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The Influence of Ammonia on Groundwater Quality during Wastewater Irrigation

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Abstract

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Currently, agriculture in many countries including the Czech Republic is increasingly facing the problem of drought. The lack of precipitation results in a reduced harvest, which implies added irrigation and freshwater requirements. One of the ways to overcome the scarcity of fresh water is to search for alternative sources of irrigation water. The paper deals with a water source, which has not been preferred yet, but theoretically provides a wide application - treated municipal wastewater. Under a pilot plant, several selected soils were tested, placed in 2.0 m high filtration columns. Our observation was focused on ammonia nitrogen and its gradual decline during the flow through the soil profile. Samples from the filtration columns (inflow = irrigation; outflow = drainage water) were periodically taken, while the collected data were used for calibration of the numerical model. The model was calibrated in two successive separate steps, both were compiled in HYDRUS-2D. In the first step the model was calibrated according to the measured soil water content of materials. Subsequently, a second calibration was performed using the measured seepage concentrations of ammonia. Despite certain simplifications caused by the focus only on ammonia nitrogen, the model shows very favourable results. The hydraulic model's goodness of fit (between observed vs. measured values of water content) is R^2 = 0.88 for sand, 0.76 for loam, 0.72 for sandy-loam with vegetation on surface and 0.74 for sandy-loam without vegetation. The calibrated hydraulic model for solute transport (between observed vs. measured values of NH_4^+-N concentration) showed the value of R^2 = 0.89 for sand, 0.95 for loam, 0.95 for sandy-loam with vegetation on surface and 0.92 for sandy-loam without vegetation. The model provides significant information on the dependence of decrease of ammonia pollution by the depth. Inflow concentration of ammonia on surface 17 ± 1 mg/l is reduced to the value of 2.0 mg/l at a depth of 110 cm. It is crucial for real application to maintain the hydraulic criteria - the field capacity should not be exceeded in praxis. The value of field capacity was deliberately slightly exceeded because of understanding of the situation: how the pollution proceeds below if this rule is not followed. As a result, if wastewater is applied, the groundwater level should not be at a depth of less than 1.5 m.

Keywords: field capacity; HYDRUS-2D; numerical modelling; nutrient; wastewater reuse; water supply

Within the last years it has become increasingly plain to most of us that the sufficiency of freshwater supplies is a significant problem not only in arid regions. It proves an apparent negative impact of climate change as a variability in precipitation amount and distribution together with rising temperatures. According to last "Statement on the State of the Global Climate" issued every year by the World Meteorological Organization (WMO 2017), the year 2016 was the warmest year on record: a remarkable 1.1°C above the pre-industrial period, which is at the same time 0.06°C above the previous record set in 2015. As a consequence, water scarcity and droughts are more and more frequent and already widespread across Europe. How the European Environment Agency refers, at least 11% of the population and 17% of the European territory have been struck by water scarcity (EEA 2012). It concerns not only the

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southern parts of Europe, but also other parts of Europe, where water deficiency may occur.

For this reason, it is imperative to protect fresh water supplies from pollution on the one hand and re-using for alternative water sources on the other hand. Regarding the alternative water sources, The World Health Organization has published Guidelines for the safe use of wastewater, excreta and greywater (WHO 2006). These guidelines offer a safety framework, and minimum requirements for the safe use of wastewater, excreta and greywater in agriculture and aquaculture.

However, in Europe, there are no regulations or guidelines on the European Union (EU) level for water reuse. There are only general formulations and rules in the European Parliament and Council directives, which are summarized by Alcalde Sanz and Gawlik (2014): "Article 12 of the Urban Wastewater Treatment Directive (91/271/EEC 1991) requires that treated wastewater shall be reused whenever appropriate" and "disposal routes shall minimize the adverse effects on the environment". The Nitrates Directive (91/676/EEC 1991) concerns water reuse for agricultural irrigation and for groundwater recharge with respect to the health and environmental impacts of nitrates, especially in vulnerable zones". In the Czech Republic, there is a basic guideline relating to water management, in which the possibilities of wastewater disposal are mentioned (Act No. 254/2001 Coll. on Waters). According to this norm, it is required to prevent the impact of urban wastewater reuse on surface water and groundwater quality.

The wastewater reuse in agriculture brings many benefits: it saves significant amounts of first-use water, it provides nutrients as a substitution of chemical fertilizers and reduce production costs. However, it may cause negative impacts such as soil salinization (Levy *et al.* 2014), as well as soil and groundwater pollution with heavy metals and organic compounds. The use of raw wastewater for crop production pose a threat to health due to its microbial content, especially parasites, viruses and bacteria, which produce a wide range of diseases (Blumenthal & Peasey 2002; Navaro *et al.* 2015). The use of pre-treated wastewater can be safer (Zavadil 2009; Yamrot *et al.* 2015) due to the absence of this risk.

