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Stable isotope ($\delta^{18}O$, $\delta^{2}H$) signature of river runoff, groundwater, and precipitation in three river basins in the center of East European Plain

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Abstract. Empirical study of the isotopic features of river runoff were conducted at three hydrological posts in three different river basins: the Zakza river in the center of East European Plane (southwest of Moscow), the Dubna river (north of Moscow) and the Sosna Bystraya river in the south of central region. Samples of river water, groundwater, and precipitation for the October 2019 - October 2021 were collected at weekly intervals. At total 332 samples of river water, 275 samples of groundwater and 194 samples of precipitation were collected. Precipitation was collected as an integral sample of all precipitation fallen during the week before sampling date. For each precipitation samples, the total amount of precipitation and air temperature, weighted by precipitation amount, are given according to weather station in river basin. During the observation period, there were two completely different conditions in terms of runoff formation. First, from October 2019 to October 2020, there was an unusually low spring freshet followed by a big rain flood in July. From October 2020 – October 2021, there was a normal intra-annual flow pattern with high spring freshet. A significant supply of melted snow during spring freshet is the key factor influencing water regimes in these three river basins; varying degrees of anthropogenic flow regulation are also present. The new height frequency and complete data of stable isotope signature of river runoff component can help to study the response of a river runoff to climate change.

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1 Introduction

Recent climate change causes a shift in the hydrological system, and the isotopic signature of water, both on an annual and multi-year scale, is an essential indication of this reaction. Stable isotopes are the effective tools for a wide range of studies in hydrology, such as determining sources of flow and pathways of water across landscapes, as well as residence time (McDonnell et al., 1999; Rodgers et al., 2005), quantifying surface - atmosphere water exchanges and surface-groundwater interactions (Kirchner and Allen 2020; Sprenger et al., 2016, Négrel et al., 2003), identifying the source of groundwater recharge (Harvey and Sibray 2001; Matiatos et al., 2014) and studying runoff generation (Brown et al., 1999; Cable et al., 2011; Klaus and McDonnell, 2013). Stable isotope tracers can thus be utilized to increase our understanding of hydrological processes and the creation of quantitative water resource information. Isotope tracers may be applied for small catchments with a sensitive reaction to each precipitation event. On a wider scale, isotope markers can be used to understand how precipitation infiltrates into soil water or recharges into groundwater. Hydrograph separation performed by using the isotope data could show the impact of groundwater level on runoff. Furthermore, it aids in deriving characteristics from this relationship for years with varying climatic circumstances, i.e. quantitatively establishing the groundwater/precipitation input to runoff ratio (Klaus and McDonnell, 2013). In the last 5 years, anomalous hydrometeorological conditions have been observed on the East European Plain, including European Part of Russia (EPR). These conditions in turn were responsible for anomalous changes in the river regime. One example of these changes was the absence of a high spring freshet for rivers previously demonstrating this behavior, which was later compensated by summer rain floods. Understanding of the processes governing the isotope values of precipitation has been advanced by regional and global collection networks of precipitation data (Darling et al. 2003, Kortelainen and Karhu 2004, Katsuyama et al. 2015, Yang et al., 2020), leading to the development of global and regional scale models of precipitation isotopes (Bowen 2010, Baisden et al. 2016).

The Global Network of Isotopes in Precipitation (GNIP) and the Global Network of Isotopes in Rivers (GNIR) are the two most important global databases of isotopes in precipitation and rivers. While the data of GNIP is globally distributed, GNIR has obvious limitations in spatial coverage. Furthermore, GNIR typically comprise data sets with a monthly resolution and just a few time series with a weekly frequency. Recently, some of isotope data from rivers in Germany and Switzerland available for scientific exchange for group members in WATSON project (WATer isotopeS in the crucial zONe - from groundwater recharge to plant transpiration - https://watson-cost.eu/). The incomplete spatial coverage of GNIR network hinders the utilization of river isotopes to study global hydrological cycle. Data for locations not covered by regular observations are also needed to estimate the response of river water isotopic signature to meteorological conditions and climate change. This knowledge gap is most pronounced for the vast territories of Eurasia such as EPR.

In recent years, frequent measurements of isotope composition of oxygen and hydrogen in precipitation in Moscow has started. These precipitation data are included in the GNIP database (Vasilchuk et al., 2020, 2022). Nevertheless, for rivers that form the runoff of the largest water arteries of the EPR, measurements of stable isotopes are completely absent. The Hydrometeorological Service of Russia provides regular observations of river runoff at 2092 gauging stations. These data may be used to approximate generic runoff models; acquiring measurements of isotope properties offers up new avenues for research into the precipitation-groundwater-runoff interaction. For this purpose, 3 river basins were selected: the basin of the Sosna Bystraya River (hereinafter Sosna River) in the south part of central region, the basin of the Dubna River in the north of the Moscow region and the Zakza river in the southwest of the Moscow region. Thereby, we expanded the available isotope data with the high sampling frequency and completeness datasets.

2 Study area

The Sosna River (catchment area 16300 km^2) is located in the south of the Central Federal District of Russia, near the Lipezk and Voronezh cities. It is a tributary of the large Don River and has a large catchment area. An anthropogenic impact is expressed in pollution via surface runoff, but the anthropogenic factor does not affect the flow rate. The second river, the Dubna (catchment size 2100 km2), runs over the surface with an abundance of lakes in the Moscow region. In the upper reaches of the river there is an anthropogenic factor in the runoff regulation - a hydroelectric power station reservoir, which constantly discharges water into the river. The third river, the Zakza River, is a small river (catchment area 17 km^2) in the south of the Moscow Region on the territory of residential development. The runoff of Zakza River is regulated by the discharge of household water from a residential complex. Thus, three rivers with varying degrees of anthropogenic regulation are represented.

