

## THE POLLUTION LOAD BY NITROGEN AND PHOSPHORUS IN THE CETINA RIVER

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### ABSTRACT

*The objective of the investigations of the Cetina River, located in southern Croatia, was to record specific characteristics and properties of the Cetina waters at nine (9) stations. In addition to measurements undertaken in the Cetina River, the water quality of its most significant springs and tributaries, such as: Kosinac, Šilovka, Studenci and Mala Ruda, Velika Ruda, Grab has also been measured. The water quality in the Cetina watershed has been evaluated in the following storage reservoirs: Peruča, Buško Blato, Prančevići. The nitrogen compounds and phosphorus concentrations have been estimated at all these sampling sites over a 3 year period (2005-2008). Concentration levels at the Cetina - Vinalić sampling site for total N (from August 2005 to December 2008) ranged from 0 to 1.759 mg/L, for NH<sub>3</sub> – N from 0 to 0.374 mg/L, for NO<sub>3</sub> – N from 0.063 to 0.916 mg/L, for PO<sub>4</sub> – P from 0 to 0.099 mg/L. The results prove that the Cetina – Vinalić sampling site is not polluted by nitrogen and phosphorus compounds. The river section from Trilj to the Prančevići dam, where the water is used for the water supply of Omiš, Makarska and Dalmatian islands, has been polluted by waste water because the majority of agricultural area, roads, industry and settlements are located upstream of it. The highest concentration for total N of 1.128 mg/L and of 1 527 total coliforms in 100 mL, expressed as a mean value for a 3-year period of investigations was found at the sampling site Trilj. The results of concentration changes at the Čikotina Lađa and Cetina Radmanove Mlinice sampling sites show no regularities. The highest concentration for total N of 0.941 mg/L was measured at the Cetina Radmanove Mlinice during 2007. The highest concentration for NO<sub>3</sub> – N of 0.916 mg/L was measured at the same sampling site. According to the investigations of the water quality of the Cetina springs and tributaries the bacteriological most polluted river spring is Kosinac and the bacteriological most polluted river tributary is Grab. With reference to the water quality in the Cetina storage reservoirs it may be concluded that the lowest quality standard has been found within the Prančevići storage reservoir regarding nitrogen compounds and phosphorus concentration levels.*

**Keywords:** total nitrogen, ammonia, nitrate, total phosphorus, Cetina River, Croatia

### 1. INTRODUCTION

Nitrogen and phosphorus are important nutrients in the environmental systems of the water resources. Consequently, they are indispensable for the growth and reproduction of organisms. The interruption in the circulation of these two elements can result in water eutrophication (Berbenni and Galassi 1978) and can destroy the environmental living conditions. Consequently, the nitrogen and phosphorus compounds are important parameters which exhibit the sanitary-chemical, i.e. hygienic state of water quality. The presence of ammonia in water is an indicator of direct pollution. The presence of nitrates proves that the pollution is not recent, that the methods for nitrogen compounds removal are efficient and that the ability for autopurification in the river is high. The increased presence of phosphorus and nitrogen compounds in water is mainly caused by the use of artificial fertilizers and insufficient monitoring of the release of domestic waste water containing detergents as a source of phosphates (Maki et al. 1984; Graham 1995; Stanners and Bourdeau 1995). The Cetina River is a karst river so that the permeability of the surrounding soil in the entire catchment (Štambuk-Giljanović 1999, 2006; Bonacci 1987) and the low ability for autopurification of the groundwater make it possible for the river to be easily polluted. Human activities increasingly pollute the Cetina River. Their influence can be observed by the release of increased concentrations of nitrogen and phosphorus into the streamflow. Considering the importance of the Cetina River for

the water-supply of the Makarska and Omiš regions and Dalmatian islands (Brač, Hvar and Šolta) the objective of the investigations was the systematic monitoring of the changes in the nitrogen and phosphorus natural concentration levels and increased concentration due to natural and/or pollution processes from the Cetina Spring to its estuary carried out over a 3 year period (2005-2008). The Mediterranean region to which Croatia belongs is specific due to a nonhomogeneous pedologic environment, different hydrological systems and temperature regime characteristics of various climatic regions, together with the distribution of the population and human activities in national territories (CEC 1994). According to the opinion of various authors (Sequi and Vianello 1993) the process of identification of springs in the risk zones implies the absorption of substances from isolated terrains which are transported by water and the classification of areas according to the water circulation regime which is especially important for obtaining data on different water pollution sources. The draining of agricultural areas, particularly in karst regions, can lead to pollution of both surface water and groundwater (Štambuk-Giljanović 2003) so that special attention should be paid to this problem.