For the aforementioned purposes, our investigation is focused on agricultural use of pre-treated wastewater and its influence on the groundwater quality. There are many mathematical models for simulating water balances in soil under irrigation. However, we need a model comprising solute transport processes in soils. A lot of analytical (e.g., CXTFIT and STANMOD) as well as numerical tools have been developed. We have chosen a HYDRUS-2D model, which follows its predecessors UNSA, SWATRE, SWMII and SWMS_2D (Šimůnek *et al.* 2008). The HYDRUS programs have various related models and tools and wide range of applications: over 850 and 550 references of HYDRUS-1D and HYDRUS (2D/3D) applications, respectively (Šimůnek *et al.* 2016) According to Dudley *et al.* (2008), the HYDRUS-2D model is the most frequently used and the most accessible simulation tool for water and solutes dynamics in soil.

The main aim of this research was to verify, that the ammonia nitrogen will not have any impact on groundwater quality. The HYDRUS-2D software was applied for numerical simulation of the transport of dissolved materials in the soil. For the proper calibration of the model, four filtration columns were built and filled with different types of soil. Subsequently, the system was loaded with the treated wastewater and the soil water content and ammonia concentration were measured.

MATERIAL AND METHODS

The field experiment was conducted on a plot within a wastewater treatment plant (850 PE) in the village of Dražovice, which is located in a rural agricultural area. The Dražovice treatment system consists of mechanical pretreatment (screens, grit trap and slotted sedimentation tank) and constructed wetland (CW), which works extensively, based on natural processes. The experimental device (Figure 1) was located next to the mechanical pretreatment and the sole source of electricity.

Wastewater for the experiment was pumped from the sedimentation tank outlet. To achieve pollutant rates in common treated wastewater 80–110 mg/l for chemical oxygen demand (COD), less than 20 mg/l of ammonia nitrogen (NH_4^+-N) and 3 mg/l of total phosphorus (P_{tot}), a small vertical flow constructed wetland (VFCW) was specially constructed for this experiment. The pump was turned on six times per day, which means that 36 l/day of wastewater from slotted sedimentation tank were supplied to the surface of the vertical flow constructed wetland. The VFCW was loaded at a constant hydraulic rate of 100 mm/day, and rainfall was not included. The overall treatment efficiency in the VFCW was 78.66 ± 5.48% for COD,

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Table 1. Statistical evaluation of inlet NH_4^+-N concentration (mg/l)

Variable	*n*	Mean	SEM	SD	Q_1	Median	Q_3
NH_4^+-N concentration in irrigation water	81	21.58	0.44	3.97	19.25	21.8	24.1
NH_4^+-N concentration after rainfall dilution	81	16.97	0.97	8.71	11.62	19.2	23.3

n – number of samples; SEM – standard error of the mean; SD –standard deviation; Q_1 – first quartile; Q_3 – third quartile

56.46 ± 18.10% for N-NH_4^+ and 28.13 ± 17.75% for P_{tot}. Treated wastewater outflowing from the VFCW was accumulated in a special tank. Samples of this water were analyzed weekly and the mean concentrations of the observed parameters were 61.86 ± 13.00 mg/l for COD, 21.58 ± 3.97 mg/l for N-NH_4^+ and 3.30 ± 0.49 mg/l for P_{tot}. Statistical evaluation of N-NH_4^+ concentrations is mentioned in Table 1.

Treated water was pumped to the surface of the experimental filtration columns. The pump was controlled by an automatic switch depending on the current moisture in filtration column L02. The detection frequency of volumetric soil water content was detected by a sensor every 2 h. If the volumetric soil water content fell below the required value, the switch turned the pump on for 1 minute. The required value of volumetric soil water content for the first term (35 days) was 35%, and after that was raised up to 40%.

The treated wastewater was applied as irrigation to the surface of four experimental 2000 mm high filtration columns made of PVC, with an inner diameter of 300 mm. The inner column surface was roughened to prevent short-circuit flow. The soil in the column reached as the height of 1670 mm, and a 30–50 mm height filtering layer of fine gravel fraction 2/4 mm and 100 mm of gravel fraction 4/8 mm were on the bottom of each column. The experimental columns were protected against rainfall and sunlight. Each experimental column was filled with different soil: column L01 with sand, column L02 with loam, column L03 with loamy sand (planted with grass) and L04 with loamy sand (planted without grass). The volume density *q0* of the soil materials was measured at the start of the experiment. Homogeneous distribution across the entire profile was assumed. Inside each column, a Virriblogger sensor (Figure 1 – No. 8) was located to obtain the volumetric soil water content. The sensors were set at a depth of 750 mm below the ground level and the soil water content was logged every 24 hours, with a measurement error of less than 1%.