The Sosna is a river in the European part of Russia, a right tributary of the Don River (Figure 1). Geomorphologically, the Sosna River Basin is a plain dissected by deep river valleys, gullies and branching ravines This area is characterized by significant slopes, well-pronounced deep and lateral erosion. Bedrocks are represented by Devonian limestones. The most grandiose limestone outcrops are in the river valleys. The water-bearing rocks are represented by weak clayey limestones, slightly cavernous, in some interlayers conglomerate-like, platy. Limestones alternate with marls and argillite-like clays. The depth of the first groundwater level varies from 4.5 to 72.0 m depending on relief, 50 - 70 m prevails. The impervious locally water-bearing Evlanovsko-Livenskaya carbonate-terrigenous suite (D3 ev-lv) is developed in the northeastern and northwestern parts of the region and is confined to the upper part of the deposits of the Upper Devonian Evlanovskaya suite. The distribution of carbonate rocks, as well as the obvious karst development, provide favorable circumstances for precipitation infiltration. Most of the region's exploited Upper Devonian layers and complexes lie directly beneath permeable Quaternary sediments, ensuring aerial supply via infiltration. Vegetation is represented by broad-leaved forests, mainly in river valleys, gullies, and on watersheds and steppes. The hydrological gauge is located in Elets city. According to observation from 01/01/2005 on meteorological station in Elets city (Архив погоды в Ельце (rp5.ru)), the average monthly temperature of the warmest month (July) is 20.9˚C, the coldest month (January) is −7.1˚C. The winter in the region is moderately cold. The frost-free period lasts around 153 days, the longest was 209 days. The amount of precipitation in the region is 458 mm per year, 338 mm falls in the warm season. The maximum amount of precipitation falls in June and July.

Dubna is a river in the center of the European part of Russia, in the Vladimir and Moscow regions, the right tributary of the Volga River. Springhead located on the slopes of the Klin-Dmitrovskaya ridge, flows along the Upper Volga lowland.

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In the upper reaches, Dubna flows in a valley with steep banks, indented by the mouths of small rivers and streams. On the Upper Volga lowland, the valley becomes wider. Geomorphologically, the basin is mainly a flat lowland with occasional low moraine hills and ridges. The sediments are represented by a thick layer of lacustrine-alluvial and fluvio-glacial deposits. Typical vegetation is pine-spruce forests with a large admixture of aspen, vast expanses of arable land with small groves of small-leaved and pine forests. The hydrological gauge on the Dubna River is located in the Verbilki village. The climate of basins of the Dubna and Zakza rivers are close to the Moscow region. Long-term (1960-2020) monthly statistics on precipitation quantity and air temperature in Moscow offers yearly averages of 702 mm and 5.6°C, respectively. Average temperature (from 2005 to 2022) in January is −7.5°C and in July is 19.2°C according to nearest to Dubna river weather station (Архив погоды в Дмитрове (rp5.ru) . Half of the annual precipitation amount falls from June to October.

The Zakza River flows through a hilly, strongly rugged plain, where typical landscape is composed of arable lands and forests. The right slope of the valley is steep, sometimes up to 15-20 m high, dissected by ravines. The vegetation is represented by mixed broad-leaved forests and shrubs. The riverbed is winding, gravelly-sandy, slightly deformable. In severe winters, the river does not freeze, the ice formation is unstable, and there are many polynyas. The geological structure of sediments is represented by interbedding of upper moraine loams, on which there are supra-morainic sands of different thickness within the catchment area - from 1.5 to 7-8 m. The artesian waters of the Podolsk-Myachkovsky and Aleksinsko-Protvinsky aquifers are above 80 meters deep. The hydrological gauge is located near Bolshoye Sareevo settlement. The depth of the wells using for water supply in the Bolshoye Sareevo village's area ranging from 83 to 102 meters. Thus, we believe that the water supply of the residential complex Bolshoye Sareevo is represented by the artesian waters of the Podolsk-Myachkovsky and Aleksinsko-Protvinsky aquifers. And these waters enter the river system as wastewater.

3 Methods and results

3.1 Sampling and analytical procedures

Samples of river water, groundwater, and precipitation from September 2019 to October 2021 were collected at weekly intervals at 3 hydrological gauges (Figure. 1). River water and groundwater were sampled at 10 a.m. in the morning; precipitation samples reflect an integrated sample of all precipitation that occurred during the week preceding the sampling date. Daily precipitation samples were collected with unheated precipitation collectors O-1 (Tretyakov rain gauge) installed at a height of 2 meters from the ground. After every precipitation event, this portion was poured into a polyethylene canister for storage. During the week, each new portion of daily precipitation was added to it. At the end of the week, an integral sample from the canister was poured into a polyethylene 10 ml tube. Samples of River water were collected by hand using sampling tube along a vertical shaft, which was lowered into the water. Groundwater at Dubna river gauge at Verbilki was sampled by bucket from a water well with a depth of 10 m, located on a high river bank 10-12 m above the river edge. At Sosna (Elets) and Zakza (Bolshoye Sareevo) from a deep well (80 m depth) for water supply. The samples were not filtered, placed in polypropylene 10 ml tubes, sealed with paraffin tape to protect samples from evaporation. Samples were stored at room

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condition (25 °C and 50-70% relative humidity) until sent to the laboratory. In the laboratory samples were stored until the analysis in refrigerators with a fixed temperature and a humidity of 4 °C and 70%, respectively.

