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Letter to Editor

Wastewater Purification by Muck Soil and Willow (*Salix Americana*)

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Abstract

Our paper presents results of a study concerning ammonium and nitrate(V) uptake by soil irrigated with communal wastewaters (1 and 2 doses) and estimation of the possibility of using organic soil and willow (*Salix americana*) for wastewater cleaning. It was found that the highest biological and physical purification effect by studied soil and plant was observed after the first 24 hours from the moment of the wastewaters application. It was also demonstrated that a single irrigation dose was better utilized than a double dose. An effect of the season of the year on the final purification effect was observed. It was concluded that the studied soil and the plant applied showed very high capacity of binding ammonium ions (up to 96%), and lower in the case of nitrates(V) (up to 69%).

Keywords: muck soil, hydrophysical properties, willow, wastewater purification

Introduction

Water is a fundamental abiotic element of the environment, influencing numerous interdependent processes occurring in ecosystems. The rational disposal of natural water resources, their transformation into disposable resources and their protection against pollution have an impact on water circulation in the biosphere and are aims of water management.

The protection of water resources, both quantity and quality, is a global problem and it constitutes an integral element of environmental protection. One of the main factors influencing agricultural development is the optimum management of water resources for plant production and municipal purposes with consideration of environmental protection.

Wastewater treatment is probably more focused today on removing phosphorus and nitrogen than pathogens, since these elements contribute to eutrophication and deterioration of our natural water ecosystems. A great number of biological wastewater treatment techniques exist, from natural and constructed wetlands at one end to high-technology solutions based on the activated sludge process at the other end [1-4]. The future challenge to sustainable wastewater treatment is to design techniques that would recycle the content of valuable plant nutrients. The strongest effect on the quality of waters is that of biogenic substances of various origin that cause their eutrophication [5, 18]. Inorganic nitrogen occurs in water in notable amounts, in two forms that are components of its natural circulation, i.e. in the form of ammonium nitrogen and of nitrate nitrogen, an increase of which in e.g. deep waters has a negative effect on the quality of potable water.

Wastewater drainage to the ground is the oldest and the most natural form of its utilization. Studies on the elimination of biogenic compounds have been conducted for a long time [6]. One of the important methods of biogenic compound elimination is the use of wastewater treatment that utilizes soil and plants and the activity of specific microflora inhabiting the soil and roots of so-called "root-reed" treatment plants [7-13].

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During recent years it has become obvious that it is both environmentally and economically appropriate to use vegetation filters of short rotation willows (*Salix* spp.) to purify waters and soils [14]. In Poland, the development of wastewater treatment plants of the root-reed type began towards the end of the 1970s [15, 16]. Currently in Poland there are about 300 sewage treatment plants of this type in operation, while in the United States there and about 150 plants of the root-reed type in one states alone [8, 17-21].

With the application of proper irrigations techniques, certain organogenic soils can be used for the third stage of wastewater purification. Worthy of recommendation is wastewater irrigation of muck soils and dried peat soils. Experiments performed on peat soils showed very good effects of wastewater purification and good effects of meadow production [22]. Drainage of municipal wastes to the ground and their agricultural utilization is an alternative solution for the problem of protection of water reservoirs.

Determination of optimum conditions for plants, under which they can uptake water, as well as limit values of the uptake, is done by means of water retention curves for soils, i.e. characteristics defining the relation between the content of water in a soil and its potential. The curves can be used to read the character of the air-water relations in soil that have a deciding effect on the growth and development of plants. Ensuring a steady supply of water to plant roots causes the plants to transpire water through open stomatal apparatus, which intensifies the process of assimilation and uptake of nutrients from the soil, including nitrogen.

The aim of the present work was to investigate ammonium and nitrate(V) fixation by soil irrigated with communal wastewaters and estimation of the possibility of using organic soil and willow (*Salix americana*) for wastewater purification.

Table 1. Physicochemical parameters of the treated wastewaters [23].

