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Geostatistical evaluation of the design of the precipitation stable isotope monitoring network for Slovenia and Hungary

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ABSTRACT

A detailed knowledge of the stable isotope signature of precipitation is the basis of investigations in a variety of scientific fields and applications. To obtain robust and reliable results, the representativity of the currently operating (at least, as of 2018) precipitation stable isotope monitoring stations across Slovenia ($n = 8$) and Hungary ($n = 9$) was evaluated on the basis of amount-weighted annual averages with the aim of revealing any redundantly (i.e. over-) represented or un(der)represented areas. In the case of the latter, optimal locations for additional sites were suggested in Slovenia and Hungary. The networks of both countries are *design-based* systems that need to be fine-tuned for long-term optimized operation. The evaluation of the monitoring network was performed taking into consideration the stations operating in Slovenia and Hungary, as well as closely situated ones operating in neighboring countries. The evaluation was carried out in nine different combinations, using spatial simulated annealing, with regression kriging variance as a quality measure. The results showed that (i) there are over- and un(der)represented areas in the network, an issue requiring remedial action, (ii) the mutual information exchange of the precipitation stable isotope monitoring networks of Slovenia and Hungary increases the precision of precipitation $\delta^{18}O$ estimation by ~0.3‰ in a 15–30 km wide zone near the borders, and (iii) by an even greater degree in the neighboring countries' stations. The current research may be termed pioneering in the matter of the detailed geostatistical assessment of spatial representativity of a precipitation stable isotope monitoring network, and as such, can serve as an example for future studies aiming for the spatial optimization of other regional precipitation stable isotope monitoring networks.

1. Introduction

The hydrochemical and isotopic composition of precipitation is a powerful tool, and an invaluable source of information for hydrological, hydro-meteorological and climatological research (Bowen et al. 2019; Zhai et al. 2013). The most basic isotopic hydrological parameters are the ratios of the heavy to the light stable isotopes of hydrogen and oxygen ($^{2}H/^{1}H$; $^{18}O/^{16}O$) in water molecules (Craig 1961; Riesenfeld and Chang 1936). Stable isotope ratios in precipitation are primarily determined by kinetic and equilibrium isotopic fractionation associated with the processes of evaporation and condensation and atmospheric transport, as parts of the global water cycle (Dansgaard 1964; Eriksson 1965). For several decades, precipitation stable water isotopes have been used as important natural tracers (Craig 1961; Dansgaard 1964) in the study of the water cycle e.g. Fórizs (2003); Yoshimura (2015), in the discipline of hydrogeology e.g. (Clark and Fritz 1997), and more recently, in food

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authentication (Kelly et al. 2005) or in wildlife forensics e.g. Bowen et al. (2005); Hobson (1999) on global, regional and local scales.

The spatial design of meteorological monitoring networks has been thoroughly studied, e.g. Mauger et al. (2013), but the focus has been primarily on the optimization of precipitation gauging networks developed for recording precipitation amount (Kassim and Kottegoda 1991) (e.g. Goovaerts (2000); Papamichail and Metaxa (1996); Pardo-Igúzquiza (1998)). A global isotope-hydrometeorological monitoring network (GNIP – Global Network of Isotopes in Precipitation) has been jointly operated by the International Atomic Energy Agency (IAEA) and the World Meteorological Organization (WMO) since 1961. It is a basic source of information on the present-day degree of temporal and spatial variability of the isotopic composition of monthly precipitation on a global scale (Araguas-Araguas et al. 1996). The GNIP database is supplemented by information gathered through national meteorological services, national authorities and scientific volunteers. In a few notable cases it is supplemented by the national monitoring networks of precipitation water stable isotopes, e.g. in Switzerland (Schürch et al. 2003), Spain (Díaz-Teijeiro et al. 2009), USA (Welker 2012), Germany (Stumpp et al. 2014), China (Liu et al. 2014) and Austria (Hager and Foelsche 2015).