Isotope analysis was conducted at the Climate and environmental research laboratory in Arctic and Antarctic Research Institute using a Picarro L2130-i isotope analyzer. This laboratory specializes in isotope analysis of Antarctic ice cores, and its excellent measurement accuracy is certified by the IAEA's regular Inter-laboratory Comparison (WICO). The accuracy was 0.04 ‰ for the $\delta^{18}O$ value measurements and 0.5 for the $\delta^{2}H$ value measurements. The isotopic abundances of ^{18}O and ^{2}H are reported using the δ notation relative to the IAEA standard Vienna Standard Ocean Water (VSMOW) following Eq. (1):

$$\delta = \left(\frac{R_{sample}}{R_{VSMOW}} - 1\right) \times 1000\ \text{‰}\ , \qquad (1)$$

where R is the ratio of the heavier isotope relative to the lighter isotope (i.e., $^{18}O/^{16}O$ or $^{2}H/^{1}H$). We have used international and commercially available standards of the International Atomic Energy Agency IAEA (VSMOW2 and SLAP2) to validate our isotope measurements, so that isotope measurements were comparable across laboratories and instruments. Every measure sequence was accompanied by measuring two international standards and three laboratory reference standards (VOS-4 $\delta^{2}H$ = −439,7±0,3 $\delta^{18}O$ = −56,813±0,02; VSPB-2 $\delta^{2}H$ = −207,0±0,3 $\delta^{18}O$ = −26,703±0,03; SPB-2 $\delta^{2}H$ = −74,1±0,3 $\delta^{18}O$ = −9,660±0,03) every 20 to 25 samples to determine instrument drift. For measured data of every sequence a multipoint linear regression was used to normalize the measured δ-values of samples to the true δ-values in the isotope reference scale (Paul et al., 2007).

3.2 Data Records

Local precipitation in Sosna river basin has a range of $\delta^{18}O$ and $\delta^{2}H$ values from 1.99 to −18.71 ‰ and from −11 to −142.4 ‰, respectively, following a distinct seasonal pattern with high values in summer and low values in winter.

The local meteoric water line (LMWL), determined by least-squares regression is $\delta^{2}H = 6.32 \times \delta^{18}O - 12.68$, R^2 = 0,95 (Figure 2), which reflects the evaporation of rain during the summer months, due to southerly position of the basin.

For several precipitation samples collected in the summer of 2019, 2020 and 2021, extremely low values of d-excess were obtained. As a rule, negative d-excess values in precipitation are associated with sub-cloud evaporation until raindrops reach the ground. For this, the low relative humidity of the air is an important parameter. The highly negative d-excess values (lower than −10‰) have been reported for samples 28.06.2020, 6.06.2021, 13.06.2021, 4.07.2021 and 25.07.2021. These samples represented thunderstorms and strong showers precipitation events. We cannot state that this is just sub-cloud evaporation effect or intracloud evaporation associated with vertical ascending air flows in a thundercloud. Perhaps the isotope composition ($\delta^{18}O$, $\delta^{2}H$) of these samples could be changed by evaporation during storage and transportation and these samples cannot be considered as completely reliable. However, they can explain the effects of evaporation trend on $\delta^{18}O$-$\delta^{2}H$ diagram for river water as a reaction to the addition of such low d-excess precipitation to the river (see Figure 2). The $\delta^{18}O$ values of precipitation correlated with the mean weighted air temperature were as follows $\delta^{18}O = 0{,}57\ t - 13{,}17$‰, R^2 = 0,63.

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Runoff average values of $\delta^{18}O$ and $\delta^{2}H$ at Sosna gouge for the entire observation period were −10.95 ‰ (from −8.86 to −12.48 ‰) and −81.3 ‰ (from −74.5 to −91.4 ‰), respectively (Figure 3). There was no time of high spring water in the 2019-2020 hydrological year, which was caused by exceptional climatic circumstances in winter. Low $\delta^{18}O$ values of river runoff were associated with the direct input of fall and winter precipitation into the river flow. There is no direct effect of isotopically heavy precipitation on the increase in $\delta^{18}O$ values of river runoff in October 2020. Most likely, the increase in $\delta^{18}O$ values of the runoff occurred as a result of rainfall in the river basin upstream, entered the river with a time lag. In the summer of 2021, the $\delta^{18}O$ values increased due to the participation of summer rains. In 2021, a decrease in $\delta^{18}O$ of the river runoff was noted during high spring water due to the inflow of snowmelt. A linear $\delta^{18}O$-$\delta^{2}H$ regression based on isotope data of river runoff also indicate the processes of evaporation of river water (see Figure 2), or input of evaporated rain to river flow.