Parameter	Unit	Range of value
pH	–	6.47-8.41
COD[#]	g $O_2 \cdot m^{-3}$	30.1-56.3
BOD_5[##]	g $O_2 \cdot m^{-3}$	8.3-22.6
$N\text{-}NH_4^+$	g $N \cdot m^{-3}$	1.1-7.1
$N\text{-}NO_3^-$	g $N \cdot m^{-3}$	20.2-38.4
N-tot	g $N \cdot m^{-3}$	22.3-43.6
$P\text{-}PO_4$	g $P \cdot m^{-3}$	3.1-6.8
P-tot	g $P \cdot m^{-3}$	3.7-7.0
Na^+	g $Na \cdot m^{-3}$	24.3-69.4
K^+	g $K \cdot m^{-3}$	11.8-27.7
Ca^{2+}	g $Ca \cdot m^{-3}$	59.7-95.2
Mg^{2+}	g $Mg \cdot m^{-3}$	12.6-19.7
SO_4^{2-}	g $SO_4 \cdot m^{-3}$	43.6-116.3
Cl^-	g $Cl \cdot m^{-3}$	67.8-121.6
Zn	mg $Zn \cdot m^{-3}$	18-800
Cu	mg $Cu \cdot m^{-3}$	6-198
Pb	mg $Pb \cdot m^{-3}$	7-96

[#]COD (chemical oxygen demand),
[##]BOD_5 (5 days biological oxygen demand).

Material and Methods

Studies on processes taking place under conditions of intensive irrigation of soil and plants with purified municipal wastewaters were conducted on an experimental object located in the Bystrzyca River valley. The object was irrigated with purified wastewaters from the city of Lublin, treated at the "Hajdów" treatment plant. Wastewaters from the particular plots of the experimental object were drained via a system of drainage pipes to a drainage ditch. The experiment included a number of plants and covered the area of 8 ha. This paper presents the results concerning willow (*Salix americana*), a species that is recommended for cultivation on stands in habitats degraded by agriculture. The willow is used in industry, among others for the production of salicylic acid and certain medicines (aspirin). Most often, however, it is used as a raw material for the production of baskets, wicker furniture, or special products. The experimental field was divided into three plots (A, B and C); plot A played the role of the control object and was not irrigated with wastewaters, plot B was irrigated with a single dose of wastewater (optimum dose), while plot C received a double dose of wastewater (double optimum dose). The plots were separated from one another by means of dykes. To prevent water infiltration from the sedimentation pools of the treatment plant and from the river waters, the object was surrounded with a belt ditch on the southwestern and southeastern sides. On the northeastern side the role of the belt ditch was played by the drainage ditch collecting drainage waters from the particular plots of the experimental field. The drainage system covered the whole area of the object, the surface of which played the draining role. Outflow from the drainage ditch was directed on to the river. Wastewaters used for irrigation were supplied by means of a pipeline to the supply ditch, then to the irrigation ditch that ran between dykes, and then, via inlets, to plots B and C.

The experimental object was located on a muck soil developed from low sedge peat. The muck layer had a depth of 40 cm, overlying peat of medium degree of warp with fresh organic elements and carbonates. Organic matter content in the muck varied from 37 to 46 g/100g, $CaCO_3$ – 35-47 g/100g, and pH in KCl was from 7.11 do 7.31. Density of the soil was low, at 0.5 (± 0.03) g cm^{-3}. The groundwater table was located at a depth of 70 cm. The water content of the soil at different water potentials was determined with the standard Richards method in low- and high-pressure chambers (Soil Moisture Equipment Comp., Santa Barbara, CA, USA). Water conductivity in the saturated zone of the soil was measured by means of a permeameter (Eijkelkamp Comp., The Netherlands), whereas unsaturated conductivity of the soil was measured using a TDR-meter (Easy Test Comp., Lublin, Poland).

