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Quantification of water and sewage leakages from urban infrastructure into a shallow aquifer in East Ukraine

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Abstract

Leaky water supply and sewer mains can become unmanaged sources of urban groundwater recharge and contamination posing environmental and health risks. Stable isotopes of water and hydro-chemical tracer were applied to quantify water and sewage leakages in a shallow aquifer of a large Ukrainian city. Binary and ternary mixing models were used based on the d -excess and chloride concentrations of tap water, rural and urban groundwater to estimate fractions of natural recharge, urban seepage, volumes of water supply and sewage leakages in urban springs. Water supply leakages that recharge aquifer were ~3% ($6.5 \text{ Mm}^3 \text{ a}^{-1}$) of the total water supply and strongly correlated with failures on the water infrastructure. Sewage leakages ($1.4 \text{ Mm}^3 \text{ a}^{-1}$) to the aquifer were less in amount than water supply leakages but induced nitrate and associated contaminants pollution risk of urban groundwater. The proposed method is useful for the pilot evaluation of urban groundwater recharge and contamination and can be applied in other regions worldwide to support the decision making in water management.

Keywords: deuterium; oxygen isotope; water losses; sewer; urban groundwater; Ukraine.

Acknowledgments

The research was carried out in the framework of projects CRP F33020 “Environmental isotopes methods to assess water quality issues in rivers impacted by groundwater discharges” and CRP F33021 “Evaluation of human impacts on water balance and nutrients dynamics in the transboundary Russia/Ukraine river basin” and CRP F33024 “Isotope Techniques for the Evaluation of Water Sources for Domestic Supply in Urban Areas” partly funded by the International Atomic Energy Agency. Additional thanks to Mr. Yuriy Vergeles and Ms. Olga Reshetova for the samples collection.

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Introduction

Water shortage and contamination have been recognized as main environmental and societal threats worldwide (UN Habitat 2010). A special attention of water managers focuses on prevention of water scarcity and pollution in densely populated areas, which requires understanding of water mixing, recharge identification and distribution of contaminants in the context of urbanization and climate variability (Garcia-Fresca 2005). Groundwater is an important drinking water source which is sensitive to anthropogenic pressures, especially, in urban areas (UN Habitat 2010). In spite of some previous attempts to trace urban seepage in groundwater (e.g. Nisi et al. 2016; Penckwitt et al. 2016; Grimmeisen et al. 2017; Roehrdanz et al. 2017), understanding how aquifers are impacted by urbanization is a relatively poorly studied phenomenon, while unmanaged recharge of urban aquifers is a global problem (Houhou et al. 2010; Tubau et al. 2017; Grimmeisen et al. 2017), which needs to be emergently addressed in urban groundwater risk management.

It was confirmed that water supply and sewage leakages from damaged mains had a strong effect on urban hydrology (e.g. rising of groundwater levels and changing groundwater flow directions) (Bob et al. 2016) and groundwater quality (i.e. nitrate contamination, salinization) (Grimmeisen et al. 2017; Vystavna et al. 2017a). Leaks may occur due to the system aging, improper maintenance (Fenner 2000), stress from traffic and vibrations (Davies et al. 2001), excessive pressure, water hammer, frost loads, ground settlement, inefficient corrosion protection and other factors (Kesteloot et al. 2006). Leakages from water supply and sewage networks pose a considerable risk for urban water management resulting in a reduction of water supply and sewage networks efficiency, infiltration of contaminated water into pipes (Staufer et al. 2012), demanding water and power for pumping and treatment, soil contamination, and water saturation of soil profile that can generate structural collapses (Lai et al. 2016). Therefore, it is important to quantify urban groundwater recharge from leaky utility systems and to assess the relative contribution of individual sources and their impacts on urban water cycle (Lerner 2002).

Quantification of groundwater recharge by leaky water supply and sewer mains are generally done applying the flow modelling (Tubau et al. 2017), hydro-chemical tracers, such as chloride (Barret et al. 1999; Tubau et al. 2017), nitrate (Grimmeisen et al. 2017; Yakovlev et al. 2015), emerging contaminants (Jurado et al. 2012), and environmental isotopes (Grimmeisen et al. 2017; Nisi et al. 2016). The selection of an appropriate method is site-specific and strongly depends on quality and availability of hydrogeological and hydro-chemical data (Tubau et al. 2017). Chemical tracers are mainly appropriate to trace sewer leaks, but not the drinking water what is generally not contaminated (Barret et al. 1999).

Recently, environmental isotopes are widely applied to trace and quantify urban groundwater recharge (Nisi et al. 2016; Penckwitt et al. 2016; Grimmeisen et al. 2017). While, the application of stable isotopes of boron, nitrate and sulfate can be time consuming, expensive and requires special knowledge to interpret the data (Russow et al. 2002), stable deuterium (^2H) and oxygen-18 (^{18}O) isotopes of water molecule are conservative tracers that provide quick and economically reasonable information on water origin (Kendall et al. 2010; Asmael et al. 2015) and groundwater mixing (Penckwitt et al. 2016; Grimmeisen et al. 2017; Roehrdanz et al. 2017; Vystavna et al. 2018). When water isotopic signatures of drinking water supply and sewage leakages overlap, it does not allow quantifying individual contributions of these components (Penckwitt et al. 2016; Grimmeisen et al. 2017) and requires the use of ^2H and ^{18}O isotopes with other tracers. In many cases, chloride has been the most commonly investigated chemical indicator of sewage leakages in aquifers (Tubau et al. 2017; Roehrdanz et

al. 2017; Vystavna et al. 2017a,b) due to its relative mobility and transport with a minimal retention in the subsurface.

The objective of this study is to quantify leakages from drinking water supply and sewage networks that contribute to groundwater recharge of selected urban springs. These leakages are quantified by coupling analyzed values of deuterium (^2H) and oxygen-18 (^{18}O) isotopes and chloride concentration in tap water, sewage, surface water, urban and rural groundwater with data on water supply network failures. The results establish a framework for risk assessment processes that can help urban water managers to make more realistic and reliable decisions.

Study site

Hydrological, hydrogeological and climate settings

The study site, the Kharkiv city (1.4 million of the inhabitants) and its surrounding area (Fig. 1) is located in the temperate continental climate zone (with snowy winters and dry summers) with a 30-year average air temperature of 8.9 °C, annual precipitation amount of 512 mm and evaporation rate of 500 mm (Geological Survey 2007). The Kharkiv city is situated in the Seversky Donets water basin at the confluence of perennial Udy, Lopan and Kharkiv rivers (Fig. 1). The topography of the study area is gently sloping from north to south and is underlain by permeable and loose sedimentary deposits. The uppermost layers comprise loams, sands and clay loams of Quaternary, Pliocene and Oligocene age up to 30 m of thickness. These layers are lying over the unconfined Obukhiv aquifer which is made of fissured fine-grained sandstones substituted laterally for alluvial sands of riverine terraces (Geological Survey 2007) (Fig. 2). The water table of the Obukhiv aquifer lies from 2 to 30 m below the terrain (Fig. 2) with an estimated average hydraulic conductivity of $2.9 \times 10^{-4} \text{ m s}^{-1}$ (Geological Survey 2007). The groundwater flows towards perennial rivers forming numerous springs in flood plains and along river banks. The groundwater of the Obukhiv aquifer has a preferential recharge by snow melt and rainfall in March–April (Vystavna et al. 2018).