Groundwater average values of $\delta^{18}O$ and $\delta^{2}H$ were −11.51 ‰ (from −11.04 to −11.85 ‰) and −84.3 ‰ (from −81.13 to −86.27 ‰), respectively. These values indicate that groundwater recharge in the Sosna catchment was supplied by an increased amount of flood waters associated with the melting of snow cover, which have lower $\delta^{18}O$ values compared to the average annual $\delta^{18}O$ values. Average $\delta^{18}O$ value of precipitation fallen from January to February is equal to −15‰, and for the rest of the year it is about −8‰. Solved isotope mass balance equation provides up to 50% of the annual recharge of the aquifer with the contribution of winter precipitation. The small amplitude of $\delta^{18}O$ values of groundwater is associated with significant smoothing of the isotope signal of precipitation, which occurs when water stays in the aquifer for a long time.

Local precipitation at Dubna gouge has a range of $\delta^{18}O$ and $\delta^{2}H$ variations from −3.49 to −17.51 ‰ and from −21 to −129.5 ‰, respectively. The mean values of the $\delta^{18}O$ showed a seasonal trend with higher value in the summer and lower in the winter. The local meteoric water line (LMWL) determined by least-squares regression is $\delta^{2}H = 7.62 \times \delta^{18}O + 3.45$, $R^2 = 0,98$, which closely follows the Global Meteoric Water Line (GMWL, $\delta^{2}H = 8 \times \delta^{18}O + 10$). The relationship between $\delta^{18}O$ values of precipitation and the mean weighted air temperature is expressed as $\delta^{18}O = 0,44\ t - 13,64‰$, $R^2 = 0,53$. The seasonal runoff and groundwater isotope amplitude is damped compared to that of precipitation due to mixing with water storage in catchment.

Runoff average values of $\delta^{18}O$ and $\delta^{2}H$ at Dubna gouge for the entire observation period were −10.36 ‰ (from −8.56 to −12.88 ‰) and −78.8 ‰ (from −65.9 to −95.3 ‰), respectively. High $\delta^{18}O$ values of river runoff were noted for the beginning of summer 2020, when strong summer rains caused the flood. Low $\delta^{18}O$ values of river runoff were associated with spring high water in 2021 (Figure 4). On a long-term scale, Dubna river is characterized by a significant supply of melted snow during spring high water. In June 2020, the rain flood significantly exceeded the spring high water, which was associated with a snowless winter of 2019-2020.

Groundwater average values of $\delta^{18}O$ and $\delta^{2}H$ were −11.48 ‰ (from −9.89 to −14.53 ‰) and −86.4 ‰ (from −79.3 to −110.7 ‰), respectively. The lowest $\delta^{18}O$ value = −14.53 ‰ obtained once on 05/05/2021 represented an accidental hit of the isotope signal of snow melt into groundwater. Excluding this event, the average $\delta^{18}O$ value was −11.49 ‰. Obviously, the groundwater has lower average values of $\delta^{18}O$ and $\delta^{2}H$ than river waters. This suggests that winter/spring precipitation contributes more to groundwater recharging than yearly precipitation. The pronounced difference between the $\delta^{18}O$ values of

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river runoff and the $\delta^{18}O$ values of groundwater throughout the year (see Figure 4) indicates that the river runoff has always had an isotopically heavy component. This means that during the observation period there were no conditions when the river runoff was completely supplied by groundwater. On the contrary, in winter, the river underflow is the source of supply for the upper groundwater. Anyway, the seasonal groundwater isotope amplitude is large, which indicates a direct relationship with infiltration of precipitation without temporary retention in the aquifer. Isotope parameters of groundwater is associated with seasonality, infiltration of precipitation, and mixing with water from different source. A part of the isotope data on $\delta^{18}O$-δ^2H diagram is located along the meteoric water line. The other part of the data is described by a slope of 3.9, this linear trend may indicate the mixing with evaporated water. All groundwater isotope data are described by a linear trend $\delta^2H = 4.01 \times \delta^{18}O + 40.2$, $R^2 = 0.85$ (see Figure 2).

Local atmospheric precipitation at Zakza gouge has a range of $\delta^{18}O$ and δ^2H values from −3.15 to −28.54 ‰ and from −23.8 to −221.8 ‰, respectively, following a distinct seasonal pattern with heavier isotopes in summer and lighter isotopes in winter. The local meteoric water line (LMWL), determined by least-squares regression is $\delta^2H = 7.91 \times \delta^{18}O + 3.5$, $R^2 = 0.98$, which closely follows the Global Meteoric Water Line. The relationship between $\delta^{18}O$ values of precipitation and the mean weighted air temperature is expressed as $\delta^{18}O = 0.48\,t - 13.77$‰, $R^2 = 0.69$.

Runoff average values of $\delta^{18}O$ and δ^2H at Zakza gouge for the entire observation period were −10.76 ‰ (from −6.94 to −13.6 ‰) and −81.92 ‰ (from −59 to −101 ‰), respectively. In the 2019-2020 hydrological year, there was no period of high spring water, which was caused by unusual meteorological conditions in winter, but a big flood in June 2020 due to intensive rains (Figure 5). The high $\delta^{18}O$ values of river runoff was associated with summer rains in 2020. The low $\delta^{18}O$ value of the river runoff was noted during high spring water in 2021 caused by snowmelt. The isotope parameters of the river runoff are greatly influenced by the inflow of groundwater.