The total amount of water supplied as irrigation on the surface of the experimental columns was measured by a water meter (Figure 1 – No. 4) on a weekly basis. The precipitation was measured on the experimental field plot by using an ombrometer. Measured values of rainfall were compared and adjusted to the data from the automatic meteorological station at village Bohaté Málkovice located 1.5 km away. The average hydraulic load of irrigation was 9.94 ± 5.62 mm/day, and including rainfall it was 11.20 ± 6.48 mm/day. The estimated evapotranspiration was calculated as the difference between the inflowing and outflowing water volume, but there was some observational error because the storage was not included.

Sampling of water was carried out every 7 days for 13 weeks. The samples were taken from the three parts of the experimental setup: from the slotted sedimentation tank (Figure 1 – No. 1), the storage tank (Figure 1 – No. 3) and the outflow of each experimental column (Figure 1 – No. 10). During the experiment, we carried out overall 13 observations including the sampling and analysis of water, rainfall meter readings, and water meter readings. For chemical and physical sample analyses, samples that were freshly dripped out from the columns were used.

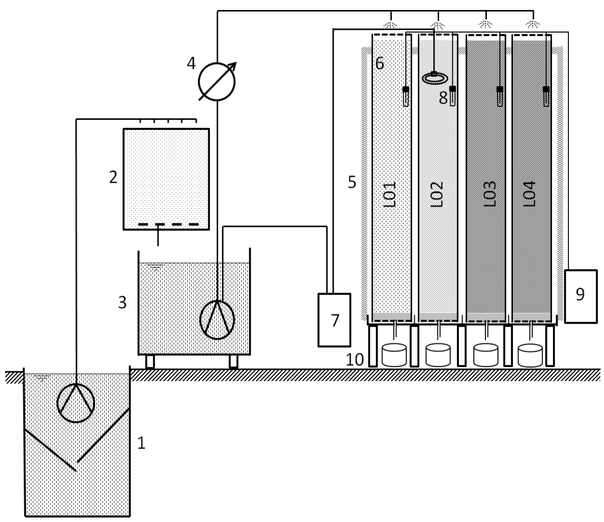

Figure 1. Scheme of the experimental column setup: 1 – slotted sedimentation tank, 2 – vertical flow constructed wetland, 3 – storage tank, 4 – water meter, 5 – experimental filtration columns (L01–L04), 6 – sensor for water content regulation, 7 – automatic moisture control, 8 – virriblogger sensor, 9 – datalogger, 10 – drip tray sampling

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One-dimensional non-equilibrium transport of NH_4^+-N was simulated by HYDRUS-2D. The geometry in HYDRUS-2D was set up and adjusted according to the real column dimensions. The hydraulic parameters (residual soil water content θ_r (m^3/m^3), saturated soil water content θ_s (m^3/m^3), coefficient in the soil water retention function a (1/m), exponent in the soil water retention function n (–), saturated hydraulic conductivity K_s (m/day), pore-connectivity parameter l (–) were optimized by inverse simulation. HYDRUS-2D uses a similar procedure to that of the single-porosity model, with the assumption that the parameters used in the single-porosity model were accurately predicted.

The seepage face boundary condition was assigned to the bottom of the flow domain and the atmospheric boundary condition was assigned to the soil surface. The atmospheric boundary condition on the surface was described using meteorological input data (i.e. amounts of rainfall and evapotranspiration), including irrigation water. The numerical model in HYDRUS-2D was created with 81 time-variable boundary conditions. For each day there was a value comprising the irrigation, precipitation and evapotranspiration. The mean inlet NH_4^+-N concentration was 16.99 ± 8.71 mg/l, including the dilution factor of precipitation.

The initial condition for water content was set as the soil water content. The value was specific for each of the materials used. For NH_4^+-N transport, the initial concentration was considered as zero in full scale.

The measured parameters were simulated using the modified form of the Richards equation (Šimůnek *et al.* 2012) for water movement in unsaturated soil under one-dimensional uniform flow.

Nonequilibrium transport of solutes is involved in the sequential first-order decay reaction. Sorption processes of ammonia (adsorption/desorption) are considered to be an instantaneous reaction between the soil solution and the soil matrix (Nakasone *et al.* 2004). Transformation processes of NH_4^+-N (nitrification, volatilization) are assumed to be first-order kinetic rate processes (Li *et al.* 2015). We are considering a strong simplification, the influence of biodegradation per nitrification bacteria, dissolved oxygen, organic carbon, volatilization to atmosphere, temperature etc., that is included in only two parameters. As reported by (Šimůnek *et al.* 2012), the first-order rate constants may be used to represent a variety of reactions or transformations, including biodegradation, volatilization, and precipitation. The mass balance equation when considering sorption and degradation is given as (Šimůnek *et al.* 2008):

$$\frac{\partial \theta c}{\partial t} + \frac{\partial \rho S}{\partial t} = \frac{\partial}{\partial z}\left(\theta D_w \frac{\partial c}{\partial z}\right) - \frac{\partial qc}{\partial z} - \mu_w \theta c$$

where:

C – NH_4^+-N concentration in the liquid phase (mg/l)
S – solute concentration in the solid phase (mg/g)
θ – volumetric water content (cm^3/cm^3)
ρ –is the dry bulk density (g/cm^3)
D_w – dispersion coefficient (cm^2/day) for the water
q – volumetric flux density (cm/day)
μ_w – the first-order rate constant for solute in the liquid phase (1/day)

The adsorption isotherm relating S is described by nonlinear equation:

$$S = \frac{k_s c^{\beta}}{1 + \eta c^{\beta}}$$

where:

k_s – adsorption isotherm coefficient for the material (l/g)
β – empirical coefficient considered as 1.0 (–)
η – empirical coefficient considered as zero (l/g)

Pearson's coefficient of determination (R^2) was used to assess the level of compliance between the predicted and observed pressure head data.

RESULTS AND DISCUSSION

Before the installation of the Virriblogger sensor, we performed sensor calibration for each material. The calibration was based on gravimetric water content measurement on disturbed soil samples, compared to real water content. Following the successful sensor calibration, these sensors were installed in the soil profile on each of the experimental lysimeters.

Figure 3 shows the measured soil water content at a 750 mm depth for all the lysimeters. The initial time for numerical simulation was the irrigation start time (14 days after the start of the experiment). The duration of experimental and also numerical model is 81 days, as data for calibration serve 81 values of water content for each lysimeter, respectively. The measured soil water content (%) are 17.0 ± 1.2, 40.0 ± 3.7, 29.6 ± 2.8 and 26.1 ± 4.3 for L01, L02, L03 and L04 respectively. The result of calibration using the observed data are soil characteristics shown in Table 2. As expected, the highest hydraulic conductivity is obtained using lymeters filled with sand (L01, K_s =8.29 × 10^3 mm/day), on the contrary the

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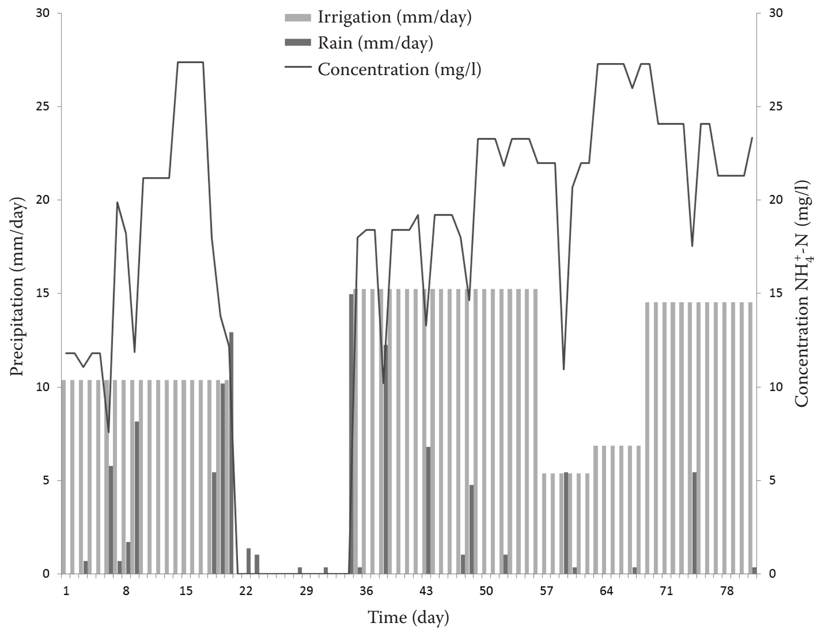

Figure 2. Boundary conditions for top layer of the model's weekly precipitation, irrigation and inlet

Table 2. Van Genuchten soil physical parameters obtained by inverse solution

Indication	Material	R^2 (–)	θ_r (mm³/mm³)	θ_s (mm³/mm³)	a (1/mm)	n (–)	K_s (mm/day)	l (–)
L01	sand	0.88	0.14	0.44	0.014	2.68	8.29×10^3	0.53
L02	loam	0.76	0.11	0.44	0.001	1.51	0.21×10^3	0.69
L03	sand:loam (1:1)	0.74	0.07	0.41	0.001	1.58	0.30×10^3	1.11
L04	sand:loam (1:1)	0.72	0.07	0.42	0.002	1.63	0.34×10^3	0.07

θ_r – residual soil water content; θ_s – saturated soil water content; a – coefficient in the soil water retention function; n – exponent in the soil water retention function; K_s – saturated hydraulic conductivity; l – pore-connectivity parameter

lowest $K_s = 0.21 \times 10^3$ mm/day is achieved in the lysimeter filled with clay (L02). When mixing sand and soil, the values are slightly higher than for clay; $K_s = 0.30 \times 10^3$ mm/day for L03 and 0.30×10^3 mm per day for L04 respectively.