Water supply and sewage infrastructure

In the Kharkiv city, urban water supply and sewage infrastructure were built during the Soviet period (1960s–1980s) and were not completely renewed till now. About 97% of the total population of the Kharkiv city uses centralized drinking water supply system with a total length of 1,867 km and 76% of the population has access to centralized sewage works with a total length of 1,493 km (Ukrainian Government 2015). About 24% of population uses pit latrines and septic tanks which are drained by special trucks of the Kharkiv Municipal Water Supply and Sewage Works (KP Voda) and emptied at the Kharkiv wastewater treatment plants (WWTP). Drinking water supply network consists of pressurized pipes that are located at a depth of 0–5 m below the terrain and distributes treated portable water from the man-made water reservoir at the Seversky Donets River to domestic and industrial users (Fig. 2). Mixed industrial and domestic (15 and 85% of the total amount, respectively) wastewaters are collected by district sewer pipes that are located at the depth of 1–4 m below the terrain and discharged into deep sewage mains located at the depth of 10–40 m (Fig. 2). Wastewater is treated by mechanical and biological processes in two WWTPs with a total daily capacity of 1 Mm^3 of wastewater and discharged in Lopan and Udy Rivers (Fig. 2).

To date, the water infrastructure of the Kharkiv city has been deteriorating due to the lack of financing, labor and equipment and causing numerous leakages, which cannot be usually eliminated in a short time (Ukrainian Government 2015; KP Voda 2017). In 2016, water losses were accounted based on paid and unpaid

water use as 24% of the total water supply (KP Voda 2017). The reported by KP Voda (2017) number of failures on water supply network was 2.6 cases per 1 km of pipeline and on sewage networks it was 1.0 case per 1 km of main. According to the location of water supply pipes (Fig. 2), water leakages occur in the unsaturated and saturated soils and by gravity reach the underlying aquifer. Deep sewage mains are mostly situated within the saturated zone (Fig. 2); hence leakages from them are unlikely to recharge the shallow aquifer. Moreover, KP Voda (2017) reported the dilution and increased discharge of wastewater in sewage collectors that may be due to the infiltration of groundwater, similar to other studies (e.g. Bareš et al. 2009; Stauffer et al. 2012). Therefore, in the case of the Kharkiv city, sewage leakages can mainly derive from septic tanks, pit latrines and partly from the shallowly built district sewage systems (Ukrainian Government 2015; Vystavna et al. 2017a).

Urban runoff waters generated by rain, washing and irrigation of lawns and gardens are collected by an independent network that cover up to 90% of the city area and are directly discharged in rivers without any pre-treatment. Urban rivers and ponds are situated at elevations lower than recharge zones of groundwater-fed springs, which preclude their contamination from surface water.

Materials and methods

Water sampling and analysis

The groundwater and surface water sampling has been organized during low flow period (July–August 2016) (Vystavna et al. 2018) with the minimal precipitation rate to exclude the influence of natural recharge sources (e.g. snow melt and rain) and separate the artificial recharge from groundwater baseflow. In total, 17 urban and 11 rural groundwater sites were sampled within the Kharkiv city administrative border (Fig. 1). Surface water samples (rural SR1–SR4 and urban SU1–SU3 sites) were taken to represent the contamination status and water isotopic signature of the Severky Donets basin (Fig. 1) as a principle water source (90% of the total water supply) of the city. Simultaneously, two tap water samples (TW1 and TW2) were taken from the drinking water supply network at different locations (Fig. 1).

Rural groundwater sites (GR1–GR11) (Fig. 1) were selected to represent the natural background information on water origin (by stable isotopes of water) and contamination (by nitrate and chloride ions). Urban groundwater samples (GU1–GU17) were collected at 17 gravity-driven springs what are used for drinking and recreation purposes. The recharge zones of 17 urban springs cover 15% of the Kharkiv city area.

Groundwater temperature (T) and spring discharge were measured *in situ*. River discharge rates were obtained from the Hydrometeorological Institute of the Kharkiv city. Nitrate and chloride concentrations were analysed by the potentiometric method. The difference between two replicates was less than 5%. For analysis on stable isotopes of water, samples of surface water and groundwater were collected in 50 mL high density polyethylene (HDPE) bottles and analysed using the L2120i laser instrument (Picarro Inc.). Hydrogen and oxygen isotope analyses were calibrated against primary reference material V–SMOW (Vienna Standard Mean Ocean Water) and were reported in the δ notation in per mille (‰) deviations from the V–SMOW. Typical precisions were better than $\pm 0.1\text{‰}$ and $\pm 1.0\text{‰}$ for $\delta^{18}\text{O}$ and $\delta^2\text{H}$, respectively. The deuterium excess (*d*-excess, ‰; Dansgaard 1964) was calculated from the measured $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values as:

$$d\text{-excess} = \delta^2\text{H} - 8 \delta^{18}\text{O} (\text{‰}) \quad [1].$$

Recharge zones and mapping

The recharge zones of 17 urban springs were obtained from previous study by Yakovlev (2017), which utilized available data of terrain elevation, spring discharge, flow gradients and hydraulic conductivity of the

aquifer and deposits of the vadose zone. Shape, size and location of the recharge zones were mapped along with the data of KP Voda (2017) on water supply network failures (~1480 failures in 2017). The spatial density of registered failures was estimated by geoprocessing using the Kernel Density tool in ArcGIS Spatial Analyst (Environmental Systems Research Institute, Inc.) which calculates a magnitude-per-unit area from point features using a Kernel function to fit a smoothly tapered surface to each point.

End-member mixing analysis

Stable isotope and chloride data were applied in two- (binary) and three-component (ternary) mixing models of natural and urban recharge for the selected 17 urban springs. These mixing models are simplified cases of the end-member mixing analysis and have been widely utilized in the isotope hydrology (McGuire and McDonnell 2006; Stewart et al. 2017). A binary mixing model was used with the estimated d -excess values of distinct natural and urban recharge components as:

$$(f_1 + f_{2+3}) \times d\text{-excess} = f_1 \times d\text{-excess}_{\text{gw}} + f_{2+3} \times d\text{-excess}_{\text{rw}} \quad [2]$$

where d -excess [‰] is the estimated d -excess in groundwater of urban springs, $d\text{-excess}_{\text{gw}}$ [‰] is the average d -excess value of groundwater of rural springs representing natural recharge, $d\text{-excess}_{\text{rw}}$ [‰] is the d -excess value of tap water, and f_1 and f_{2+3} are fractions of natural (precipitation and snow melt) and unmanaged artificial recharges, respectively. The sum of f_1 and f_{2+3} is equal to 1. The potential infiltration of groundwater into the sewage mains can change the water isotopic signature of the sewage (Houhou et al. 2010). Therefore, we assume that d -excess of the tap water can be more descriptive for quantification of sewage leakages from district sewage pipes, septic tanks and pit latrines similar to previous studies (Penckwitt et al. 2016; Grimmeisen et al. 2017).