Groundwater average values of $\delta^{18}O$ and δ^2H for the entire observation period were −11.48 ‰ (from −8.61 to −12.22 ‰) and −86.3 ‰ (from −77.7 to −89.1 ‰), respectively. On the $\delta^{18}O$-δ^2H plot there is a clear difference between isotope signature of groundwater during 2019-2020 and 2021. We attribute this effect to the inflow of wastewater from the residential complex, in which new wells for water supply were put into operation. The slope on $\delta^{18}O$-δ^2H plot for groundwater in 2019-2020 (3.28) attributed to mixing with another source of water. Starting from January 2021, the $\delta^{18}O$ values of groundwater are very uniform from −11.8 to −12.22 ‰, indicating that this is water from a deep aquifer, where the water stays for a long time and in which seasonal variations of precipitation are homogenized. Against this background of such a homogeneous isotope signal of groundwater, the $\delta^{18}O$ variations of runoff, associated with the direct contribution of precipitation are more noticeable.

4 Data availability

The presented datasets are available open access via the PANGAEA repository (Chizhova et al., 2022 https://doi.pangaea.de/10.1594/PANGAEA.942291).

The river discharge is measured by the Federal State Budgetary Institution "Central Administration for Hydrometeorology and Environmental Monitoring" (FGBU "Central UGMS") (URL: httpswww.cugms.ru, accessed 20 March 2022). These data cannot be provided with our dataset due to legal restrictions; however, they can be requested free of charge from hydrology department (URL: https www.cugms.ru; Dubna river at Verbilki - #75079; Zakza river at Bol.Sareevo #75438; Sosna river at Elets #78054). Alternatively, data on the water level of Dubna and Sosna rivers at corresponding gouges are available via open access database https://allrivers.info (https://allrivers.info/gauge/dubna-verbilki?; https://allrivers.info/gauge/byistraya-sosna-elec). Meteorological information according to record on weather station (ID 27518 for Zakza river basin, ID 27419 for Dubna river basin, ID 27928 for Sosna river basin) is available via https://rp5.ru.

Author contributions

J.Ch., K.R. and M.K. designed the study, N.P., M.Kh. and V.P. collected samples of river runoff and groundwater, O.Z. developed the study of precipitation, A.E., A.K. and A.V. performed the isotope measurements, N.V., T.S. and M.Ch. provided runoff hydrological data, J.Ch. prepared the paper with contributions from all authors.

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Table 1 Geographic locations of sampling sites and duration of observation

River (Gauge)	Latitude, longitude	Basin area, km^2	Average catchment height, m	Observation period	Number of river runoff samples	Number of groundwater samples	Number of precipitation samples
Dubna (Verbilki)	56.53 N, 37.6 E.	2100	179	02.10.2019-31.10.2021	109	107	64
Sosna (Elets)	52.62 N. 38.47 E.	16300	156	01.09.2019-01.10.2021	108	54	27
Zakza (Bolshoye Sareevo)	55.71 N, 37.18 E.	17	191	03.10.2019-31.11.2021	115	114	103