The simulated data followed the observed data and showed very good compliance in all the displayed plots (Figure 4). The goodness of fit for each of the lysimeters L01–L04 are 0.88, 0.76, 0.74 and 0.72 (Table 2); this shows the strongest relationship between the observed (measured) and simulated water content. The decrease of the water content values (days 20–34) seen in Figure 4 was caused by the pump

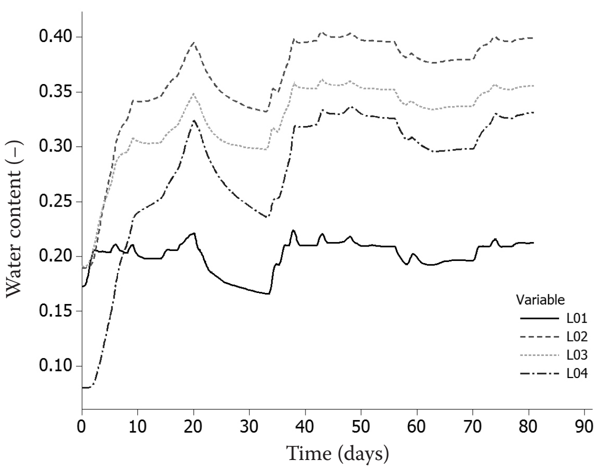

Figure 3. The time-dependent progress of the water content (%)

Table 3. Observed drainage NH_4^+-N concentration (mg/l)

Variable	n	Mean	SEM	SD	Q_1	Median	Q_3
L01	13	0.34	0.165	0.596	0.05	0.14	0.19
L02	13	0.22	0.068	0.246	0.03	0.19	0.30
L03	13	0.21	0.071	0.258	0.03	0.10	0.32
L04	13	0.70	0.193	0.697	0.17	0.44	1.28

n – number of samples; SEM – standard error of the mean; SD –standard deviation; Q_1 – first quartile; Q_3 – third quartile

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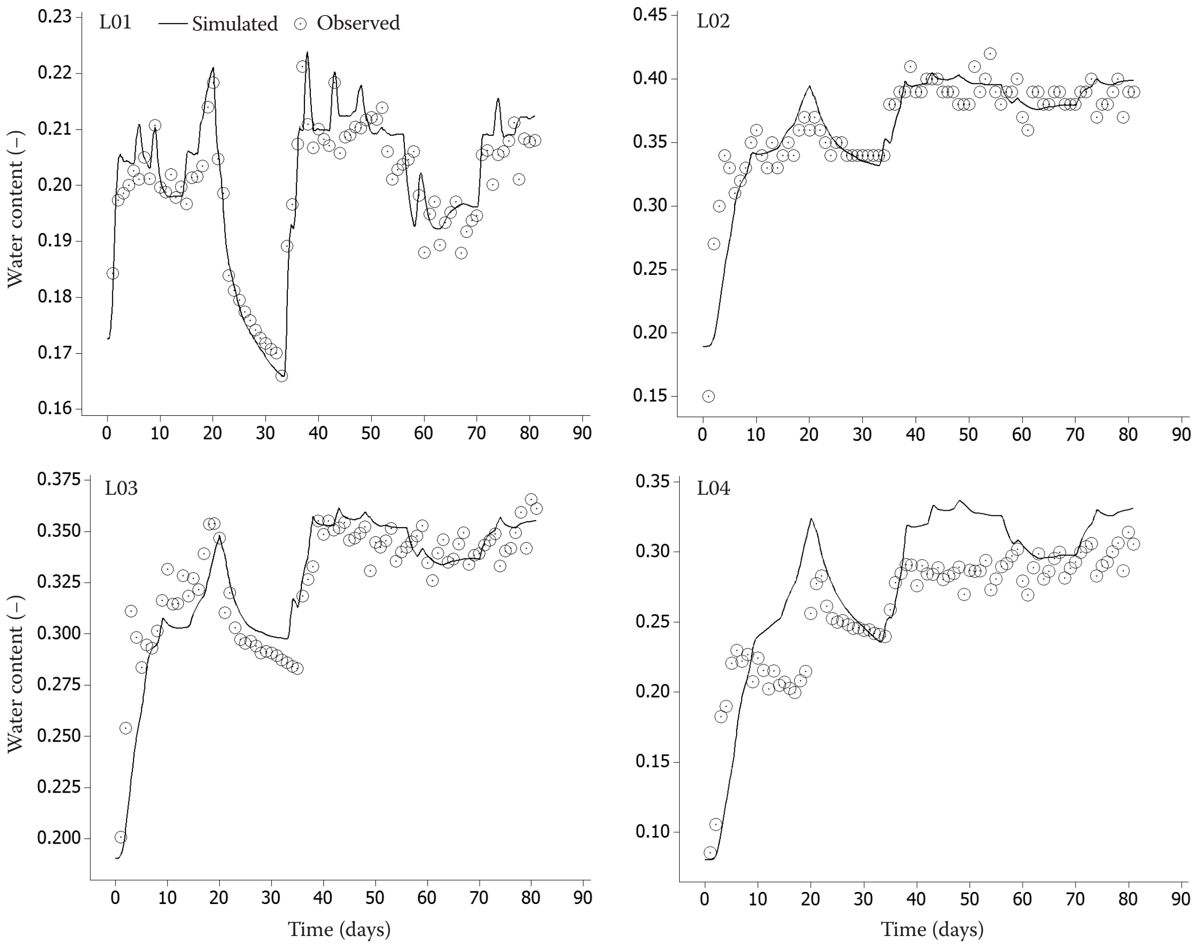