Accompanying the d -excess value, the chloride concentration was used to separate portable water and sewage leakages in urban groundwater using the ternary mixing model (adapted from Grimmeisen et al. 2017 and unpublished training materials of the International Atomic Energy Agency, IAEA):

$$(f_1 + f_2 + f_3) \times [\text{Cl}^-]_{\text{ub}} = f_1 \times [\text{Cl}^-]_{\text{gw}} + f_2 \times [\text{Cl}^-]_{\text{tw}} + f_3 \times [\text{Cl}^-]_{\text{sw}} \quad [3]$$

where Cl^-_{ub} [mg L⁻¹] is the measured chloride concentration in urban springs, Cl^-_{gw} [mg L⁻¹] is an average of measured chloride concentration in rural springs; Cl^-_{tw} [mg L⁻¹] is the measured chloride concentration in tap water; Cl^-_{sw} [mg L⁻¹] is the chloride concentration in raw wastewater. The values of f_1 , f_2 and f_3 are fractions of natural recharge, water supply and sewage leakages, respectively with the sum of f_1 , f_2 and f_3 equal to 1. Two approaches were used to estimate f_1 , f_2 and f_3 . In the first approach, the f_1 is considered as a known parameter that was simulated by the binary mixing model (Equation 2). In the second approach, the f_1 is considered as an unknown parameter and was simulated by ternary mixing model. Calculation has been carried out using Microsoft Excel and also checked using the appropriate software (Vázquez-Suñé et al. 2010).

Natural sources of Cl^- are limited within the study area (Geological Survey 2007; Vystavna et al. 2015, 2017a; Yakovlev 2017), hence Cl^- enrichment in the shallow aquifer is associated with anthropogenic sources (Kopáček et al. 2014) mainly raw wastewater that have Cl^- concentration of 350 mg L⁻¹ (Declaration 2010).

Water and sewage leakages

The simplified conceptual balance model of water supply and sewage leakages on the urban territory was used to demonstrate leakages volumes in the urban environment (Tubau et al. 2017). The synthesis of terms and parameters used in the estimation are presented in Table 1. The annual balance of water supply through the distribution network (WS, m³ a⁻¹) was formulated as:

$$\text{WS} = \text{WS}_u + \text{WU} + \text{W} \quad [4].$$

Results

Water analysis of urban and rural sites

Table 2 summarizes *in situ* measured parameters and the results of Cl^- and NO_3^- concentrations and stable isotopes analysis at rural and urban sites (see locations in Fig. 1). For urban springs, average NO_3^- concentrations are 6.6 times higher than in rural springs while average Cl^- concentrations are only 2.6 higher. Average Cl^- and NO_3^- concentrations in surface water are $67 \pm 37 \text{ mg L}^{-1}$ and $14 \pm 25 \text{ mg L}^{-1}$ (average \pm standard deviation, SD), respectively. In tap water samples, the range of Cl^- and NO_3^- concentrations is $50\text{--}60 \text{ mg L}^{-1}$ and $0.25\text{--}1 \text{ mg L}^{-1}$, respectively. From Table 2, NO_3^- concentrations of 10 urban and 2 rural springs were higher than the recommended drinking water quality standard of 50 mg L^{-1} (WHO 2011). In urban groundwater, Cl^- concentration had a significant positive correlation with NO_3^- concentration (Pearson correlation, $r=0.51$, $p<0.05$) (Supplementary Information). In rural groundwater no significant correlations were found between Cl^- and NO_3^- , also between other hydro-chemical parameters and stable water isotopes.

In Fig. 3a, stable ^{18}O and ^2H isotopes demonstrate the diverse signatures of urban and rural sites. Urban springs are more enriched in stable isotopes with average $\delta^{18}\text{O}$ of $-10.2 \pm 0.6\text{‰}$ and $\delta^2\text{H}$ of $-75.7 \pm 3.2\text{‰}$ than rural springs (average $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values were $-10.9 \pm 0.3\text{‰}$ and $-77.6 \pm 1.2\text{‰}$, respectively) (Table 2). The $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values of urban springs are plotted below the global meteoric water line (GMWL; Craig 1961) and the local meteoric water line (LMWL; Vystavna et al. 2018) (Fig. 3a) for majority of samples. However, isotopic values for most of rural groundwater samples are well plotted on the GMWL indicating natural groundwater recharge (Fig. 3a). These isotopic differences are represented by the d -excess values between urban and rural sites. The average d -excess of urban groundwater is $5.9 \pm 1.9\text{‰}$ and are much lower and spatially variable than average d -excess of rural groundwater ($9.8 \pm 1.1\text{‰}$) (Fig. 3b).

For surface water samples, the $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values of surface water and its trend line (SWL) have been below the GMWL and the LMWL (Fig. 3a). The stable isotope values with average $\delta^{18}\text{O}$ of $-7.8 \pm 0.4\text{‰}$ and average $\delta^2\text{H}$ of $-62.3 \pm 6.4\text{‰}$ are above isotope values of rural and urban groundwater sites (Table 2). Stable isotope values of two tap water samples are plotted on the SWL below the surface water sample of SU3 site, which is highly enriched with $\delta^{18}\text{O} = -5.6\text{‰}$ and $\delta^2\text{H} = -49.0\text{‰}$ (Fig. 3a). The tap water samples have the d -excess value of -1.5‰ allowing us to separate natural and urban recharge signatures.

Urban leakages in recharge zones of springs

Fig. 4 demonstrates failures of the potable water supply network in the Kharkiv city per one square km and the number of failures in the recharge zones is provided in Table 3. The recharge area of GU1, GU5–GU7, and GU14 springs has a high density of failures on the network (>10 failures per km^2). For example, the GU6 spring has the largest recharge zone of 20.1 km^2 and failures. The recharge zones of GU1 and GU14 springs have an area of 1.1 km^2 with 23 and of 3.3 km^2 with 12 failures, respectively. The GU12 and GU13 springs have the recharge zone area of 3.28 and 1.08 km^2 and the rest of springs have an area from 0.3 to 0.8 km^2 (Table 3). Number of failures on the water supply network in the recharge zones of springs significantly correlates with leakages from the water supply ($r=0.92$, $p<0.01$) and sewage ($r=0.91$, $p<0.01$) networks.

From the binary mixing model, the estimated fraction of natural recharge (f_i) in groundwater ranges from 0.33 at GU9 to 0.89 at GU10 indicating the presence of urban leakages in 17 urban spring sites (Table 3). Using the ternary mixing model, urban leakages are separated in two fractions of water supply up to 0.64 (GU9)

and of sewage up to 0.29 (GU8 and GU13) in 17 springs (Table 3, Fig. 4). Differences between pairs of natural recharge values obtained by two approaches were less than $\pm 5\%$.