In every case, the spatial representativity of the given environmental monitoring network has to be examined for redundancy or a lack of necessary coverage. These aspects are important from both a theoretical and practical – that is, cost optimization – point of view, since redundancy implies excess expenditure in the case of any given environmental monitoring network, e.g. (Nunes et al. 2004). Such an audit has not yet been completed, neither for the GNIP, nor for any of the national precipitation stable isotope monitoring networks mentioned above. In one special regional case, the GNIP records from the Iberian Peninsula combined with data from regional studies were jointly used to provide an overview of the representativity of the precipitation stable isotope monitoring stations (Hatvani et al. 2020).

The assessment of to what extent a monitoring network provides spatial coverage of an area can be considered as an optimization problem. One of the most traditional approaches to monitoring optimization for spatial classification is the application of geostatistical tools (Hatvani and Horváth, 2015) by means of an analysis of e.g. kriging/simulation variances (Kanevski et al. 2009; Szatmári et al., 2015; Szatmári et al. 2019). A commonly applied quality measure of the quantification of the geographical coverage of a monitoring network with regard to a pre-specified domain is the kriging variance (KV); see Section 3.4 and Baume et al. (2011); Melles et al. (2011) for details. The computation of the KV is part of the kriging interpolation procedure, which has several advantages in terms of network optimization. First of all, it is an unconditional variance (Chilès and Delfiner 2012), meaning that it does not depend on the data values. Thus, it can be computed before the actual sampling takes place, which makes it cost-effective. However, this independence has to be taken into consideration if one wishes to use the KV for uncertainty assessment (Szatmári and Pásztor 2019). A further advantage of the application of kriging variance to the evaluation of the representativity of a monitoring network is that it is able to make a numerical comparison and rank the alternative geometric data configurations (Journel and Rossi 1989). As an objective statistical measure of representativity, the KV may be considered as a *bottom-up* criterion (see Parr et al. (2002)) for selecting the most important sites in the environmental monitoring network being examined.

The two main approaches in selecting the location of environmental monitoring stations are (i) *model-based* and (ii) *design-based*. In the case of the former, the information provided by the monitoring network depends on the validity of the model adopted. These *model-based* networks are much more flexible than *design-based* ones, but are necessarily pre-determined by the underlying assumptions from which they are derived. Meanwhile, the operation of *design-based* networks is less flexible, as well as being costly, but because these rely on a smaller number of assumptions, they provide more robust information than the *model-based* ones (Parr et al. 2002). The precipitation stable isotope monitoring networks of Slovenia and Hungary (the subjects of the study) represent a *design-based* system that should be fine-tuned for long-term optimized operation. In the course of the optimization procedure, an objective a *model-based* approach will be considered by taking into account additional key factors that provide added value in design-based planning. The most important of these key factors that should be considered in a *post-hoc* stratification of an investigated environmental monitoring network are:

- the conservation of long-term integrated environmental monitoring at designated permanent sites (Beard et al. 1999; Bruns et al. 1991; Pryor et al. 1998; UNESCO 1993). These sites allow (i) the reliable estimation of baseline values for significant environmental variables and indicators (Bruns et al. 1991) (ii) the development of generic models of predictive value as a basis for environmental management and the use of natural resources (Parr et al. 2002);
- the conservation of the monitoring sites that provide an understanding of environmental conditions prevailing in locations of specific importance (Innes, 1988), which criterion is expressed based on professional experience as a *top-down* decision (Parr et al. 2002);
- the inclusion of monitoring sites from a neighboring network/region, since these can reduce the need to set up new sites (Parr et al. 2002).