Figure 4. Observed vs simulated water content at 750 mm depth during 81 days (L01, L02 above, L03 and L04 below)

failure. The water content was stable after the pump having been repaired. Increasing values of water content in the soil are caused by rainfall (Figure 2).

After successful hydraulic model calibration, the transport model was assembled. The goal of calibrating the transport model was to find two parameters k_s and μ_w describing the loss of NH_4^+-N in the soil matrix. For transport model calibration 13 measured values of NH_4^+-N concentration were used for each lysimeter. Observed data of NH_4-N (mg/l) concentration for transport model calibration are shown in Figure 5 and statistical evaluation of these values is mentioned in Table 3.

The results of the solute transport simulations showed a good match overall with the measured data (concentration of NH_4^+-N, mg/l). This could be partly explained by the good calibration of the hydraulic and transport models. The transport model performed with acceptable efficiency with R^2 = 0.89, 0.92, 0.95 and 0.92 (Figure 5).

The distribution coefficient (k_s and μ_w) for NH_4-N and for different soil types is shown in Table 4. Compared to the Li *et al.* (2015), k_s values are higher probably caused by more permeable material (silt loam vs. loam or sand).

As other results illustrate (Marković *et al.* 2015), this clearly demonstrates the importance of irrigation in the period of the year with high evapotranspiration values and low rainfall. This rule is applied when irrigation with clean water is carried out. If it is necessary to irrigate using another source, it is necessary to comply with additional safety requirements.

Table 4. Solute reaction parameters (k_s, μ_w) obtained by inverse solution

Variable	R^2 (–)	k_s (l/mg)	μ_w (1/day)
L01	0.89	3.26×10^{-3}	0.136
L02	0.92	5.45×10^{-1}	0.202
L03	0.95	1.19×10^{-1}	0.330
L04	0.92	2.85×10^{-1}	0.146