Estimated volumes of urban leakages that recharge individual urban spring sites vary between each site. At the city scale, estimated water supply leakages (WU, Table 1) are $6.5 \text{ Mm}^3 \text{ a}^{-1}$ (0.66 L s^{-1}) and sewage leakages (SU, Table 1) are $1.4 \text{ Mm}^3 \text{ a}^{-1}$ (0.15 L s^{-1}) (Fig. 5). The largest volume is estimated for GU6 site with $0.62 \text{ Mm}^3 \text{ a}^{-1}$ for water supply and $0.12 \text{ Mm}^3 \text{ a}^{-1}$ for sewage (Table 3). While the GU8 and GU13 sites have the same sewage leakage fraction of 0.29, the volume of sewage leakage is much larger for GU8 spring with $0.002 \text{ Mm}^3 \text{ a}^{-1}$ compared to GU13 spring with $0.017 \text{ Mm}^3 \text{ a}^{-1}$.

Discussion

Our results show different d -excess values of tap water, rural and urban groundwater (Fig. 3a,b) (Dansgaard 1964) and these values allow us to distinguish between natural and urban recharge of the shallow aquifer in the Kharkiv city. While the d -excess values with the binary mixing model was useful to estimate natural and artificial recharge of groundwater, ternary mixing model based on the d -excess and Cl^- concentration values separates fractions of leakages from water supply and sewage networks (Table 3, Fig. 4) that can be applicable in other areas with the distinct Cl^- concentration of natural and anthropogenic sources. The selection of the d -excess value instead of previously applied $\delta^{18}\text{O}$ values (Houhou et al. 2010; Penckwitt et al. 2016; Grimmeisen et al. 2017) is considered based on the assumption that $\delta^{18}\text{O}$ values are altitude and latitude dependent including seasonal variations of climate, soil saturation and water storage conditions (Kendall et al. 2010; Rossi et al. 2015; Vystavna et al. 2018; Deb et al. 2018) and, therefore, not sufficiently representing local natural recharge conditions. From the definition, the d -excess values arise due to diverse degrees of water condensation and evaporation cycling keeping the evaporation signal of local recharge conditions (Dansgaard 1964).

The enrichment of tap water in heavier water isotopes confirms its surface water-origin with a signal of evaporative losses from the river channel and open-water reservoirs before the pre-treatment and portable water distribution (Vystavna et al. 2018). The isotopic signature of urban groundwater, the position of the urban groundwater line (GWL; Fig. 3a), low and variable d -excess values (Fig. 3b) indicated mixing at least between two water sources: (i) infiltration of local precipitation and snow melt that represent natural recharge (Vystavna et al. 2018) and (ii) water of other origin that was noticeably enriched in stable ^{18}O and ^2H isotopes. Dry weather conditions were not favorable for natural recharge in July–August (27 mm of the precipitation from 15 July to 18 August 2016, Kharkiv Meteorological station WMO ID 34300). In summer precipitation event and urban irrigation, water intensively evaporated from the impermeable surfaces and topsoil (the evaporation rate \sim the precipitation rate), is uptaken by urban plants in the soil root zones (Qin et al. 2011; Yadav et al. 2016) and collected by the urban storm runoff system. Under the given conditions, water holding capacity of the vadose zone (usually 15–40 mm per 25 cm of the top soil) was enough to accommodate the surface runoff confirming the dominance of the subsurface recharge of the aquifer in this period (Deb et al. 2018). For the Kharkiv city, recharge zones of urban springs are mapped with water supply network failures (Fig. 4) confirming that water leakages are diffusive subsurface source of urban groundwater recharge (Lerner 2002). A strong correlation between failures on water supply network and sewage leakage suggests that leaky pressurized pipes can be a path flow for sewer leaks from shallow district sewer pipes, subsurface septic tanks and pit latrines.

In the Kharkiv city, leakages from water supply network that recharge the aquifer were 3% of the drinking water supply (Fig. 5), while reported water losses were 24% of the water supply. The rest of water losses (21% of the drinking water supply) can be associated with unregistered connections to water supply network (symbol X, Fig. 5) and possible groundwater discharge locations such as perennial rivers and numerous small ponds (symbol Z, Fig. 5). Minor share of water losses also can reside in subsurface infrastructure (Vazquez-Sune et al. 2010), recharging deeper sewage mains (symbol Y, Fig. 5) and aquifers. Urban leakages as a subsurface recharge source of the aquifer were reported in other studies. For example, Grimmeisen et al. (2017) found that the fraction of urban seepage in groundwater of As-Salt city, Jordan was between 30 and 64% and network losses were 53–59% of the total water supply. Tubau et al. (2017) estimated that water and sewage leakages contributed up to 48% to the groundwater of Barcelona, Spain. Houhou et al. (2010) reported that approximately 26% of drinking water was lost through leakages in Nancy, France that contributed to the recharge of groundwater and sewer pipes. Chen et al. (2008) stated that water losses in Beijing, China were 16% of the water supply. While the detailed local balance of water supply and sewage leakages was beyond the scope of this study, our results indicate the necessity to identify the contribution of water supply leakages and groundwater to deeper sewage mains and to quantify the potential share of unregistered water and sewage connections in the Kharkiv city.

While the volume of sewage leakages (Fig. 5) was notably less than that of the water leakages, elevated chloride and nitrate concentrations (Table 2) and significant correlations between these substances indicated that leaky district sewer pipes, septic tanks and pit latrines made the groundwater quality undesirable for drinking and posed a serious health threat due the nitrate pollution risk (Table 2) and potential presence of other dangerous substances (i.e. trace metals, pharmaceuticals, persistent organic contaminants, etc.). Therefore, the attention of urban water managers should be focused to inform population about the groundwater quality of the identified springs and to improve protection of recharge zones by identifying potential contamination sources.

Obtained results on water supply and sewage leakages in Kharkiv, together with other studies on evaluation of their economic, environmental, public health and social consequences could provide important information for a risk-based management of the urban groundwater. Such a powerful tool will enable municipalities and other authorities to build long- and short-term management plans and can facilitate future planning, rehabilitation and maintenance programs. While our quantitative assessment is limited only to one season and city, isotopic, hydro-chemical and data on the state of the urban infrastructure can be used for the year-round quantitative assessment of the environmental and health risks associated with the urban pressure on the groundwater in the studied region and worldwide.

Conclusions

Coupling isotopic and hydro-chemical tracers was useful to confirm and quantify water and sewage leakages into a shallow urban aquifer discharging in springs. The stable water isotope values with the binary mixing model allowed us to calculate the fractions of natural and artificial urban recharge from leaky water infrastructures while the inclusion of chloride concentration enabled the separation of leakages from water supply and sewage networks in the three component model. Using these mixing models and data on failures of water supply network allowed demonstrating a strong link between the state of the water infrastructure and occurrence of unmanaged artificial recharge of urban aquifers. The applied methodology indicates that coupling isotopic signatures with Cl^- concentrations in tap water, sewage and rural groundwater is a useful tool to estimate

mixing of waters of different origins in urban groundwater. The simplified conceptual balance model of water and sewage leakages improved understanding and validated results on the contribution of urban seepage to the shallow aquifer. Further research should be focused on the determination of urban water residence time and reduction of urban leakages in the shallow aquifer.