1.1. Development of the national monitoring network for stable isotopes in precipitation across Slovenia and Hungary

Systematic measurements of the isotopic composition (i.e. δ^2H and δ^{18}O) of precipitation were commenced in Slovenia in 1972 at three stations (Gospodarič and Habič 1976). In Hungary, sporadic measurements began in the early 1970s (Dénes and Deák 1981) and the first regular monthly measurements of precipitation stable isotopes were conducted between 1977 and 1984 at a single station (Deák 2006). The station in Hungary with the longest record of operation is located in Debrecen, and has been gathering data since 2001 (Vodila et al. 2011). In Slovenia, the longest continuously operating monitoring stations are the Ljubljana Reaktor and Portorož stations, working since since 2000 (Vreča et al. 2015); the former having been relocated twice between 1981 and 2000 in the Ljubljana area (Vreča et al. 2008). The number of monitoring stations has shown an increase in Hungary in 2012 – after the setting up of a country-wide network (Czuppon et al., 2017a) – and increased in 2009 in Slovenia (Vreča and Malenšek 2016). Stable isotopes have been monitored at more than 30 different locations in Slovenia, although only few stations have been in operation simultaneously for more than three years (Vreča et al. 2018; Vreča and Malenšek 2016). A kind of proliferation has also been observable in Hungary, since beside the continuously-operating monitoring stations, ad hoc stations (Czuppon et al. 2018) and small-scale industrial networks (Fórizs et al., 2020; Szántó et al., 2007) have also been launched at various times. This growth in the number of precipitation isotope monitoring stations gave rise to the idea of an investigation into the possibility of optimizing the current network and finding any locations where new monitoring stations could operate in the most useful way.

1.2. Aims of the study

The aim of the present study is to establish a sound scientific basis for the optimal spatial organization of the national networks for water isotope monitoring in precipitation in Slovenia and Hungary. Specifically, the goal is to evaluate the representativity of the currently operating networks across Slovenia and Hungary to reveal areas either with redundant coverage or which are un(der)represented. In the case of the latter, the optimal locations for possible further sites are suggested. The precipitation stable isotope monitoring stations are assessed in various combinations. The following questions are addressed: (i) for which country the existing precipitation stable isotope networks are