## 2. STUDY AREA

The Cetina River spring is located at 382 m a.s.l. at the bottom of Dinara Mountain in the village of Cetina, north of Vrljka. The Cetina streamflow is 105 km long and its estuary is near Omiš (Figure 1). Its upstream flow, 56 m long to Trilj, is the recipient of water from numerous large karst streamflows and tributaries, the most important being the Mala and Velika Ruda Rivers which flow into the Grab rivulet which flows into the Cetina River.

The Mala Ruda spring is an intake for the regional water-supply for settlements from Sinj, Trilj and Dugopolje to Klis and Muć. The Kosinac spring on the left bank of the Cetina River near Han is the water intake for the Sinj water-supply. In its downstream section, from Trilj to its estuary in the sea, the Cetina is a typical mountain river with a slope of 290 m. A small reservoir is located downstream from Trilj where it has been diverted from the river by a 9 km long tunnel across the reservoir at Gata to the hydroelectric-power plant in Split.

At the reservoir at Gata the Cetina's intake is used for the water-supply of Omiš and the islands of Brač, Šolta and Hvar. The Đale dam hydroelectric power plant is located in the Cetina canyon, 5.8 km downstream from Trilj. In this way it is possible to exploit the energy potential on the 21 m slope between the Sinjsko polje (field) and the Prančevići reservoir. For the production of electric energy it uses the routed streamflow of the Cetina catchment area. The Cetina River near Zadvarje has intakes for the water-supply of the Makarska riviera (from Brela and Makarska to Zaoštrog).

The Cetina River with its wider catchment area, covering ca 4000 km<sup>2</sup>, is one of the largest streamflows in the Dinaric karst. The average annual discharge at the Cetina estuary is ca 116 m<sup>3</sup>/s. The total slope from the spring to the estuary is 382 m wherein one third refers to the Gubavica waterfall. The most favourable conditions for the construction of the significant Kraljevica hydroelectric power plant in 1912, the largest in Europe at that time, were the abundance of water that the Cetina River receives by underground circulation from the karst hinterland and an exceptionally steep slope in the narrow valley. The Peruča hydro-electric power plant was constructed to exploit all available hydropotential of the Cetina. This was followed by the construction of the Split HP (Zakučac) and Orlovac HP plants. The latter plant, due to the hydroenergetic value of its Buško Blato reservoir (Štambuk-Giljanović 2001) is the most important plant in the entire Cetina catchment. Domestic and industrial waste water (Cetinka plastics factory in Trilj, Dalmatinka thread factory in Sinj, Galeb textile plant in Omiš) is released into the Cetina River. The largest point sources of pollution by domestic waste water are Sinj, Trilj and Vrljka.

Treatment plants have been built for purifying the waste water from the catchment area in order to decrease the pollution load. A biological treatment plant was built at Trilj, for the equivalent

of 10 500 (EI) (EI=water consumption of 200 L/day/person); later a mechanical and a biological treatment plant was built in Sinj for 45, 000 equivalent inhabitants. Plans for two more plants are being developed: biological treatment plant in Vrlika (6600 EI) with a submarine outfall and a mechanical treatment plant in Omiš (32,000 EI). The Cetina-Vinalić sampling site is located 5 km downstream from the Cetina spring while the Čikotina Lađa site is in the village of Čikotina on the left bank of Cetina, which is ca 45 km from the spring and ca 1.5 km downstream from the Prančevići dam. It controls the release of water from the dam and/or spillways.

At the Čikotina Lađa site the water flows into the Cetina riverbed after the intersection of two flows at the dam and transportation to the Zakučac HP. The Radmanove Mlinice hydrological sampling site is 85 km from the Cetina spring. The point pollution sources include towns, industry and tourism, while the diffusion pollution sources include agriculture, of particular importance in the Sinjsko polje, as well as pollution from intensive local and regional traffic. Traffic pollution is relevant during the rainfall period.