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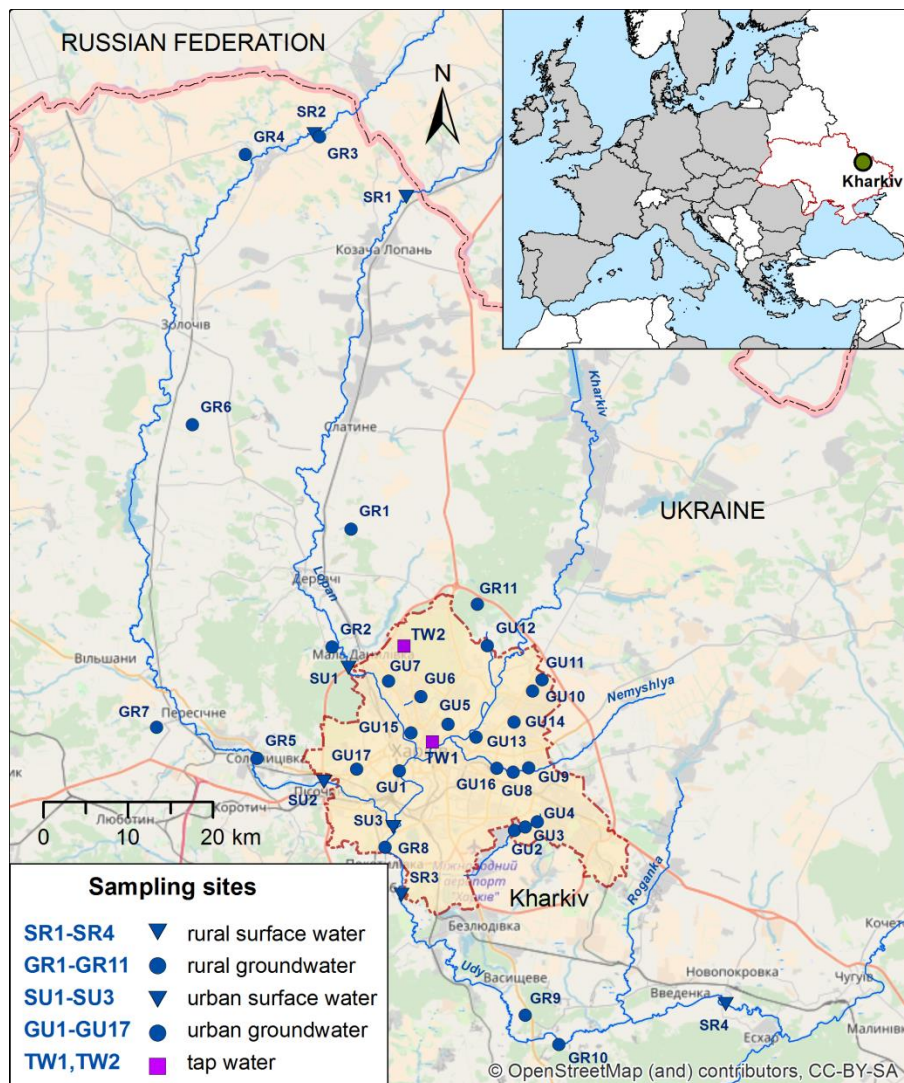
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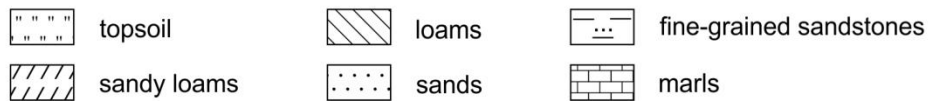
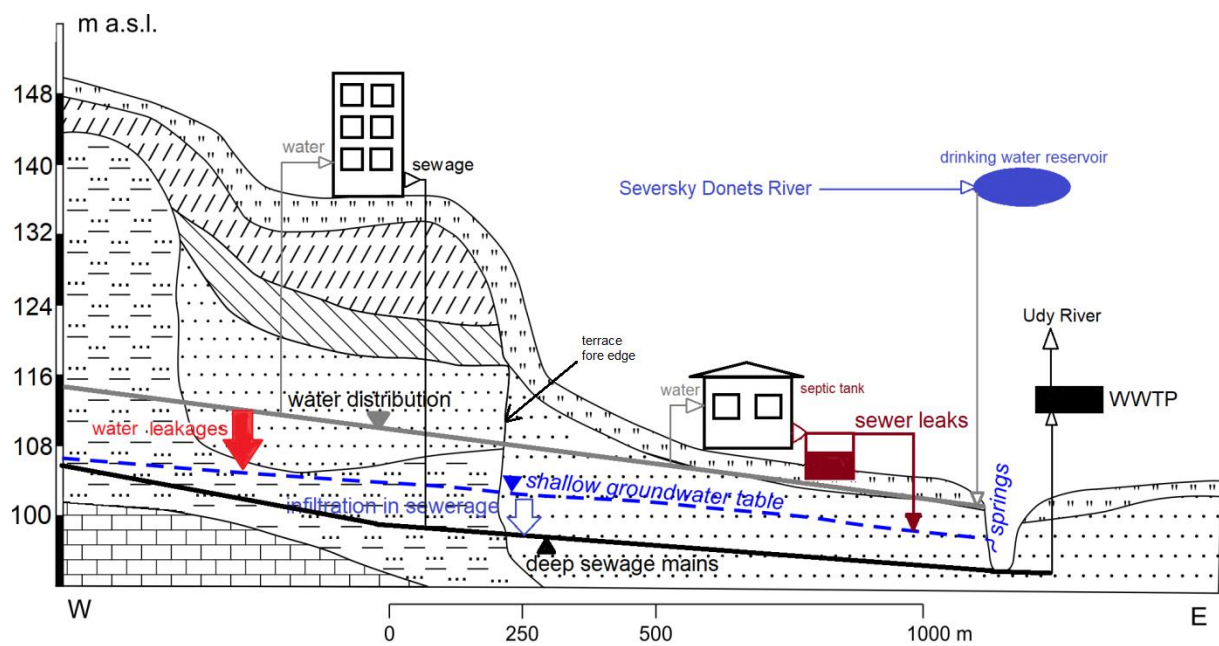
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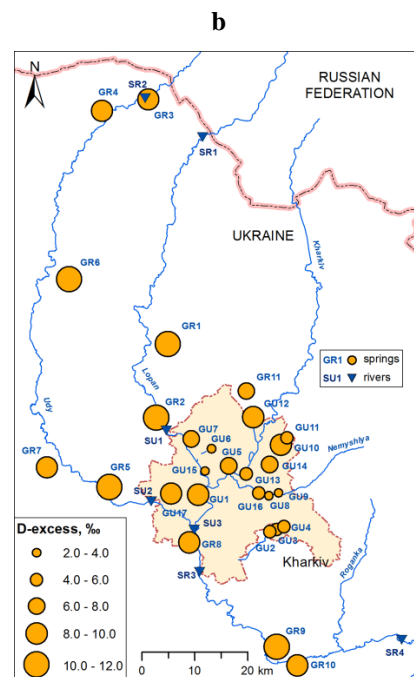
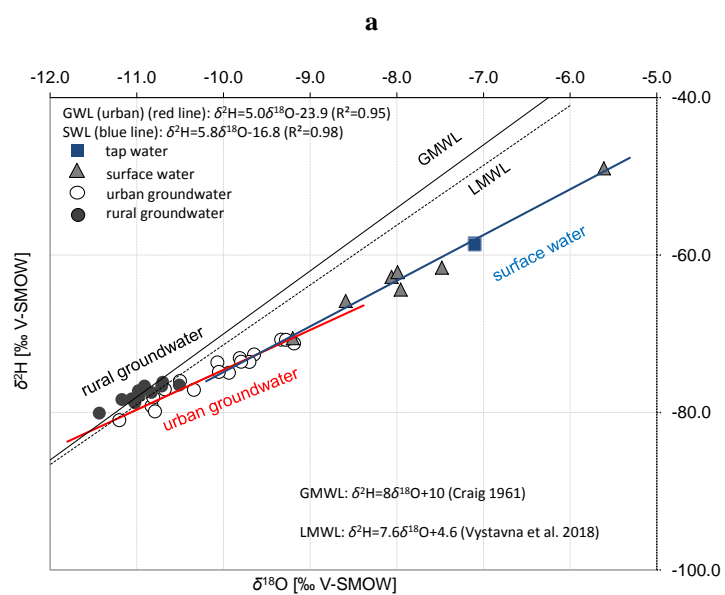