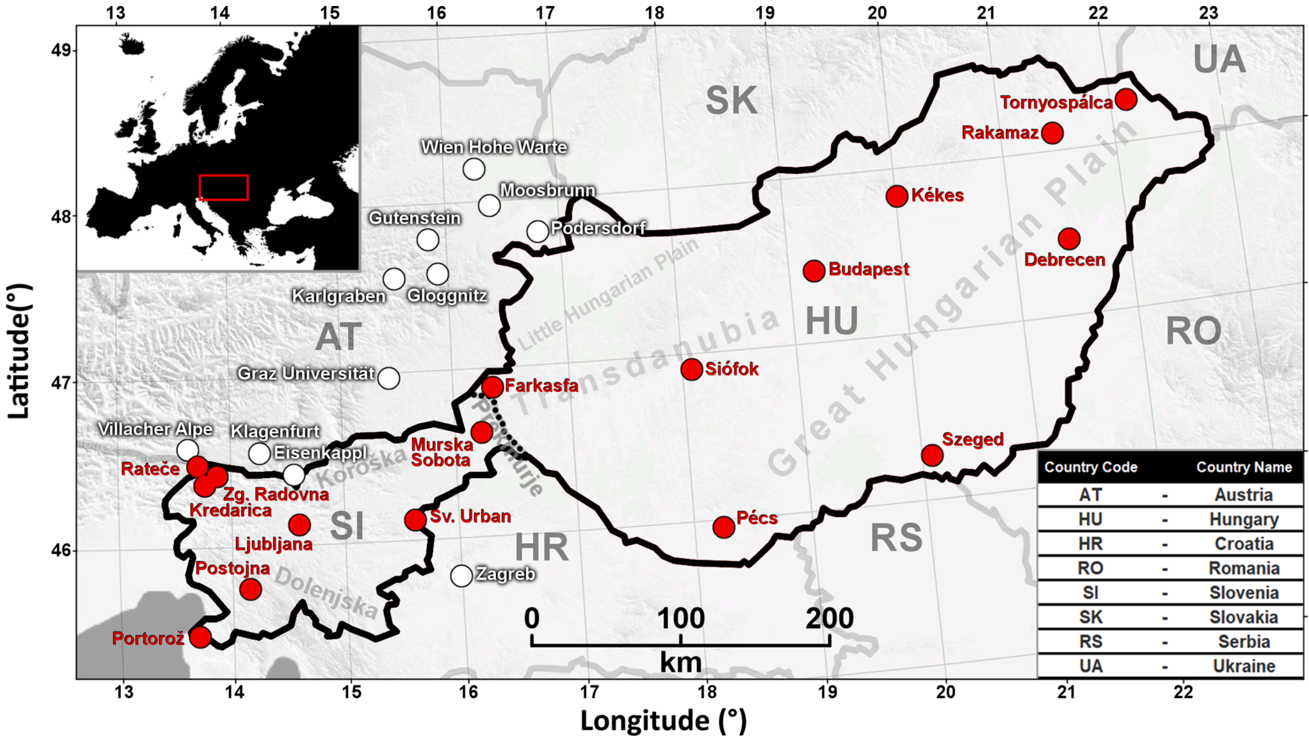

Fig. 1. Schematic map of the spatial distribution of the monitoring sites for precipitation stable isotopes in Slovenia (SI; $n = 8$) and Hungary (HU; $n = 9$) (outlined in black) (sites are indicated by red dots) operating in 2018, and in the neighboring counties (Austria: AT ($n = 10$), Croatia: HR ($n = 1$)) available in open databases (white dots). The inset map shows Europe with the region indicated by a red rectangle. The base map was taken from SRTM Worldwide Elevation Data (3-arc-second Resolution); accessed on 07.03.2020. The country codes on the map and in the inset table follow the ISO-3166–1 ALPHA-2 standard at every location mentioned. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

sufficient, (ii) which sites are less crucial to the effectiveness of monitoring, and (iii) to what extent information is available from the neighboring countries, and to what degree this is beneficial.

2. Climate of the area

Most of the studied region (Fig. 1) is characterized by a warm temperate/continental climate with warm summers and without a dry season (Köppen-Geiger code: Cfb). Humid warm temperate areas with hot summers (Köppen-Geiger code: Cfa) can be found along the Adriatic Coast and the southern border of Hungary; while a boreal climate with cool summers and cold winters (Köppen-Geiger code: Dfc) prevails in the northwestern mountainous part of Slovenia (Kottek et al., 2006).

Determined by the geographical position of the studied region, westerly winds prevail at higher altitudes, which can be deflected by orography near the ground surface (Bertalanič 2020; Bihari et al., 2018). Specifically, the winds in Slovenia are mostly westerly (Bertalanič 2020), in Hungary, the dominant wind direction is north-westerly (relative abundance 10–15%), and in the areas with highly variable relief conditions determining surface wind directions can vary from NW to S. The average annual wind speed at 10 m above the surface is between 2.5 and 3 m s^{-1} in Hungary (2000 to 2009), with higher speeds recoded on the Great Hungarian Plain and the Transdanubian Mountains (Bihari et al., 2018). The annual average wind speed, in the period from 1996 to 2005, in the lowlands of Slovenia rarely exceeds 2.0 m s^{-1}, while higher (up to 5.1 m s^{-1}) values characterize the hills and mountains (Bertalanič 2020). The highest wind speeds both in Slovenia and Hungary can be attributed to southerly winds caused by cyclones from the Mediterranean (Bertalanič 2020; Bihari et al., 2018).

In the studied area, the Western- and Central Mediterranean are the dominant marine moisture source regions throughout the year, although in summer locally recycled continental moisture is the primary source of atmospheric moisture (Czuppon et al., 2017a; Gómez-Hernández et al., 2013). The dominance of the Mediterranean moisture decreases beyond ~400 km from the Adriatic Coast (Kern et al. 2020a), and in the northern part of the study region the westerlies transport Atlantic marine moisture mixed with recycled terrestrial moisture from western and west Central Europe (Bottyán et al., 2017; Czuppon et al., 2017a).

A pronounced precipitation gradient characterizes the study area which, as in the case of the wind climate, is also determined by the geographical conditions. The mean annual sum precipitation decreases from north-west Slovenia (>1500 mm) eastwards dropping to ~540 mm in Eastern Hungary, largely mirroring the orography and continentality of the region (Fig. 2). The primary sources of moisture are the humid air masses arriving from the Mediterranean perpendicular to the orographic barrier, thus, as the moist air mass is forced to rise, the consequent cooling and condensation cause heavy precipitation events (De Luis et al. 2014).