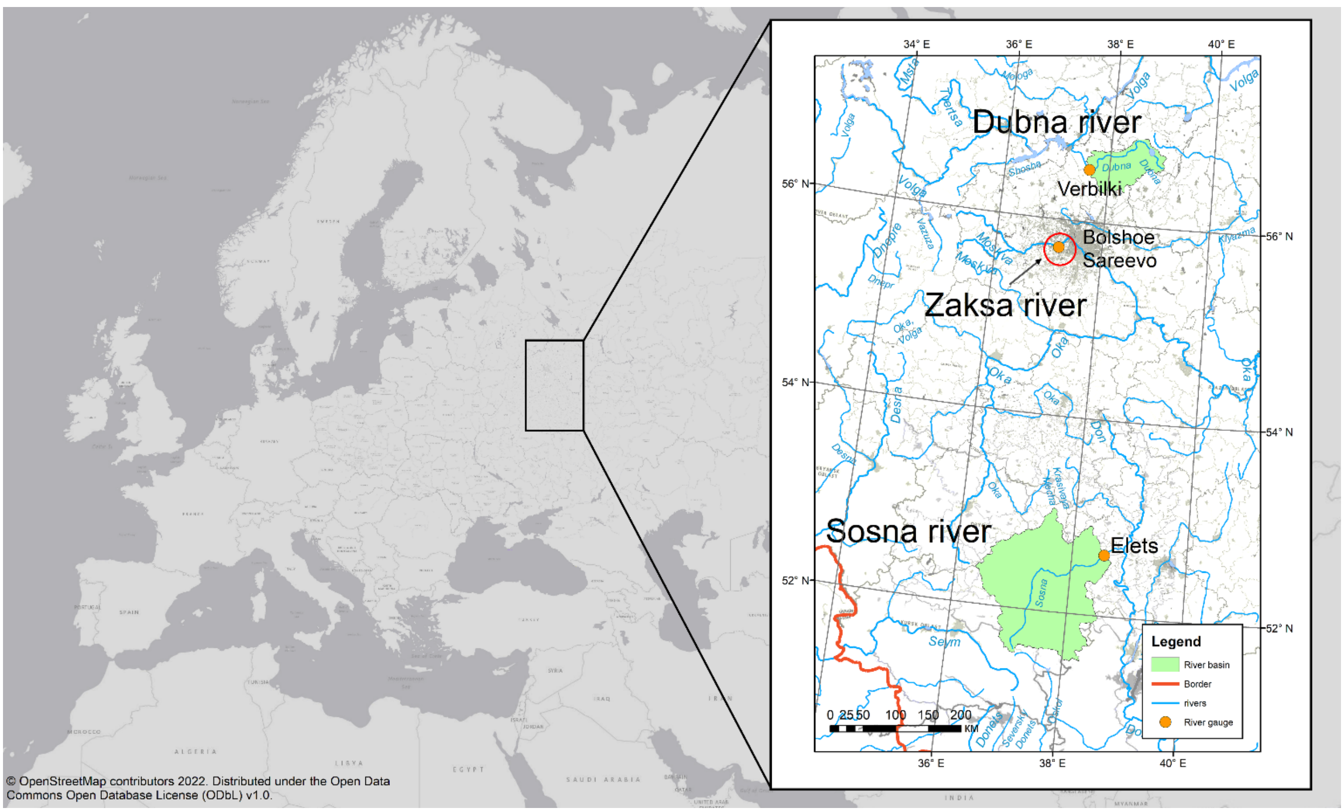

Figure 1: Location of three river basins in the center of the East European Plain.

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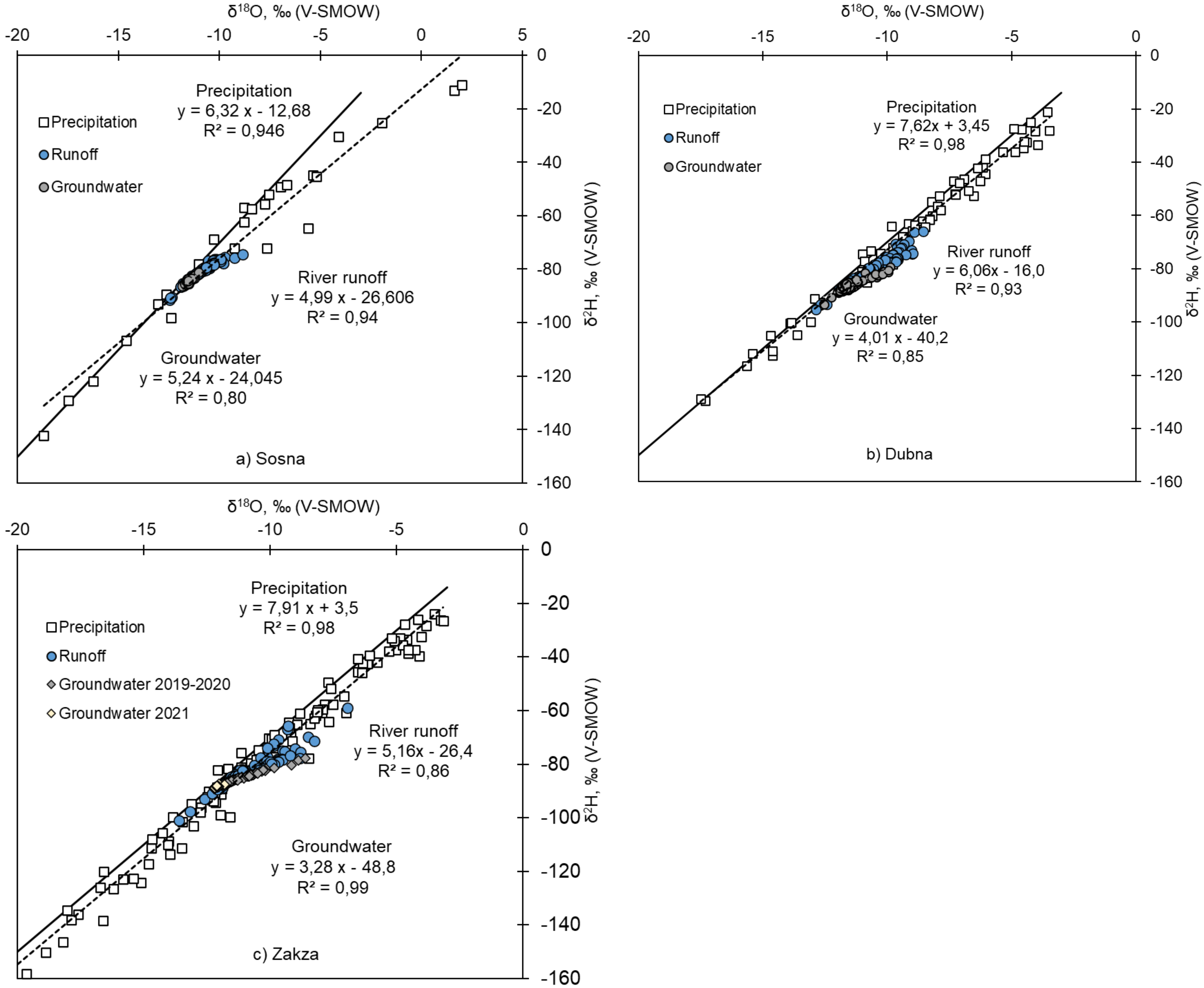

295 **Figure 2: The δ²H vs δ¹⁸O plot of river runoff, groundwater, and precipitation at gauging station of the a) Sosna river basin, b) Dubna river basin, c) Zakza river basin.**