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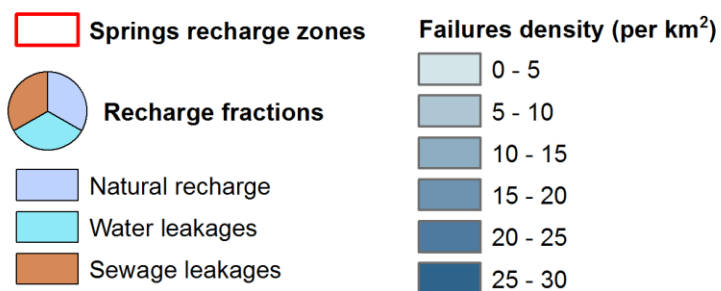
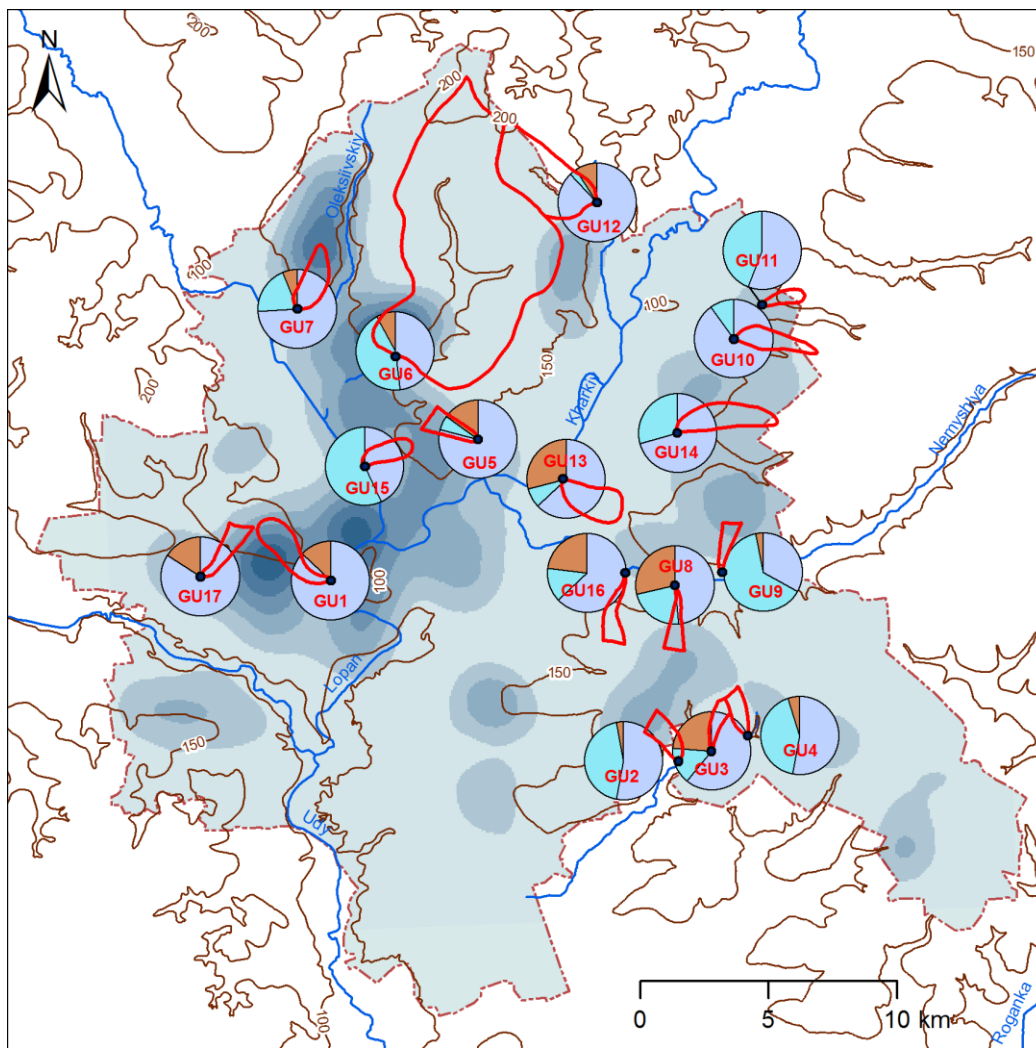
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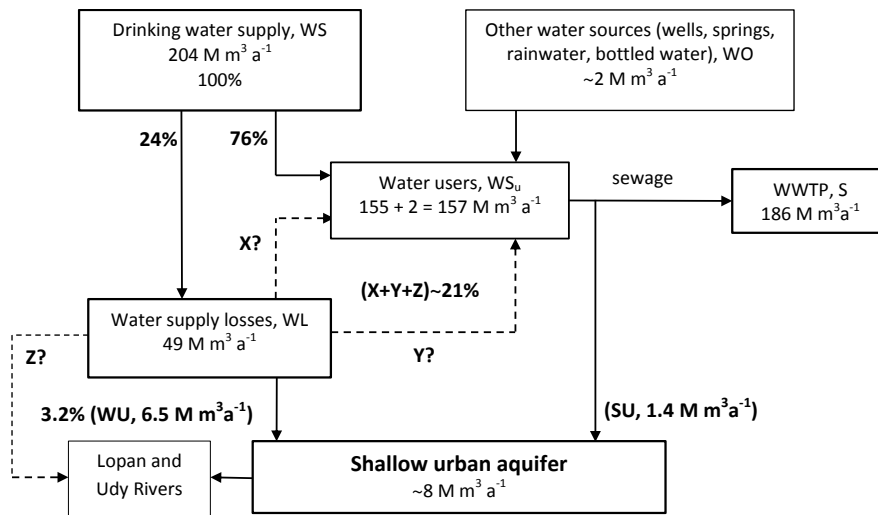
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10 Table 1. The synthesis of terms and parameters used in the estimation of volumes of water supply and sewage
11 leakages in the Kharkiv city