In the course of study the quality of communal wastewaters used for irrigation was controlled. The wastewaters were characterized by low variability of physicochemical parameters, periodically slightly exceeding the content of eutrophic substances, yet fully suitable for soil irrigation within the framework of the experiment. Within the period of analyses the concentration of ammonium ion varied within the range from 1.56 to 10.8 mg NH_4^+-N L^{-1}; that of nitrate (V) ion from 14.6 to 23.2 mg NO_3^- - N L^{-1}. Detailed characteristics of the wastewaters used for irrigation are given in Table 1 [23].

For irrigation the wastewaters were applied in suitable doses, i.e. the full single dose was 900 mm, the double dose was 1800 mm, and the number of doses applied was 12 (flooded 4 times in the spring, summer and autumn). Purified wastewaters from the "Hajdów" treatment plant can supply biogenic substances in amounts corresponding to intensive fertilization of soil; a dose of 600 mm supplies the soil with at least 180 kg N/ha, and a double dose, of 1200 mm, double that amount of nitrogen. Presented results are the average value from 4 floodings.

For calculation of hourly fixation of analyzed nitrogen ions (mg L^{-1} h^{-1}) the difference between the initial concentration and that after the first 24-hour period was adopted and then divided by 24. For calculations the first day from the moment of introducing wastewaters was adopted, as the greatest drop in the concentration of the analyzed ions was observed during the first 24 hours of the experiment.

The index of efficiency of wastewater purification (purification effect) by the studied soil and plants was calculated as the difference in the concentration of the analyzed ion between the initial concentration and the final concentration (148h), expressed as a percentage of the initial value. For comparison, the same method was applied for the calculation of the index of efficiency for the initial 24 hours from the moment of wastewaters introduced in the soil.

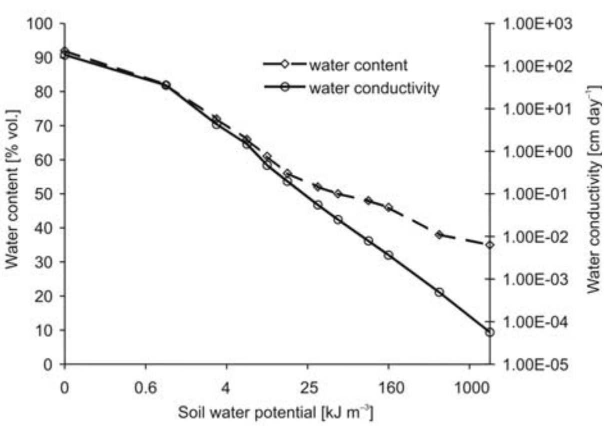

Fig. 1. Water content and water conductivity at different soil water potential.

The linear (y=a+bx) and exponential ($y=e^{a+bx}$) models were used in regression analysis and in each case the model with highest R^2 was selected as the best fit for the experimental data.

Results

Hydrophysical Characteristics of the Soil

The hydrophysical characteristics of the soil on which the experiment was performed are presented in Fig. 1. The shape of the water retention curve, i.e. the relation between water content in the soil and its potential, indicates that the studied soil is capable of holding notable amounts of water of all categories. The measured total porosity of this muck soil is about 91% vol., i.e. cm^3 cm^{-3}. The amount of free water, i.e. water subjected to the effect of gravity, is about