An integrated plan for the management and protection of the Cetina catchment area should be developed (UNEP/MAP/PAP 2000) after extensive investigations of that area. This calls for an integrated and interdisciplinary approach in developing legal regulations and international agreements.

### 3. METHODS

The quality of the Cetina water has been monitored at nine (9) sampling sites as follows: Vinalić, Peruča dam, Trilj, Đale, Prančevići dam, Čikotina Lađa, Gata, Zadvarje, Radmanove Mlinice. In addition to the measurements undertaken in the Cetina River, the water quality of its most significant springs (Kosinac, Šilovka, Studenci) and tributaries such as: Mala Ruda, Velika Ruda, Grab has also been measured. The water quality in the Cetina watershed has been evaluated in the following storage reservoirs: Peruča, Prančevići, Buško Blato. All these analyses have been carried out annually 12 times on a monthly basis during a three year period (2005-2008) and include the following parameters: temperature, pH, turbidity, conductivity, evaporation residue, alkalinity, total hardness, dissolved oxygen, percentage of oxygen saturation, biological oxygen demand, total N, ammonia, nitrates, total P, chlorides, sulphates,  $\text{KMnO}_4$  consumption, total coliforms in 100 mL. The quality parameters in the analysed water were determined according to American Standards Methods (AWWA 1995). At sampling sites Cetina – Vinalić, Cetina Čikotina Lađa and Cetina Radmanove Mlinice the measurements included, in addition to the nitrogen compounds and phosphorus concentrations, the discharges ( $\text{m}^3/\text{s}$ ) which corresponded to the sampling dates. The discharge was measured by a hydrometric propeller and was calculated by multiplying the mean velocity profiles according to the verticals of cross sections with the streamflow cross section surface (Jovanović et al. 1986). Loads for nitrogen compounds and phosphorus during a 3-year period (2005-2008) have been calculated at the sampling site Cetina – Vinalić. The loads ( $\text{kg}/\text{d}$ ) were calculated as concentrations ( $\text{kg}/\text{m}^3$ ) multiplied by discharge ( $\text{m}^3/\text{d}$ ).

### 4. RESULTS

Samples have been collected on the Cetina River streamflow, on its springs and tributaries and on the storage reservoirs in its watershed. The results of physical, chemical and microbiological investigations are presented in Tables 1 – 3 as the mean 3 – year values (2005 – 2008). The mean water temperature in the Cetina River streamflow, its springs and tributaries (Tables 1 and 2) varied from 10.4 to 13.3° C and in storage reservoirs from 11.4 to 17.7° C (Table 3). The mean values of dissolved oxygen were high at all 18 sampling sites. According to the concentrations given in the standard statistical classification of surface fresh water quality for the maintenance of aquatic life presented by the United Nations Economic Commission for Europe (UN/ECE) and used in the