3. Materials and methods

Cooperation between Slovenia and Hungary in the matter of their networks of precipitation stable isotope monitoring was established by 2016. Annual datasets of complete measurements are available from the majority of active stations in both countries from 2016, 2017 and 2018 (Fig. 1). There have been minor changes in the set of stations over the years. In the beginning, in 2016, a less complete network was operating, with the north-eastern stations (Tornyospálca and Rakamaz, Hungary; Fig. 1) not yet functioning. The complete set of stations began operation in 2017 and 2018. However, there is a notable difference between the quality of the data gathered from 2017 and 2018, although in both there were just two stations with a certain portion of fallen precipitation not represented in the stable isotope measurements. Specifically, in 2017 the percentage of actual precipitation not represented was notably higher (>8%), while in 2018 it was less than 4%, rendering the latter more representative in this aspect.

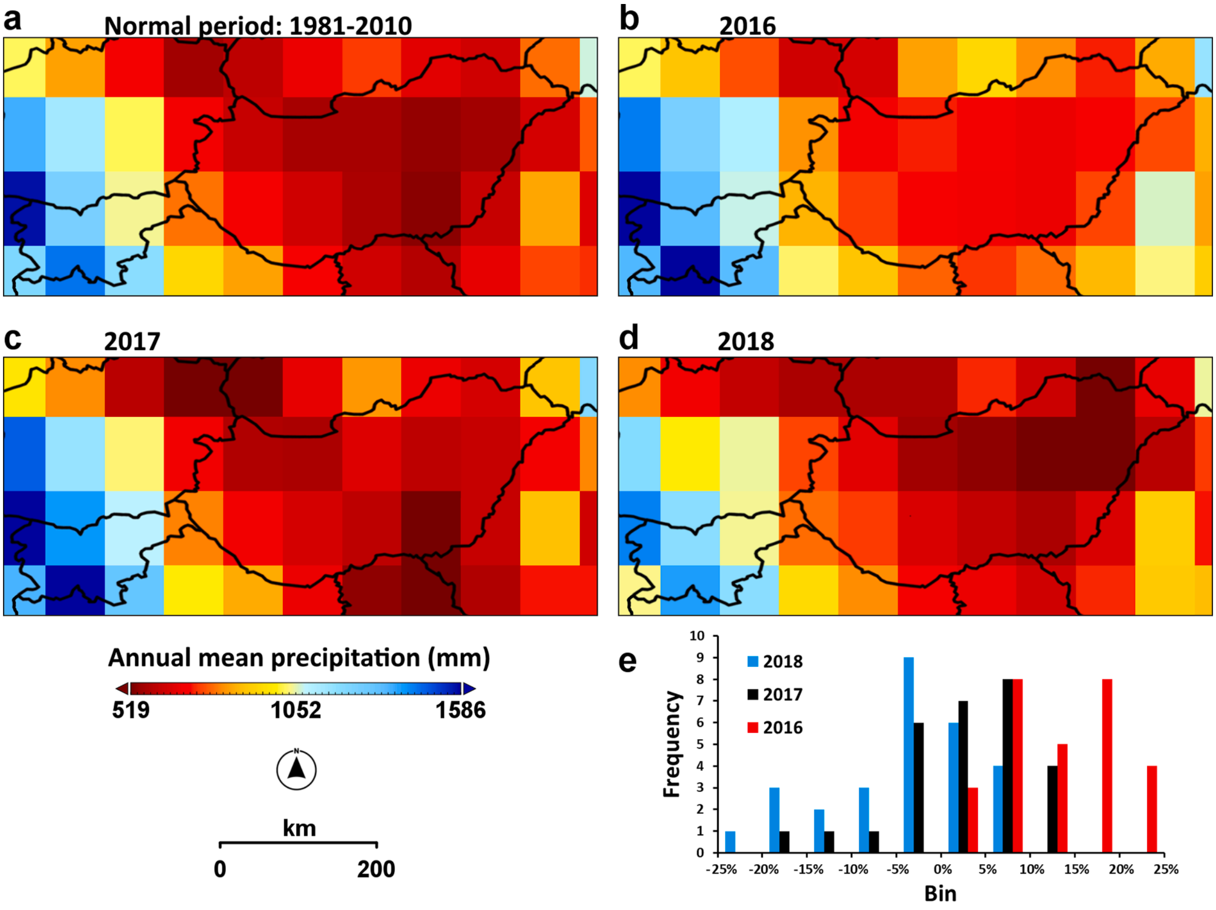

Fig. 2. Spatial distribution of annual precipitation totals in Slovenia and Hungary and their surroundings. Annual mean sum for the normal period: 1981–2010 **(a)** and annual total precipitation for years 2016 **(b)**, 2017 **(c)**, 2018 **(d)** based on the GPCC's Full Data Monthly Analysis V.2018 database (Schneider et al. 2018). Black lines represent national borders. The histograms calculated from the annual mean sum of the normal period subtracted from the annual total precipitation for 2016 (red), 2017 (black) and 2018 (blue) are expressed in percentages of the annual normal (1981–2010) precipitation of the corresponding grid cell **(e)**. The calculation was performed for the 28 individual grid cells covering Slovenia and Hungary. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

With regard to the hydroclimate of the area, the year 2018 is most similar to the average conditions (Fig. 2a,b,e) if it and the years 2016 and 2017 are considered (Fig. 2c–e). In 2018, both the areal average of the annual sum precipitation (~760 mm) and its spatial distribution (Fig. 1d) best resembled the long-term normal precipitation (~785 mm) (Fig. 2a, e). The differences in the gridded annual precipitation totals of 2018 and the normal period (1981–2010) were clustered within the ±5% interval (Fig. 2e) in more than half (~55%) of the cells, with the largest differences along the northern margin (Fig. 2a,d). In addition, in December 2016, there was no precipitation across large parts of the region, yet that year was much wetter than the normal period, as indicated by the positive bias in difference values (Fig. 2e). This suggests an unusual seasonal distribution of precipitation for 2016. In the meanwhile, 2017 was also wetter than the average conditions in Slovenia and Hungary (Fig. 2c,e).