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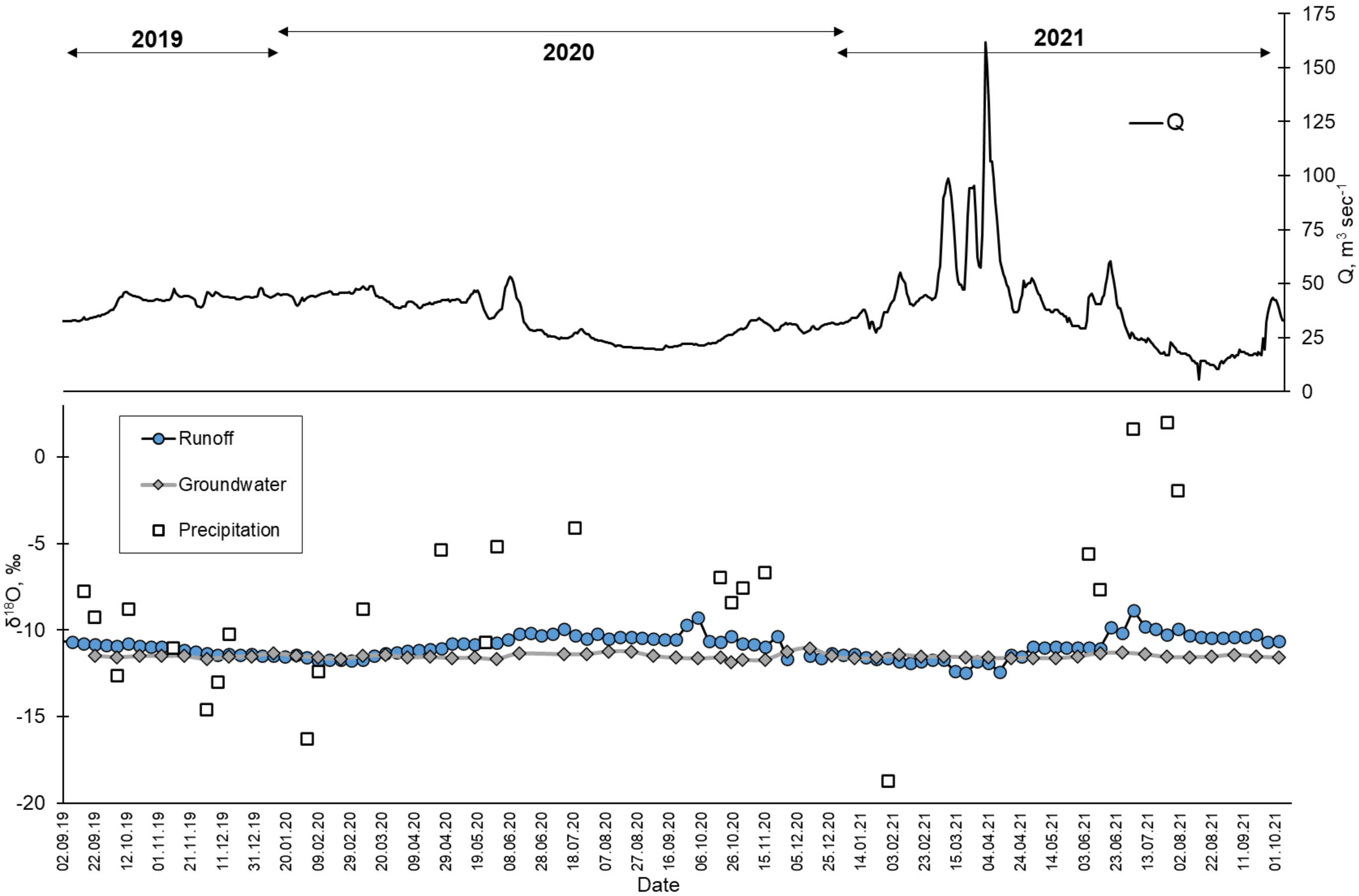

Figure 3: Runoff of Sosna river at the Elets gauging station and $\delta^{18}O$ values of river water, groundwater, and precipitation for the period from 01.09.2019 to 01.10.2021.

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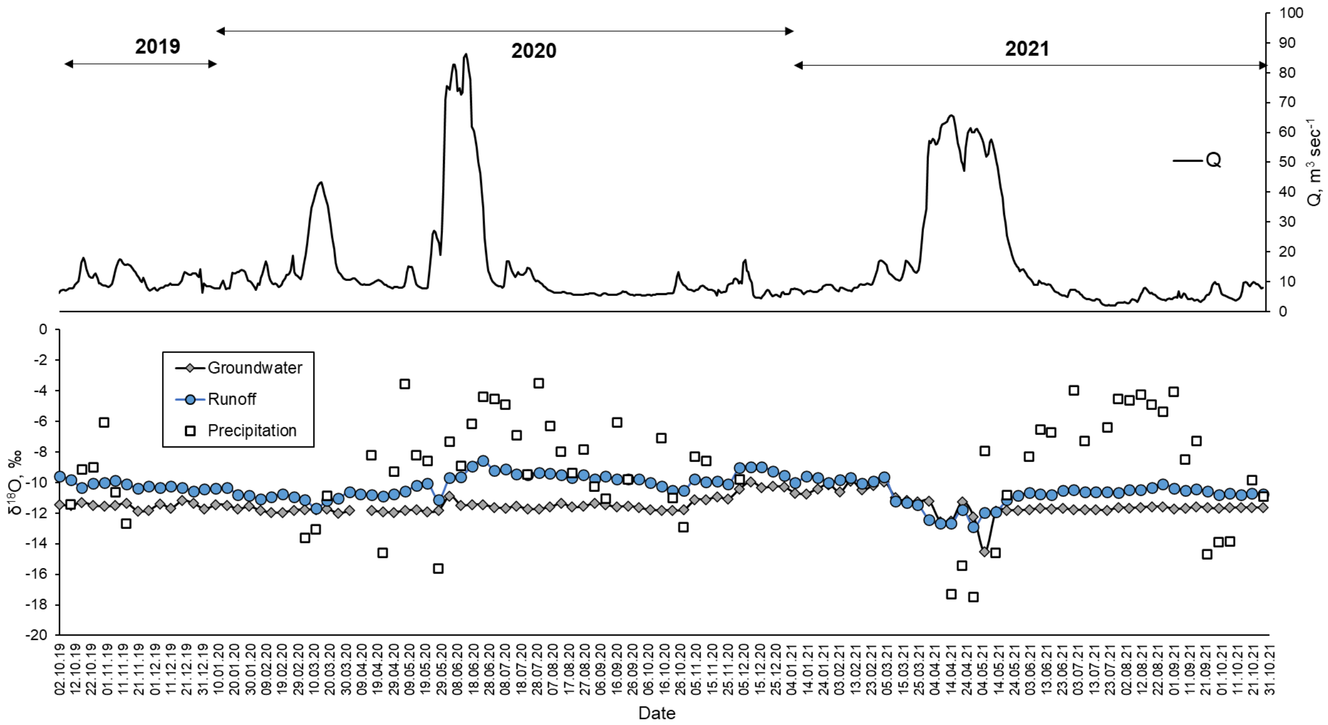