statistical compendium for the Dobris Assessment (Stanners and Bourdeau 1995) the concentrations of dissolved oxygen were usually close to 10 mg/L in unpolluted waters. All samples satisfied that criterion. According to UN/ECE criteria unpolluted rivers typically have BOD<sub>5</sub> value of 2 mg/L or less. At all sampling sites the mean BOD<sub>5</sub> values were 27% higher than 2 mg/L. There were small variations in the mean concentrations of nutrients (those of total N, ammonia, nitrates, total phosphorus) between sampling sites. The concentration levels of total phosphorus, NH<sub>3</sub> – N, NO<sub>3</sub> – N and total nitrogen at the sampling site Cetina Vinalić are presented in Figure 2 using the correct experimental discharge – concentration data pairs. According to the research results for total N for which natural concentration amounts to 1 mg/L (Official Bulletin No 46/94) it is evident that the Cetina - Vinalić site is not polluted. The highest concentration during a three year period was 1.753 mg/L and the discharge was 2.38 m<sup>3</sup> /s. If the concentration of 0.1 mg/L NH<sub>3</sub> is considered to be natural and if a concentration of 0.3 mg/L is considered as more serious pollution, then one result in three years exceeded the concentration level of 0.3 mg/L (20 August 2006). The mean value of NH<sub>3</sub> – N at this sampling site is 0.0309 mg/L (Table 1). The high value of natural concentration levels for NO<sub>3</sub> – N from 10 mg/L allows relatively high concentrations without significant pollution. The mean concentration for NO<sub>3</sub> – N of 0.508 mg/L was found at the Cetina – Vinalić site. The natural concentration for PO<sub>4</sub> – P amounts to 0.1 mg/L. At the sampling site Cetina – Vinalić its concentrations ranged from 0 to 0.254 mg/L. The average annual load values for nitrogen and phosphorus compounds expressed in t/year for this sampling site are presented in Figure 4. The load (t/year) for total N was highest in 2008 as well as the load for NH<sub>3</sub> - N which shows that the sampling site was directly threatened by faecal waste the first time ever. The highest mean value concentration for total N of 1.128 mg/L (Table 1) was found on the sampling site Trilj. From the above data it is evident that the most polluted section is the area extending from Trilj to the Prančevići dam. This area has been mainly polluted by waste waters because the majority of agricultural areas, roads, industry and settlements are all located nearby. This section has been extremely vulnerable to pollution after the formation of the water retention of the Đale and Prančevići storage reservoirs, within which the pollution has been accumulated and dissolved. In terms of water supply applications, the degradation rate of water quality has reached its maximum in this section, as it is the water from the comparatively most polluted Prančevići dam, that has been supplying the regional water lines of Makarska and Omiš – Brač – Hvar – Šolta. This is made worse by the fact that the Cetina waters of this section supply the underground waters of the Jadro spring which in turn, constitutes the water supply for 500, 000 inhabitants of the town of Split. In terms of pollution, the next most affected region is the river section crossing the Sinj field. This section has been overloaded with waste waters discharged from the town of Sinj and its local industry, in addition to pollution deriving from agricultural and badly managed urban sectors. The mean concentrations for total N of 0.21 mg/L and for total P of 0.016, i.e. 0.034 mg/L were found at the Zadvarje and Gata sampling sites. These sampling sites are captured for the water supply systems of Omiš, Makarska and Dalmatian islands. For the entire period under study NO<sub>3</sub> – N concentrations at the Čikotina Lađa site ranged from 0 to 0.950 mg/L (Figure 3). At the Cetina Radmanove Mlinice station the concentration levels ranged from 0 to 0.992 mg/L. During the entire period the NO<sub>3</sub> – N concentrations were significantly lower than 10 mg/L which is the natural concentration level (Official Bulletin No 46/94). The PO<sub>4</sub> – P concentration levels at the Čikotina Lađa site range from 0 to 0.284 mg/L (1 February 2008) while at the Radmanove Mlinice site they range from 0 to 0.181 mg/L (25 June 2008). The results for PO<sub>4</sub> – P show that the Čikotina Lađa site is not polluted by phosphorus which was also proved for the Cetina - Vinalić site. Considering the results for total N it was concluded that twelve (12) results for total N exceeded the value of 0.3 mg/L during a three year period, which is considered to be increased pollution. Comparing the results obtained at the Cetina Radmanove Mlinice site with those from the Cetina - Vinalić and Cetina Čikotina Lađa sites it is evident that the former is susceptible to the most serious threat by pollution of nitrogen compounds. If the results for NH<sub>3</sub> – N which show direct pollution by faecal coliforms are considered it can be seen that at the