Therefore, on the basis of the above – i.e. with most sites active in 2018, and the hydroclimate of this year resembling that of the normal period the most – the 17 stations operational in 2018 were used for the geostatistical evaluation of the precipitation stable isotope monitoring network design for Slovenia and Hungary considering additional stations from the neighboring countries (Fig. 1; Table A1; see Section 3.1 for details).

3.1. *Used $\delta^{18}O$ and precipitation data and preprocessing*

The ratio of heavy to light stable isotopes is traditionally expressed in delta (δ) notation as the relative deviation of the ratio of the sample from that of the standard, expressed in per mil (‰) (Coplen 1994). Monthly precipitation hydrogen and oxygen stable isotope values (δ_p) were provided for 2018 from 17 stations (Fig. 1; Table A1), eight of which were selected from the Slovenian Network of Isotopes in Precipitation (SLONIP) operated by the Jožef Stefan Institute (Vreča et al. 2017), and nine from Hungary (Czuppon et al., 2017b) curated by the Research Centre for Astronomy and Earth Sciences, with an additional station operated by the Institute for Nuclear Research (Vodila et al. 2011). There were only two stations with missing monthly δ_p values due to sampling failure in January at Sv. Urban (SI) and in December at Farkasfa (HU). In addition, to (i) improve the spatio-temporal density of the data set, and (ii) obtain a more realistic picture of how the focus area is presented in terms of precipitation δ_p, the available data for 2018 provided by the stable isotope monitoring stations within 150 km of Slovenia and Hungary (the focus area of the study) were also taken into account. These complementary data included ten stations from the Austrian Network of Isotopes in Precipitation (ANIP) (Umweltbundesamt, 2019), and one station in Croatia (HR) (Krajcar Bronić et al. 2020) (Fig. 1; Table A1). A measure of local indicator of spatial association, specifcally a sequential univariate outlier detection procedure (Kern et al., 2020b) did not detect any possible outlier among the monthly δ_p data.

For the geostatistical modeling, precipitation amount weighted annual averages were calculated using monthly precipitation amounts from the GPCC's Full Data Monthly Analysis V.2018 database (Schneider et al. 2018; Schneider et al. 2014).