305 **Figure 4: Runoff of Dubna river at the Verbilki gauging station and δ^{18}O values of river water, groundwater, and precipitation for the period from 01.10.2019 to 31.10.2021.**

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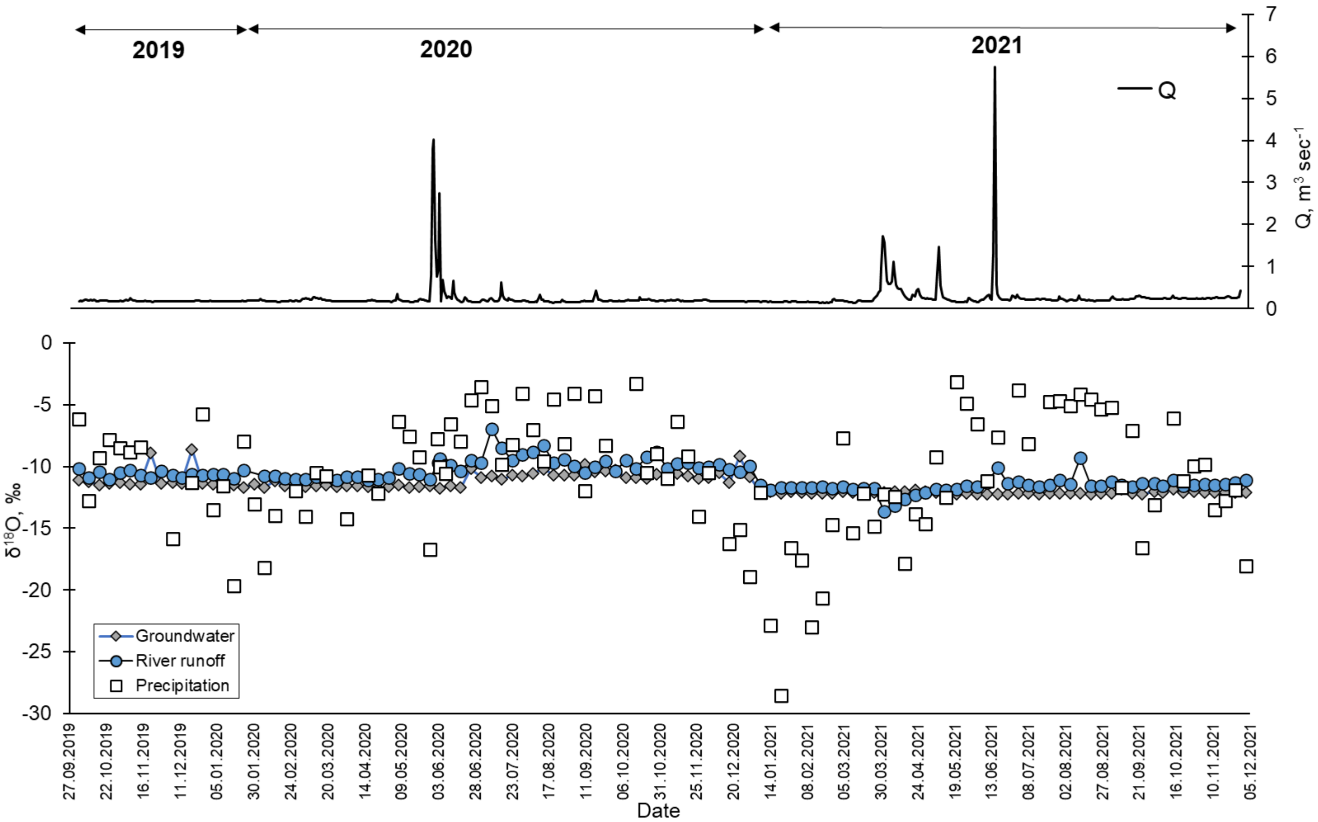

Figure 5: Runoff of Zakza river at the Bolshoe Sareevo gauging station and δ^{18}O values of river water, groundwater, and precipitation for the period from 01.10.2019 to 31.11.2021.

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