| Parameters | Abbreviation and unit | Description and source |
|---|--|---|
| Drinking water supply | WS, $\text{m}^3 \text{a}^{-1}$ | Water flowing through the distribution network, data obtained from KP Voda (204 Mm^3) |
| Water losses | WL, $\text{m}^3 \text{a}^{-1}$ | Estimated from the percentage of water losses reported by KP Voda (KP Voda 2017) (24% of the total water supply): $WL=0.24 \times WS$ |
| Water supplied to users | WS_u , $\text{m}^3 \text{a}^{-1}$ | Estimated as a difference between water supply (WS) and water losses (WL): $WS_u=WS-WL$ |
| Water supply network leakages that recharge the studied groundwater-driven springs | WG, $\text{m}^3 \text{a}^{-1}$ | Estimated as a sum of water supply network in all springs that was calculated based on fractions of water leakage (f_2) found by the ternary model (Equation 3) and discharge rate (Q , $\text{m}^3 \text{a}^{-1}$) for the each spring (i): $WG_i=f_2 \times Q$ $WG=\sum WG_i$ |
| Other water sources (wells, springs, rainwater, etc) | WO, $\text{m}^3 \text{a}^{-1}$ | Consumption of other sources of water by population what have no access to urban water supply network (42 thousands) (KP Voda 2017). Calculated based on water consumption rate of 110 L d^{-1} (WHO 2013). |
| Collected sewage | S, $\text{m}^3 \text{a}^{-1}$ | Sewage collected and transported on urban wastewater treatment plants (WWTP) obtained from KP Voda (186 Mm^3) |
| Sewage leakages that recharge the studied groundwater-fed springs | SG, $\text{m}^3 \text{a}^{-1}$ | Estimated as a sum of sewage leakages volume in all springs that was found by the ternary model (Equation 3) and based on the fraction of sewage leakages (f_3) and discharge rate (Q , $\text{m}^3 \text{a}^{-1}$) for the each spring (i): $SG_i=f_3 \times Q$ $SG=\sum SG_i$ |
| Specific water supply network leakages per area that recharge the shallow aquifer in the Kharkiv city | WU_o , $\text{m}^3 \text{a}^{-1}$ per km^2 of the recharge zone | Calculated as the proportion of the sum of water supply network leakages that recharge the studied springs (WG) to the sum of their recharge zone areas (A , km^2): $WU_o=WG/A$ |
| Specific sewage leakages per area that recharge shallow aquifer in the Kharkiv city | SU_o , $\text{m}^3 \text{a}^{-1}$ per km^2 of the recharge zone | Calculated as the proportion of the sum of sewage leakages that recharge the studied springs (SG) to the sum of their recharge zone areas (A , km^2): $SU_o=SG/A$ |
| Water supply network leakages that recharge shallow aquifer in the Kharkiv city | WU , $\text{m}^3 \text{a}^{-1}$ per area of the Kharkiv city | Calculated by multiplying specific water supply network leakages per area (WU_o) on the total area of the Kharkiv city (308 km^2): $WU=WU_o \times 308$ |
| Sewage leakage that recharge shallow aquifer in the Kharkiv city | SU , $\text{m}^3 \text{a}^{-1}$ per area of the Kharkiv city | Calculated by multiplying specific sewage leakages per area (SU_o) on the total area of the Kharkiv city (308 km^2): $SU=SU_o \times 308$ |
| Other water losses (illegal connections, inflow into sewage collectors, residence in the subsurface infrastructure, etc.) | W, $\text{m}^3 \text{a}^{-1}$ | Difference between total water losses (WL) and water leakages that recharge shallow aquifer (WU): $W=WL-WU$ |