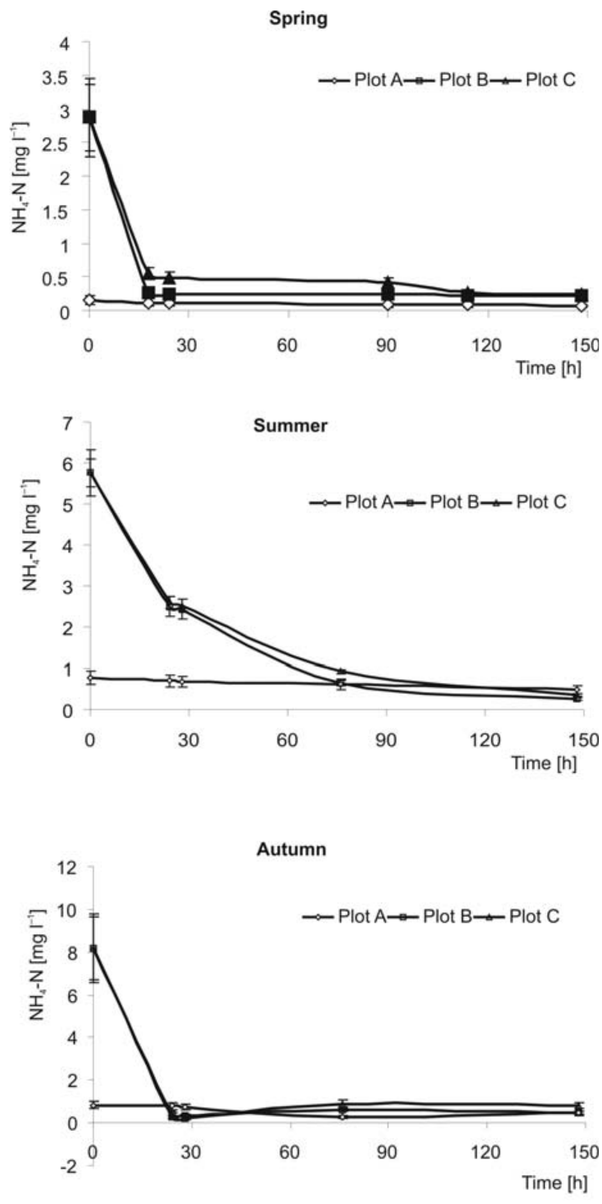

Fig. 2. Concentration of NH_4^+-N (mgL^{-1}) in drainage waters as a function of time and irrigation dose.

25% vol., that of capillary water, 66 % vol. The big part of capillary water – 35% – is water strongly bound to the soil. This part of water does not move in the soil and it is practically unavailable for plants. At full saturation of the soil with water, and thus with the wastewaters, outflow occurs at the rate of 182 cm day^{-1}. When the water ground level goes down to 30 cm, i.e. water is bound to soil with the higher potential, water can filtrate at the rate of 4.33 cm day^{-1}, and with the ground water table at 70 cm the rate of filtration, i.e. unsaturated conductivity coefficient, equals 1 cm day^{-1}. Even at considerable drying of the studied soil, when it contains only water bound with soil with potential higher than 1,500 kJm^{-3}, the cofficient (K) is relatively high - 0.01 cm day^{-1}.

Concentration of Ammonium Ion in Drainage Waters

Ammonium ion concentration in the drainage waters from plots planted with willow, flooded in the spring, summer and autumn with the single or double doses of wastewaters, is given in Fig. 2. Additionally, the Figure shows the concentration of NH_4^+ ions in drainage waters from the control object, where the concentration of the analyzed ion only slightly varied in time. In all analyzed cases the concentration of ammonium ion in drainage waters decreased to the value for the control plot, irrespective of the irrigation dose and the time of its application. This indicates high sorptive capacity of the studied soil and of willow with relation to the NH_4^+ ion. The time required for ammonium ion concentration to be reduced to a level similar to that of the control plot varied within the range from 18 to 76 hours and did not depend on the initial concentration level and the wastewater dose applied. Analysis of ammonium ion concentration in the function of time showed, in a few cases, significant correlations where the determination coefficient varied from R^2=0.57 ($P<0.05$) to R^2=0.97 ($P<0.001$).