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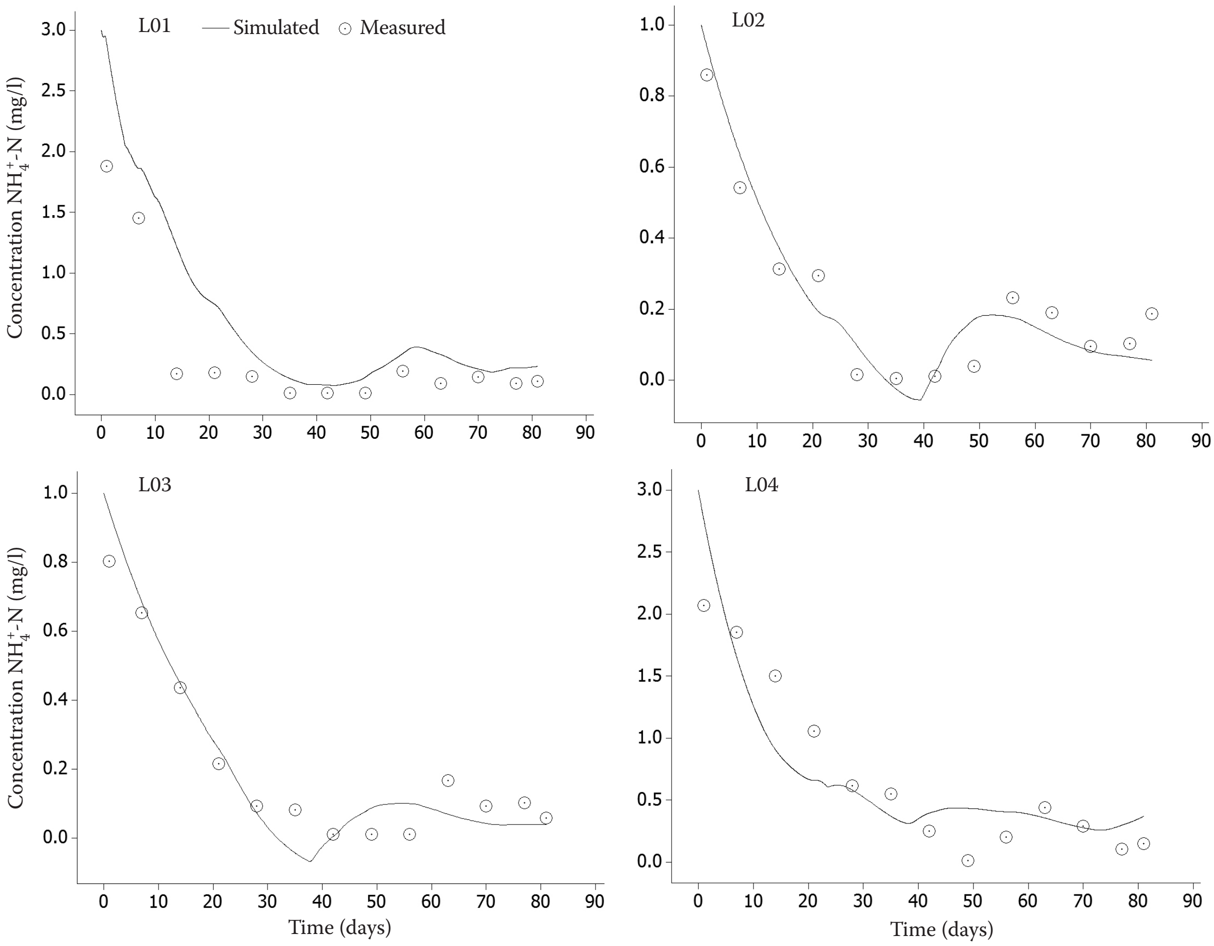

Figure 5. Observed vs simulated NH_4^+-N (mg/l) concentration at 1670 mm depth during 81 days

The main criterion for treated wastewater application (as a source for irrigation) is to avoid groundwater contamination and thus to achieve the minimum seepage into the underlying layer. Figure 3 shows the differences between water content in the different soil materials due to the installation of only one sensor for water content regulation in the lysimeter L02 (the scheme is in Figure 1). The mean of simulated outflow concentration flux in this lysimeter is 0.22 g/m^2/day (Figure 6). In the case of the same hydraulic load, lysimeter L01 (sand) shows a higher infiltration rate when the averaged specific concentration of the solute is 1.56 g/m^2/day for a depth of 1.67 m. Under the same boundary conditions, the value of specific concentration for lysimeter L03 (sandy-loam with vegetation on surface) is 0.08 g/m^2/day, and for the lysimeter L04 (sandy-loam without vegetation) it is 0.57 g/m^2/day.

Mean values of simulated outflow concentration of NH_4^+-N from calibrated model was 0.58, 0.20, 0.20 and 0.66, when compared with measured values 0.34, 0.22, 0.21 and 0.70 for L01, L02, L03 and L04 (Table. 5).

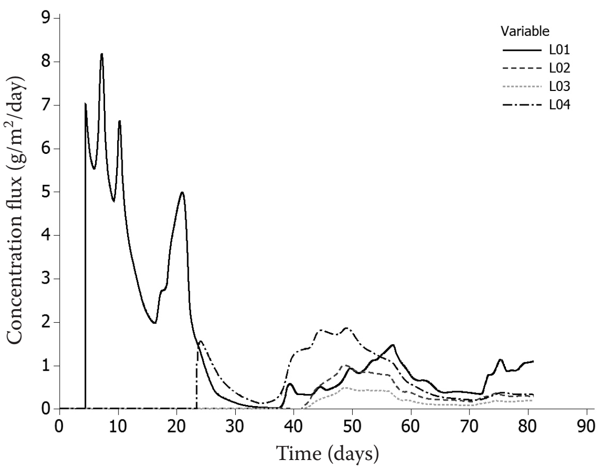

Figure 6. Simulated concentration flux of NH_4^+-N (g/m^2/day) at 1.67 m depth below the soil surface

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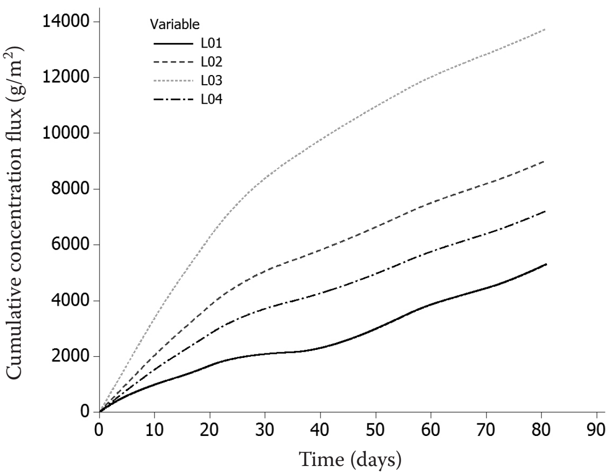

