded reservoir located in the city, near buildings, roads and industrial plants. Yet despite the significant anthropogenic impact on the Reservoir, its TOC/TN values suggest a mainly autochthonous origin for the

organic matter deposited in sediments. While BC contents were much higher than in Solina Reservoir, the shares of TOC it accounted for were not high. Account should, however, be taken of the fact that the OM content in these sediments was higher than in the other Reservoirs studied. As in Solina Reservoir, a significant correlation between TOC and BC was noted here ($r = 0.8516$, $p < 0.05$) (Fig. 2), the suggestion here being that the origin was the same.

It was in Besko Reservoir that reported BC contents in sediments were highest. The determined shares of BC in TOC ranged from 12.11 to 45.83 %, while the highest shares (of 28.35 and the aforesaid 45.83 %) were at stations B4 and B2, respectively. Station B4 is located near the main inflow to the Reservoir (the River Wisłok), while B2 is located near the village of Sieniawa in the catchment area of the Głębokki Stream, which flows between buildings in the village of

Głębokie a significant part of its length. The example provided by this Reservoir shows a clear anthropogenic effect on BC content in sediments. Very high shares of BC in TOC at stations B4 and B2 probably reflect combustion of fossil fuels and biomass on nearby farms. Like other authors [24] we found no BC-TOC correlation (Fig. 2).

For a comparison of how these BC levels relate to those in other aquatic ecosystems in different parts of the world, Table 2 assembles data from the literature. The sediments of Solina Reservoir can be seen to be characterised by low values for BC in sediments, and by the lowest share of TOC accounted for (in the upper part of the Reservoir in particular). In the case of the two other Reservoirs, BC contents and the shares of TOC they account for are within the range presented by other researchers for various types of aquatic ecosystem ranging from rivers to oceans.

Table 2

Concentrations of black carbon (BC), and percentage contents in total organic carbon (BC/TOC) they account for in the Reservoirs studied, as well as other aquatic ecosystems around the world

Ecosystem/Region	Method	BC (%)	BC/TOC (%)	Reference
Solina Reservoir/Poland	CTO-375	0.001–0.098	0.3–10.3	This study
Rzeszów Reservoir/Poland	CTO-375	0.124–0.381	9.2–14.8	This study
Besko Reservoir/Poland	CTO-375	0.135–0.557	12.1–45.8	This study
Molenplaat, Schelde estuary/The Netherlands	CTO-375	0.038–0.056	17–42	[29]
Iberian Margin, Atlantic Ocean	CTO-375	0.047–0.163	16–29	[29]
Northwestern Black Sea	CTO-375	0.197–0.815	13–24	[29]
North Sea	CTO-375	0.032–0.093	16–61	[29]
Eastern Mediterranean	CTO-375	0.112–0.729	25–39	[29]
Gulf of Maine/USA	CTO-375	0.03–0.17	3–9	[13]
Guanabara Bay/Brazil	CTO-375	0.03–0.31	2.3–12.8	[33]
Gulf of Cadiz/Spain	Simpson & Hatcher method	0.047–0.809	3.0–45.7	[11]
South Atlantic Ocean/Africa	CTO-375	0.08–0.16	3–12	[24]
South Atlantic Ocean/South America	CTO-375	0.04–0.17	8–34	[24]
China’s marginal seas	CTO-375	0.01–0.25	27–41	[20]
Lake Superior/Canadian	CTO-375	0.19–0.85		[32]
Changjiang River/China	CTO-375	0.03–0.19	10–57	[19]
Indus River/Pakistan	CTO-375	0.04–0.50		[2]
Baltic Sea/Poland	CTO-375	0.01–0.04		[35]

Many studies to date point to a large influence of the BC fraction on the sorption of hazardous organic pollutants, including PAHs and dioxins [e. g. 8, 21, 26, 35]. For this reason, the BC fraction should be considered among key factors controlling the fate of such pollutants in aquatic ecosystems.

Conclusions

First research into the black carbon content of sediments in reservoirs located in south-eastern Poland

shows that, despite the relatively limited nature of this content, the share in total organic matter accounted for may suffice to affect the migration and fate of certain dangerous pollutants. The main source of BC in the sediments analysed was most likely the combustion of fossil fuels and biomass within the Reservoirs’ catchment areas. The lack of potential sources of BC close to a Reservoir did not prove to be a determinant of low content in sediments, since deposition of this carbon fraction in surface waters, and subsequent sedimentation, may involve transfers over very large

distances from the source, depending of course on the prevailing atmospheric circulation.

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