Considering the diurnal reduction in the level of the ammonium ion in the wastewaters used for irrigation of

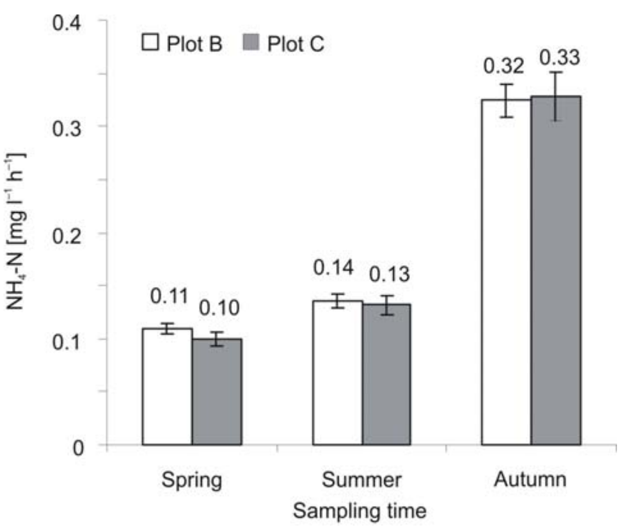

Fig. 3. Reduction of NH_4^+-N (mgL^{-1}h^{-1}) in drainage waters with relation to irrigation dose and season of the year for the first 24 hours of the experiment.

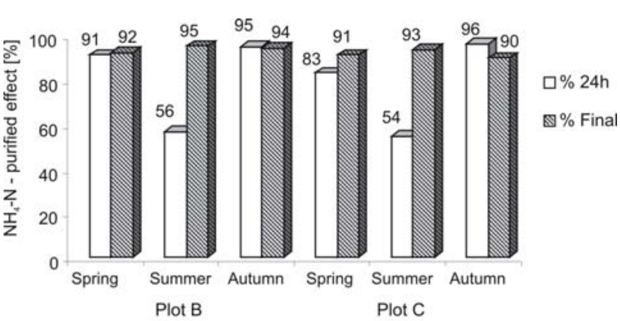

Fig. 4. Effect of wastewaters purification for ammonium ion (%) during the first day and at the end of the experiment.

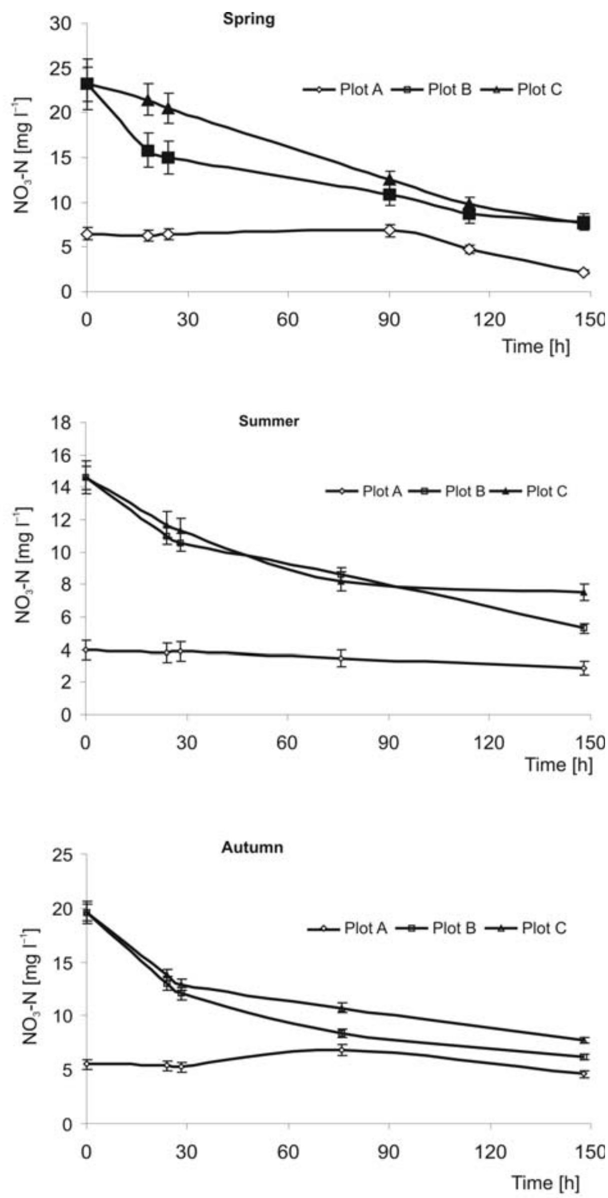

Fig. 5. Concentration of NO_3^--N (mgL^{-1}) in drainage waters as a function of time and irrigation dose.