Figure 7. Simulated cumulative flux of NH_4^+-N (g/m^2) lost by transformation processes

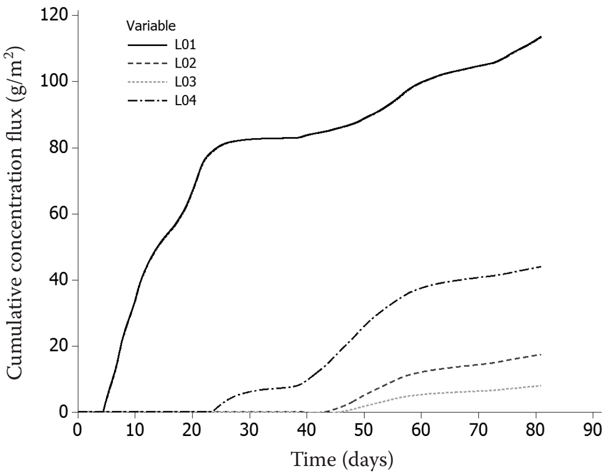

Figure 8. Simulated cumulative concentration flux of NH_4^+-N (g/m^2) at outflow of experimental lysimeters

All lysimeters without vegetation have similar results for the specific solute concentration (NH_4^+-N). Figure 7 shows cumulative flux of transformed NH_4^+-N inside each lysimeters for a period of 81 days. Final values after 81 days of wastewater infiltration are (5311.5, 9019.3, 13 740, 7212.9 g/m^2 for lysimeters L01, L02 and L04) are comparable. However, there is a noticeable difference for the lysimeter with vegetation L03, where the value is 13 740 g/m^2. In L03, the ammonia (nitrate) is absorbed by plants and therefore, more efficient nitrification is represented by a higher decomposition capability in the simulation model.

The previous conclusions are confirmed by the results in Figure 8, which shows the simulated cumulative concentration flux at outflow (the seepage face boundary condition was used at the bottom). The results of simulation for all lysimeters show expected values of cumulative concentrations flux. The best results are obtained by lysimeter L03 (with the involvement of plants), the value is 8.0 g/m^2. On the other hand, the lysimeter L01 shows high values due to the faster infiltration of waste water through sand. The value of final cumulative flux is 113.5 g/m^2. Result values for L02 and L04 are 17.4 and 43.9 g/m^2 respectively accumulated during the period of 81 days.

Table 5. Simulated concentrations of NH_4^+-N (mg/l) at 1.67 m depth below the soil surface

Variable	*n*	Mean	SEM	SD	Q_1	Median	Q_3
L01	5537	0.58	0.008	0.684	0.133	0.228	0.773
L02	1613	0.20	0.006	0.242	0.059	0.147	0.212
L03	1208	0.20	0.008	0.273	0.037	0.088	0.282
L04	3598	0.66	0.009	0.579	0.345	0.420	0.669

n – number of samples; SEM – standard error of the mean; SD –standard deviation; Q_1 – first quartile; Q_3 – third quartile

CONCLUSION

The HYDRUS-2D model using the single-porosity assumption with an included standard solute transport function was confronted with the semi-field measurement of water content using Virriblogger sensors in four irrigation columns. Column L01 was filled with sand, L02 with loam; L03 and L04 were filled with a mix of both materials (1:1), while only L03 was planted with grass as a control.

Despite a very strong simplification of the ammonia transformation process, we gained high quality results approaching the measured values. We did not consider the temperature effect since only irrigation during the vegetation seasons is assumed, where the subsoil temperatures are approximately similar.

The hydraulic model was calibrated with satisfactory R^2 values of 0.88, 0.76, 0.72 and 0.74 (single-porosity model) and 0.89, 0.95, 0.95 and 0.92 (solute transport model) for L01, L02, L03 and L04, respectively.

Our measured and simulated results show the possibility of utilizing treated wastewater for irrigation by respecting the field capacity value.

On the other hand, the use of treated wastewater as a source of irrigation water has its disadvantages, such as soil contamination by pathogenic organisms, drugs or heavy metals. Likewise, there is a danger for people working and operating with the irrigation system.

This article focuses on monitoring and simulating the ammonia nitrogen transport during irrigation using treated wastewater. The presented results il-

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lustrate that there is a minimal risk of groundwater contamination by NH_4^+-N (the depth is minimum 2 m below the ground level) if only the required amount of water is supplied to the soil.

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