Cetina Radmanove Mlinice location six concentrations exceed natural concentration levels of 0.1 mg/L so that the pollution threat is greater at the Cetina Radmanove Mlinice (Figure 3) than at the Cetina Vinalić and Čikotina Lađa sites. The third section, beside the Cetina River mouth, is the most endangered area. This section has undergone important changes and is densely populated. The pollution loads transferred by the river have been compounded by local pollution; this resulted in the extinction of a number of species previously found in the river mouth, an area where the river flow mixes with seawater. Studenci, Kosinac and Šilovka are the springs in the river watershed which are still used for water supply purposes without prior water treatment except for water disinfection. The concentration levels of total N and total P in these springs do not exceed the concentration levels of serious pollution (Table 2). The construction of the Šilovka water supply system had started prior to the recent hostilities in Croatia based on utilisation of the Šilovka spring which is located next to the Cetina River at the foot of the Peruča storage reservoir. The system is relatively simple with two pressure pipelines connecting the Šilovka pumping station with the river banks. It supplies all settlements within the area extending from the Peruča dam to the town of Sinj. In tributaries of the Cetina River the mean concentrations for total N of 0.513 mg/L and for total P of 0.039 mg/L have been increased in Velika Ruda with reference to Mala Ruda and Grab where the concentrations of total N and total P have been lower (Table 2). Considering the water resources management the major project undertaken within the entire watershed is the construction of the Orlovac HP plant with its storage reservoir Buško Blato in the territory of Bosnia and Herzegovina. In the storage reservoirs of the Cetina River : Peruča, Prančevići and Buško Blato the water quality was satisfactory. The lowest water quality has been found within the Prančevići reservoir (Table 3).

## 5. DISCUSSION

The physical- chemical characteristics of the Cetina River are typical for karst water of calcium- hydrogencarbonate type (Štambuk- Giljanović 2002). The relationship between the average evaporation residue, hardness, alkalinity, chlorides and sulphates is defined mainly by natural factors; over a several year period it remained constant. However, the sanitary-chemical hygienic features are not only influenced by natural conditions but also by antropogenic factors. Nitrogen and phosphorus display such an influence (Benedetti et al. 1995; Smith et al. 1992). Although these are the most important nutrients for the growth and reproduction of organisms, the excessive concentrations of their compounds can result in the accelerated growth of algae (Urlich et al. 1996; Várallyay et al. 1995). The concentrations of nutrient salts in the Cetina River are relatively low when compared with the rivers in Western and Central Europe (Stanners and Bourdeau 1995). The concentrations of total phosphorus in many European rivers exceed 0.1 mg/L for total P and 1 mg/L for total N. Those values resemble the values in the rivers in Finland (Niemi 1998). The concentrations of total phosphorus, ammonia, nitrates and total N at the sampling site Cetina – Vinalić comply with the concentrations defined in the statistical classification of the European Economic Commission of the United Nations (UN/ECE) presented in the Dobris Assessment (Stanners and Bourdeau 1995). The worst situation considering water quality is evident in the Trilj (Table 1) area and downstream of Trilj, where the total pollution of the upstream flow accumulates in the reservoirs of the Đale and Prančevići HP plants. The water quality in these sections is almost unfit for consumption, due to significant concentrations of nitrates, bacteria and phosphorus. The necessary protection measures have been undertaken by the implementation of water treatment systems in Trilj. In the entire watershed there are still no plants for waste water treatment and no sewage systems. This situation is definitely responsible for the pollution of the waters in the watershed. Thus, the main pollution sources are the settlements. A positive aspect is the fact that this region is not densely populated. The total number of inhabitants in the watershed area is about 200, 000. This region is still rather poorly developed in the industrial sense. The sum of waste quantities is still small in proportion to the existing water quantities, i.e. water abundance so that the release of waste