plots B and C for the first 24 hours from the moment of introduction of the wastewaters, it varied from 0.10 to 0.33 mg NH_4^+-N L^{-1} h^{-1} (Fig. 3). The greatest reduction in the level of ammonium ion during the first day from the moment of flooding was observed in the autumn period, and the lowest in spring. Utilization of the single dose of wastewaters (Plot B) was slightly more effective compared to the double dose (Plot C).

From the viewpoint of groundwater protection, it was of interest to determine the purified effect by the studied soil and the plant, especially after the first 24 hours from the moment of the wastewater application, as in this case time is also a significant factor in the aspect of environment protection. Analyzing the percentage of absorbed ammonium ion, one can find that the studied object had a very high capacity for reducing the concentration of NH_4^+ ions in the wastewaters. That percentage varied from 56 to 95% and from 92 to 95%, respectively, for the first day from the moment of wastewater introduction and the last day for plot B, and from 54 to 96% and from 91 to 93% for analogous times of analyses for plot C with relation to the dose introduced in the soil with the wastewaters. Worthy of emphasis is the very high efficiency of wastewater purification by the studied object already after 24 hours from the moment of wastewater application, comparable at most times of analysis with the final effect (Fig. 4).

Concentration of Nitrate(V) Ion in Drainage Waters

Nitrate(V) ion concentration in drainage waters from plots planted with willow, flooded in the spring, summer and autumn seasons with the single and double doses of wastewaters is presented in Fig. 5. Additionally, the Figure shows the concentration of NO_3^- ion in drainage waters from the control plot, where the concentration of the ion in question varied only slightly within the period of analyses. While analyzing the reduction in the level of nitrate(V) ion in drainage waters, an extension of the process in time was observed as compared to ammonium ion. Therefore, the concentration of nitrate(V) ion in the function of time showed a very high coefficient of determination (varying from R^2=0.87, P<0.01 to R^2= 0.99, P<0.001) for both the single and the double irrigation doses. The studied soil showed a somewhat higher activity in transformations of NO_3^- ion in the first 24-hour period from irrigation, especially in the case of the single dose of wastewaters. The time after which the soil attained NO_3^- concentration in drainage waters similar to that in control was also extended in relation to the ammonium ion and varied depending on the season of the year.

Considering the diurnal reduction in the level of NO_3^- ion in the wastewaters for the first 24 hours from the moment of their introduction (Fig. 6), a notable effect of the season of the year and of the irrigation dose applied on the rate of NO_3^- immobilization was observed. The highest rate of NO_3^- reduction in the plot with the single dose was recorded in the spring season, and the lowest in the summer. In the case of the double irrigation dose the highest rate of nitrate(V) ion reduction was observed in autumn, while in other seasons of analyses it remained at comparable levels. In the spring period the strongest effect was observed of the irrigation dose on the rate of immobilization of nitrate(V) ion. In all analyzed cases a higher rate of NO_3^- reduction was observed for the single irrigation dose.

Considering the reduction in the level of nitrate(V) ion (purified effect) (Fig. 7), a much lower effect of wastewater purification was observed in comparison to the ammonium ion. The percentage of immobilization of the nitrate(V) ion varied from 24 to 35% and from 63 to 68%, respectively, for the first day from the moment of wastewater introduction, and the last day for plot B, and from 11 to 29% and from 48 to 66% for analogous times of analyses for plot C with relation to the dose introduced in the soil with the wastewaters. In general it can be stated that NO_3^- ion immobilization was higher in autumn, and lower in summer and in spring. A higher purified effect of the wastewaters was observed with the application of the single dose, both in the case of immobilization during the first day from the wastewaters application and in the final effect.

