ORIGIN, TRANSFORMATION AND ROLE OF DISSOLVED ORGANIC(HUMIC)SUBSTANCES IN LAKE BALATON

PhD Theses

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1. Scientific preliminaries

The research of aquatic humic substances was not important in limnology, in fact it was not until the mid 70's that aquatic humic substances were brought into focus in environmental science. The event trigging was the discovery that it could be precursors of carcinogen substances (trichalomethane) (BELLAR *et al*., 1974), while photochemical degradation is known to be involved in the breakdown of toxic chemicals including oxygen radicals can be formed (COOPER *et al*., 1989). In most natural waters a major portion of DOC is dominated by dissolved humic substances which may rich up to 80% of DOC in the lakes (STEINBERG & MÜNSTER, 1985). Dissolved humic substances complex or sorb major cations and due to the catalysis and potolytical reaction could change both bioavailability and geochemical cycling of other organic (algaetoxin, pesticide) and inorganic (trace metals, nutrients) compounds (ALLARD *et al*., 1991). Humic substances strong absorb the lowest wavelength of the light, hereby they change the underwater light climate (KIRK, 1976; BRICAUD *et al*., 1981).

Previously the brown coloured water inflow from the peat lands was thought negligible in respect to the water quality of the Lake Balaton, although the change in the view took place with two decades lag in the middle of 90's. At the flooding of lower Kis-Balaton reservoir (in the year of 1993) the concentration (as Pt-colour) of dissolved organic substances increased with order of magnitude at the mouth of the River Zala according to the upper section of the River Zala (V.-BALOGH & VÖRÖS, 1996). At this time it was thought that the source of the brown coloured humic substances is the peat marshy land of the Kis-Balaton reservoir, which is washing out in some years. It was measured that the contribution of humic substances to the DOC is around 40% in the upper section of the River Zala, while it increases to 75% at the outflow of lower Kis-Balaton reservoir and it hardly decreases in some cases at the Keszthely basin (V.-BALOGH & VÖRÖS, 1999). This results suggested that the importance of dissolved organic(humic)substances became more important in aspect of the water quality of Lake Balaton. The knowledge of quality and quantity of the dissolved organic matter load of Lake Balaton reaching it through the River Zala, the qualitative transformation of oganic(humic)substances until the water residence time in the lake, their biological availability and of their role in the formation of the underwater light climate were insufficient.

2. Aim of the study

The aim of my study was to determine the humic properties of dissolved organic matter load of Lake Balaton reaching it through the River Zala under changing hydro-meteorological circumstances (1). The changes of concentration and qualitative transformation of organic (humic) substances was measured along the longitudinal axis of Lake Balaton (2). The aim was to determinate the qualitative transformation of the dissolved organic (humic) substances due to the photochemical (UV-radiation) and bacterial degradation (3), as well as their biological availability (4). Moreover the aim of my study was to determinate the contribution of dissolved humic substances to the attenuation of the underwater photosynthetically active radiation (PAR) and particularly to the extinktion of the ultraviolet – UV-A and UV-B - radiations, comparing with other influencing parameters - suspended solids, phytoplankton (as chlorophyll-a) and dissolved coloured organic substances (Pt-colour) (5).

3. Materials and methods

The experiments and the measurements were carried out with the water samples taken from the River Zala (main inflow of Lake Balaton) and from the 5 sampling stations of Lake Balaton along the longitudinal axis (Keszthely basin, Szigliget basin, Balatonszemesi basin, Siófok basin (at Tihany and at Balatonfűzfő) between 1999 and 2004. The organic carbon concentration was measured by an Elementar High TOC analyser. The humic substances (HS) were isolated by low pressure chromatographical XAD-method (STANDARD METHODS, 1995). Amberlite® XAD-7 (®Rohm and Haas Co) (Aldrich Chemical Company, Inc.) non ionic (20-60 mesh) polyacrylic (acrylic-ester) resin and Pharmacia C type low pressure chromatography column were used. The colour of the water was determined by a Shimadzu UV 160A spectrophotometer (at the absorbance 440 nm) (CUTHBERT & DEL GIORGIO, 1992). The fluorescence intensity of dissolved organic substances was determined by a Hitachi F-4500 fluorescence spectrophotometer. The size fractionation of organic substances was carried out with frontal ultrafiltration procedure. Amicon cell and Millipore membranes were used. The dissolved organic carbon (DOC) availability (bacterial degradation) experiment was carried out according to SERVAIS *et al*. (1989) and Waiser & Robarts (2000). For the enumeration of bacterioplankton epifluorescence microscopy (Nicon Optiphot) and acridin orange

fluorochrom were used. The water temperature and the underwater light intensity (PAR, UV-A and UV-B) were measured in situ by a PUV-2500 Radiometer (Biospherical Instrument). The water discharge data of the River Zala which was used to the calculation of organic carbon load was given from the West-Transdanubian District Environmental and Water Authority.