materials and substances is very large and the concentrations of pollutants low. The water of the Cetina River could be used for drinking after adequate treatment. The Cetina River possesses a substantial water supply and energy supply potential. Despite the fact that the majority of its waters originate in karst springs, the river constitutes a potent flow, characterised by powerful and rapid fluctuations. Cetina's most important uses related to hydro-electric applications have long been recognized as the river most resourceful potential. In addition to the regulation of its utilisation and the structures required for its hydro-electric exploitation, several other measures have been undertaken to ensure the water supply of the wider area, including agricultural irrigation and the prevention of flooding. Furthermore, Cetina and its tributaries are the recipients of all waste water generated in the watershed. The construction of the most significant hydro-electric stations had started by the end of 19<sup>th</sup> century. By 1912, the first phase of the Kraljevac hydro-electric power plant has been completed. Its second phase was completed in 1932. The second phase of construction in the Cetina watershed started after the Second World War. In 1960, the storage reservoir and HP plant in Peruča were constructed as a multi-purpose dam plant, given that in addition to its electricity generation function it has influenced positively the Cetina hydrological regime by reducing flood wave generation, as well as by improving water discharge from the protected surface area of the Sinj field. Furthermore, the accumulated water in its storage reservoir facilitated the establishment of an irrigation system, as well as the water supply for both towns and industry. The additional construction of the storage reservoir and the Peruča HP plant enabled the regulation of the Cetina's water level, whilst harnessing its energy to the needs of the Kraljevac HP plant located downstream of the river, and from 1961 onwards, to those of the Zakučac HP plant, and for HP plant Đale from 1989. With the operation of the Zakučac HP plant, the hydro-electric utilisation of the Cetina River was continued in its downstream segment with the largest existing river inclination. The first phase of the Zakučac HP plant started its regular operation in 1962, and its second phase in 1980. In addition to its energy potential, the Zakučac HP plant had also been recognised for its water supply capacity. This fact justifies its significance as a multi-purpose project. With reference to water resources management, the major project undertaken within the entire watershed, was the construction of the Orlovac HP plant, in the territory of Bosnia and Herzegovina. This HP plant system was completed in 1973. Apart from the above multi-purpose stations, the river energy potential has been utilised in the area between the Sinj field and the Prančevići storage reservoir by the Đale HP plant since 1989, when its regular operation started. The sustainable development management and protection of the Cetina River watershed requires a new development structure and the mobilisation of all factors (international, national, regional and local) in order to create new foundations for the development and prosperity of the area. It will not be possible to achieve this goal through partial consideration of individual sectors such as energy production, water resources management, environmental protection and urban expansion and economic activities. An integrated and inter-disciplinary approach is needed which can in turn serve as the basis for a common system of natural and international legislation (agreements).

It is necessary to define hazardous areas and their catchments and protect the Cetina River from pollution (Coccosis et al. 1999; OECD 1982, USEPA 1995, 1997; UNEP/MAP/PAP 2000) for both surface water and groundwater.

## 6. CONCLUSIONS

1. The water of the Cetina River investigated at nine (9) sampling sites is a typical karst water of calcium hydrogencarbonate type, of relatively low mineralisation ( evaporation residue approx. 300 mg/L), of small total carbonate hardness (from 182 – 222 mg/LCaCO<sub>3</sub>). It is a moderately hard water and contains low concentrations of sulphates (from 12 – 30 mg/L). The chemical composition of the river water does not change very much during the year. The water temperature is within the range of 10.4 – 13.3 °C.

2. Concentration levels at the Cetina Vinalić sampling site for total N for a 3-year period (2005-2008) ranged from 0 to 1.759 mg/L, for  $\text{NH}_3 - \text{N}$  from 0 to 0.374 mg/L, for  $\text{NO}_3 - \text{N}$  from 0.063 to 0.916 mg/L, for  $\text{PO}_4 - \text{P}$  from 0 to 0.099 mg/L which proves that this sampling site is not polluted by nitrogen and phosphorus.

3. The deterioration of water quality at the river section from Trilj to the Prančevići dam poses serious problems in water supply since the water from the Prančevići dam is used for the water supply of Omiš, Makarska and Dalmatian islands: Brač, Hvar, Šolta. The highest concentration for total N of 1.128 mg/L and of 1 527 total coliforms in 100 mL, expressed as mean value for a three year period of investigations, was found at the Trilj sampling site.

4. The results of changes in concentration levels for several years monitoring (from 2005 to 2008) at the Cetina Čikotina Lađa and Radmanove Mlinice show no regularities. The highest annual concentration level of 0.941 mg/L for total N was measured at the Cetina Radmanove Mlinice during 2007. The highest concentration level of  $\text{NO}_3 - \text{N}$  of 0.916 mg/L was measured at the same sampling site.

5. According to the investigations of the water quality of the Cetina springs (Kosinac, Studenci, Šilovka) and tributaries (Mala Ruda, Velika Ruda, Grab) the bacteriologically most polluted river spring is Kosinac and the bacteriologically most polluted river tributary is Grab.

6. With reference to the water quality in the Cetina storage reservoirs it can be concluded that the lowest water quality has been found within the Prančevići storage reservoir.

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