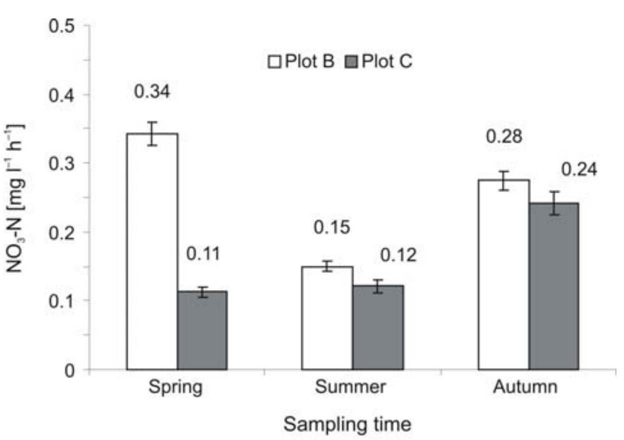

Fig. 6. Reduction of NO_3^--N ($mgL^{-1}h^{-1}$) in drainage waters with relation to irrigation dose and season of the year for the first 24 hours of the experiment.

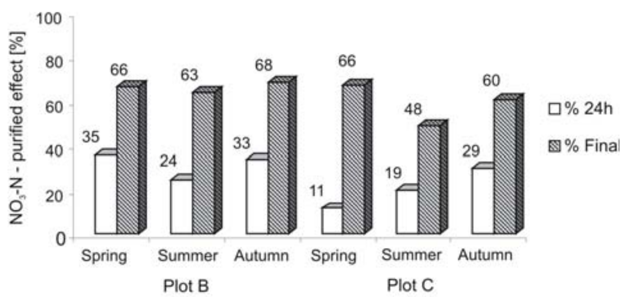

Fig. 7. Effect of wastewater purification for nitrate(V) ion (%) during the first day and at the end of the experiment.

Discussion

The static and hydraulic water properties cause the dynamics of soil waters in the experimental object to be favourable and they are in agreement with the results obtained by Baranowski et al. [24], Witkowska-Walczak et al. [25], and Włodarczyk and Witkowska-Walczak [26].

The ammonium ion may be removed from the soil solution along one of the following pathways:
1) uptake by plant roots,
2) uptake by microorganisms,
3) adsorption on the surface of soil colloids,
4) chemical binding to organic substances and may be oxidized to NO_2^- and NO_3^- (nitrification).

The ammonium ion which penetrated into the soil with wastewater under investigated conditions during infiltration through the soil profile was uptaken by plant roots and by microorganisms, especially in the upper soil layer in the rhizosphere zone. Earlier studies conducted on the same object [27] showed that NH_4^+ concentration in the soil solution of the profile depended on the time of filtration with a clear downward trend in the level of its concentration. This fact is evident for ammonium ion utilization by plants and microbes and for its facility for entering into a biochemical reaction. The high effectiveness of NH_4^+ ion uptake by willow irrigated with municipal wastewater was confirmed by Michałojć's and Nurzyński's [28] investigations. They studied the experimental object in Hajdów at the same time and found higher nitrogen content in willow's leaves planted on the plots irrigated with single and double doses of wastewaters than on the control object. A study by Elowson [29] showed high nitrogen content in willow leaves and biomass. It indicated that investigated willows were well supplied with nitrogen and that the stands functioned as a vegetation filter. Simultaneously with biological ammonium ion sorption, physical sorption by soil colloids could take place, especially in the deeper mineral part of the profile.

After flooding, the oxidative conditions in the investigated soil changed drastically but in a few millimetres deep upper layer there were suitable conditions for nitrification. Some part of the ammonium ion might be oxidized to the nitrate ion. With proceeding wastewater filtration in the soil profile, the upper pores filled with the air and nitrification could develop.