14 Table 2. Stable isotopes of water and chemistry analysis of studied surface and ground waters

| Site ID | Coordinates | | Elevation, m a.s.l. | Discharge, L s ⁻¹ | T, °C | Cl ⁻ , mg L ⁻¹ | NO ₃ ⁻ , mg L ⁻¹ | δ ² H, ‰ | δ ¹⁸ O, ‰ | d-excess, ‰ |
|----------------|-------------|--------------|------------------------|---------------------------------|-------------|---|--|------------------------|-------------------------|----------------|
| | Latitude, N | Longitude, E | | | | | | | | |
| SR1 | 50.3645 | 36.2162 | 135 | 30 | 21.2 | 71 | 2 | -70.6 | -9.2 | 3.0 |
| SR2 | 50.4068 | 36.119 | 148 | 80 | 20.4 | 22 | 4 | -62.2 | -8.0 | 1.8 |
| SR3 | 49.8916 | 36.2112 | 96 | 2,600 | 22.7 | 101 | 70 | -61.6 | -7.5 | -1.8 |
| SR4 | 49.8171 | 36.5544 | 90 | 1,800 | 21.3 | 99 | 8 | -64.4 | -8.0 | -0.8 |
| SU1 | 50.0459 | 36.1549 | 107 | 200 | 19.6 | 58 | 3 | -62.8 | -8.1 | 1.7 |
| SU2 | 49.9685 | 36.129 | 99 | 30 | 16.1 | 30 | 1 | -65.8 | -8.6 | 2.9 |
| SU3 | 49.9376 | 36.2023 | 98 | 600 | 22.1 | 91 | 10 | -49.0 | -5.6 | -4.1 |
| average | - | - | 110 | 800 | 20.5 | 67 | 14 | -62.3 | -7.8 | 0.4 |
| GR1 | 50.1404 | 36.1572 | 139 | 0.40 | 10.4 | 15 | 20 | -78.4 | -11.2 | 11.0 |
| GR2 | 50.0602 | 36.1371 | 123 | 0.01 | 15.0 | 10 | 6 | -76.7 | -10.9 | 10.6 |
| GR3 | 50.4055 | 36.1239 | 152 | 0.05 | 14.0 | 18 | 10 | -76.7 | -10.7 | 9.0 |
| GR4 | 50.3933 | 36.0454 | 153 | 0.05 | 13.8 | 17 | 9 | -77.8 | -11.0 | 9.9 |
| GR5 | 49.9846 | 36.0578 | 107 | 1.00 | 12.5 | 26 | 13 | -78.3 | -11.1 | 10.2 |
| GR6 | 50.2109 | 35.9892 | 147 | 1.50 | 8.3 | 10 | 5 | -80.1 | -11.4 | 11.3 |
| GR7 | 50.0058 | 35.9515 | 138 | 1.00 | 11.0 | 21 | 6 | -76.1 | -10.7 | 9.4 |
| GR8 | 49.9243 | 36.1933 | 100 | 0.02 | 13.8 | 55 | 9 | -78.8 | -11.0 | 9.4 |
| GR9 | 49.8097 | 36.3417 | 110 | 0.03 | 13.6 | 17 | 1 | -77.2 | -11.0 | 10.6 |
| GR10 | 49.7896 | 36.3771 | 95 | 0.03 | 14.6 | 52 | 6 | -77.5 | -10.8 | 9.2 |
| GR11 | 50.0893 | 36.2908 | 141 | 8.0 | 10.0 | 20 | 1 | -76.5 | -10.5 | 7.6 |
| average | - | - | 128 | 1.10 | 12.5 | 24 | 8 | -77.6 | -10.9 | 9.8 |
| GU1 | 49.9763 | 36.2086 | 105 | 0.64 | 11.4 | 117 | 161 | -77.1 | -10.7 | 8.3 |
| GU2 | 49.9358 | 36.3303 | 137 | 0.25 | 11.4 | 43 | 57 | -75.0 | -9.9 | 4.5 |
| GU3 | 49.938 | 36.3417 | 143 | 0.25 | 11.4 | 100 | 87 | -73.1 | -9.8 | 5.4 |
| GU4 | 49.9414 | 36.3544 | 152 | 0.03 | 14.5 | 48 | 46 | -72.6 | -9.6 | 4.5 |
| GU5 | 50.008 | 36.2601 | 121 | 0.30 | 11.0 | 64 | 103 | -79.2 | -10.8 | 7.4 |
| GU6 | 50.0266 | 36.2311 | 124 | 45.0 | 11.9 | 56 | 10 | -70.8 | -9.3 | 3.9 |
| GU7 | 50.0372 | 36.1969 | 120 | 1.50 | 12.4 | 40 | 29 | -73.7 | -10.1 | 6.9 |
| GU8 | 49.9752 | 36.3289 | 120 | 0.20 | 10.6 | 119 | 49 | -73.6 | -9.7 | 4.0 |
| GU9 | 49.9781 | 36.3453 | 132 | 0.13 | 14.7 | 52 | 46 | -71.2 | -9.2 | 2.2 |
| GU10 | 50.0306 | 36.3495 | 130 | 1.25 | 13.0 | 18 | 24 | -81.0 | -11.2 | 8.6 |
| GU11 | 50.0381 | 36.3594 | 131 | 1.50 | 12.7 | 29 | 28 | -73.6 | -9.8 | 4.8 |
| GU12 | 50.0611 | 36.3017 | 121 | 0.25 | 10.0 | 43 | 33 | -78.1 | -10.8 | 8.4 |
| GU13 | 49.9991 | 36.2898 | 113 | 1.80 | 11.4 | 121 | 26 | -77.1 | -10.3 | 5.6 |
| GU14 | 50.0094 | 36.3298 | 125 | 1.80 | 11.3 | 28 | 22 | -79.9 | -10.8 | 6.4 |
| GU15 | 50.0019 | 36.2205 | 114 | 0.45 | 12.6 | 28 | 48 | -70.8 | -9.3 | 3.4 |
| GU16 | 49.978 | 36.3114 | 118 | 0.25 | 10.6 | 95 | 57 | -74.8 | -10.0 | 5.6 |
| GU17 | 49.9772 | 36.163 | 116 | 0.50 | 10.3 | 88 | 63 | -76.0 | -10.5 | 8.0 |
| average | - | - | 125 | 3.1 | 11.8 | 63 | 53 | -75.7 | -10.2 | 5.9 |
| TW 1 | 49.9958 | 36.2433 | nd | nd | 15.0 | 60 | 0.25 | -58.5 | -7.1 | -1.5 |
| TW 2 | 50.061 | 36.2136 | nd | nd | 14.0 | 50 | 1 | -58.7 | -7.1 | -1.5 |

nd means 'not determined'

17 Table 3. Recharge zones area, number of failures on water supply network, fractions of natural recharge, water
18 supply and sewage leakages in urban springs

| Site ID | Recharge zone area, A, km ² | Number of failures of water supply network in the recharge zone | Fractions by mixing models (<i>d</i> -excess and Cl ⁻)* | | | Water supply leakages, (WG), m ³ a ⁻¹ | Sewage leakages, (SG), m ³ a ⁻¹ |
|---------|--|---|--|--|------------------------------------|---|---|
| | | | Natural recharge, <i>f</i> ₁ | Urban leakages | | | |
| | | | | From portable water, <i>f</i> ₂ | From sewage, <i>f</i> ₃ | | |
| GU1 | 1.05 | 23 | 0.87 | 0.00 | 0.13 | 0 | 2644 |
| GU2 | 0.58 | 6 | 0.53 | 0.44 | 0.03 | 3438 | 260 |
| GU3 | 0.31 | 0 | 0.61 | 0.15 | 0.24 | 1152 | 1917 |
| GU4 | 0.40 | 1 | 0.53 | 0.42 | 0.05 | 396 | 47 |
| GU5 | 0.57 | 10 | 0.79 | 0.06 | 0.15 | 574 | 1435 |
| GU6 | 20.5 | 64 | 0.48 | 0.44 | 0.08 | 622152 | 118804 |
| GU7 | 0.83 | 10 | 0.74 | 0.20 | 0.06 | 9340 | 2800 |
| GU8 | 0.41 | 4 | 0.49 | 0.23 | 0.29 | 1425 | 1812 |
| GU9 | 0.31 | 6 | 0.33 | 0.64 | 0.03 | 2578 | 116 |
| GU10 | 0.73 | 1 | 0.89 | 0.10 | 0.01 | 3821 | 366 |
| GU11 | 0.28 | 1 | 0.56 | 0.44 | 0.00 | 20814 | 0 |
| GU12 | 3.28 | 0 | 0.88 | 0.03 | 0.09 | 248 | 729 |
| GU13 | 1.08 | 2 | 0.63 | 0.08 | 0.29 | 4573 | 16525 |
| GU14 | 1.25 | 12 | 0.70 | 0.29 | 0.01 | 16501 | 579 |
| GU15 | 0.53 | 4 | 0.43 | 0.57 | 0.00 | 8089 | 0 |
| GU16 | 0.48 | 1 | 0.63 | 0.14 | 0.23 | 1125 | 1806 |
| GU17 | 0.55 | 6 | 0.84 | 0.00 | 0.16 | 0 | 2523 |

19 *Presented results are from the fixed f_1 as identified by the binary mixing model.
20
21
22