4. Theses

(Summary of new results)

1. The annual dissolved organic carbon (DOC) load of the River Zala was significantly higher (2617 tons) in the rainy 2004 year than in the droughty 2003 (1138 tons). The DOC concentration has not revealed any positive correlation with the increasing discharge in the rainy year, while it decreased by the increasing discharge in the droughty year. The contribution of humic substances (HS) to the total DOC was lower in 2004 than in the droughty 2003. A close correlation was found between the temperature and the DOC concentration in both years. The obtained results suggest that the increased discharge of the River Zala plays a higher roll in the washing out the organic substances from the Kis-Balaton reservoir in 2004 than the dilution of their concentration has less importance on the DOC load.

2. The allochthonous dissolved organic substances getting into the Lake Balaton (and the autochthonous, partially those developed there) go through quantitative and mainly qualitative changes in the course of their way along the longitudinal axis of the lake from the mouth of the River Zala to the outflow. The DOC concentration and the fluorescence and the colour intensity as well as the contribution of the HS concentration to the total DOC pool decreased, while the relative amount of the largest molecular size fraction of DOC incresed. The contribution of humic substances to the total DOC was dominant both in 1999 (75%) and in 2003 (65%) at the mouth of the River Zala, which decreased to 55% and 52% to the eastern basin of the lake. The contribution of fulvic acids to the pool of humic substances increased from 75% to 99% during the residence in the lake in 1999, while it varied from 80% to 99% in 2003.

3. In the background of these changes (Thesis 2.) are the photochemical and microbial degradation processes. Due to the photolysis (direct and indirect) the DOC concentration, the intensity of fluorescence and colour and the contribution of HS decreased. The contribution of HS by the microbial degradation decreased, too. In connection with the change of nominal molecular weight cut-offs of the organic

substances were the largest DOC fraction in both the microbial (> 10000 Da) and the photolytical (3000-10000 Da) degradation.

4. According our results the biological available DOC was between 1,4 – 1,8 mg l^{-1} (9 - 14%) in the River Zala. There were no significant differences found in the BDOC values of the experimental variants (original water and with inorganic nutrients addition). The BDOC concentration was between 0,4 - 0,8 mg l^{-1} (6 - 9%) in the water of the Keszthely basin, the lowest value was found in the winter and the highest one in the autumn. No significant differences were found in the values of the BDOC concentration between the treatments. The maximum BDOC concentration (0,52 mg l^{-1}; 5%) has been found in the original water of Siófok basin in summer, when the bacterial growth was limited by the inorganic nutrient. The maximum BDOC value has been found in summer, due to the photolytical degradation, which increases the biological availability of organic substances. The decrease of the biologically available dissolved organic carbon from the mouth of the Zala River to the eastern basin of the lake demonstrates that the organic substances became more refractory and more unavailable during their residence in the lake.

5. In the water coloumn the depth of the penetration of the light increased from the mouth of the River Zala to the eastern part of the lake, but it decreased by the decrease of the wavelength in all cases. This means that the light penetration depth is the highest at the eastern basin of the lake and the lowest at the mouth of the River Zala. The 1% depth (depth of penetration to 1% of the sub-surface irradiance) of the photosynthetically active radiation (PAR) was 3.5 m, UV-A 2 m and UV-B 1 m at the eastern basin of the lake. The 1% depth of PAR was 2.5 m, UV-A 0.3 m and UV-B 0.1 m at the mouth of the River Zala. These results suggest that there are higher differences between underwater UV-climate of open water areas of Lake Balaton and mouth of the River Zala than between their PAR climate. It was shown that the role of dissolved humic substances is dominant in the underwater light climate at the mouth of the River Zala. The role of suspended solids is dominant in Lake Balaton, but in addition to this, the contribution of dissolved humic substances is often equal with that of the algae in forming the light climate.

5. Relevant publications

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