Ammonium ion concentration in drainage waters from the control plot was virtually constant within the period of the experiment, which indicates a certain equilibrium between the processes of mineralization and of immobilization of the ammonium ion.

The ultimate purified effect of the wastewater, expressed in the content of nitrate(V) ion in the drainage waters, is a result of the biological activity of the soil profile over the whole extent of its infiltration path.

A much lower effect of wastewater purification observed in the case of nitrate(V) ion in comparison to the ammonium ion caused – among other things – by its mobility related with the negative charge of the NO_3^- ion. Nitrate(V) ion, being an anion, is not fixed by the sorptive complex of the soil, and thus we are dealing here mainly with its biological immobilization, which is notably slower and affects the rate of the reduction of NO_3^- in the wastewater during its migration down the soil profile. The nitrate(V) ion may be removed from the soil solution along with one of the following microbiological pathways:
1) assimilatory nitrate(V) reduction, or
2) dissimilatory nitrate(V) reduction.

Both involve the transfer of electrons to nitrogen compounds, but they differ in the ultimate fate of the reduced nitrogen atom. Assimilatory nitrate(V) reduction is the process of NO_3^- incorporation into biomass. In contrast to assimilatory reduction for dissimilatory nitrate(V) reduction, the nitrogenous compounds accept electrons in support of cellular respiration. When the dissimilative reduction produces the dinitrogen or nitrous oxide compounds, the process is termed denitrification. In case of the nitrate(V) ion under investigated conditions we can speak of its reduction due to the process of denitrification, among others. Earlier studies [30] showed a significant relationship between the redox potential and nitrogen transformation taking place in soil irrigated with wastewater after the 2nd stage of treatment. Irrigation with wastewater results in a decrease of Eh value in the whole soil profile, causing a reduction of redox potential value below the level of +200 mV, especially in lower horizons, corresponding to dissimilative reduction of nitrate(V) to the forms of N_2O and N_2. Studies by Nosalewicz et al., [31], conducted under the conditions of the experimental object in Hajdów, indicated N_2O emission up to 21.01 mg m^{-2} h^{-1} from *Salix* vegetation following irrigation with a double dose of wastewaters. The capacity of that soil for dissimilative reduction of nitrates was also demonstrated by Brzezińska [32] in a model experiment. Results obtained by Yang et al. [33] showed that soil of 0-60 cm depth is an active rhizoplane, with a strong capability to remove nitrogen. During the past two decades of integrated research in Sweden and Poland on plant nutrition, it has been demonstrated that willows have the capacity for efficient uptake both of macro and micro nutrients, which is reflected in their high productivity [14].

Conclusions

1. Total porosity of the investigated soil is about 90% vol. The amount of water subjected to the effect of gravity is about 25% vol., that of capillary water 65% vol.
2. At full saturation of the soil with water, and thus with the wastewaters, outflow occurs at the rate of 182 cm day^{-1}. When the water ground level goes down to 30 cm, water can move at the rate of 4.33 cm day^{-1}, and with the ground water table at 70 cm - unsaturated conductivity coefficient equals 1 cm day^{-1}.
3. Concentration of analyzed ions (NH_4^+ and NO_3^-) in drainage waters from the control plot varied only slightly in time.
4. Concentration of the studied ions in drainage waters dropped to the level of the control plot (NH_4^+) or almost

to control value (NO_3^-), irrespective of the irrigation dose and of the season of application.
5. An effect of the season of the year on the final purified effect was observed.
6. The studied soil and the applied plant (*Salix americana*) showed very high capacity of fixing ammonium ions (up to 96%), but a lower capacity in the case of nitrates(V) (up to 68%).
7. The highest purified effect by the studied soil and the plant was observed after the first 24 hours from the moment of wastewater application.
8. The decreasing of ammonium and nitrate(V) content in the drainage water was correlated with time filtration.
9. The single dose was utilized to a greater extent than the double dose.

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