For calculations, the geographical coordinates (latitude and

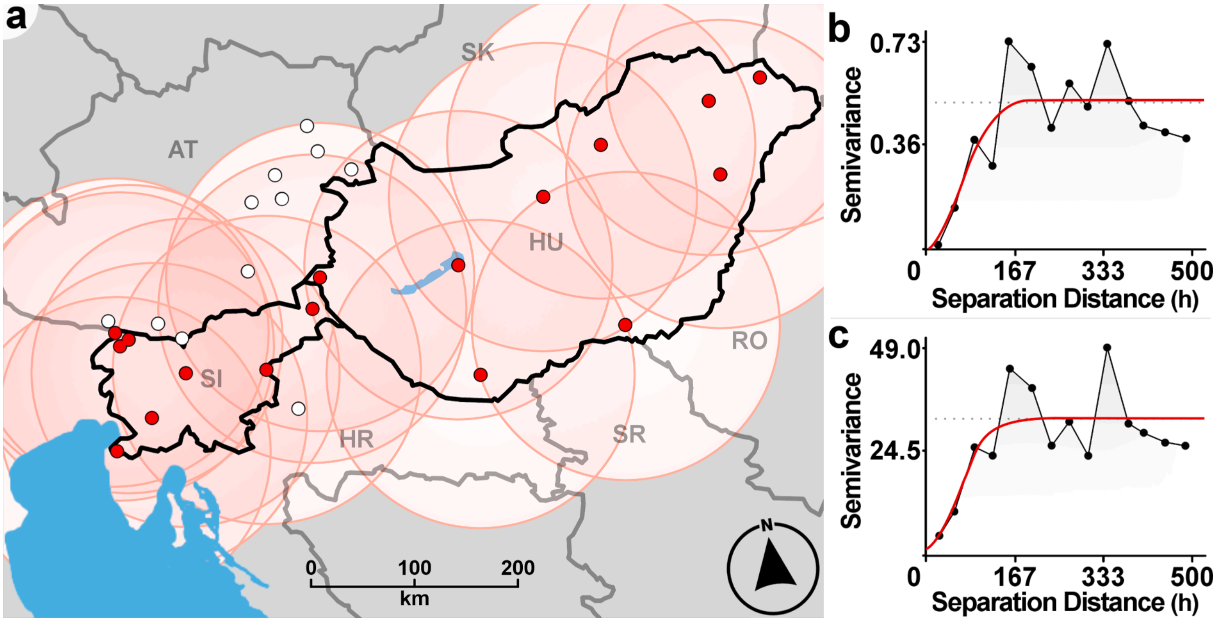

Fig. 3. Spatial range union and semivariogram of the precipitation stable isotope monitoring network of Slovenia and Hungary for 2018. The monitoring sites of the Slovenian – Hungarian precipitation stable isotope network are indicated with red, and those of other countries with white dots **(a)**. The empirical semivariograms for δ^{18}O **(b)** and δ^{2}H **(c)** are indicated by the black dots, and the red line represents the theoretical semivariogram. The dotted horizontal line indicates the average variance. The underlying properties of the Gaussian δ^{18}O and δ^{2}H theoretical semivariograms are the following – range: 154.1 and 140.3 km; $C_0 + C$: 0.523 and 32.4; C_0: 0.001 and 2.05; fit: $r^2 = 0.651$ and 0.586, RSS = 0.182 and 722, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

longitude) of the stations were converted from a geographic coordinate system (EPSG: 4326, WGS84; Table A1) to a metric coordinate system (EPSG:3857, WGS84 / Pseudo-Mercator projection), since exploratory variography (see Appendix) should be performed on a metric scale. It should be noted here that in the study the reported ranges are planar distances (d_{planar}) in km (EPSG: 3857), unless otherwise reported; the estimated average conversion in the region is given by $d_{planar} \times 0.678 \approx d_{geodetic}$.

3.2. Methodological approach

The first step after preprocessing the dataset was the removal of the governing effect of spatial trend with the use of multivariate regression on amount weighted annual mean δ_p values. This was necessary to obtain a semivariogram (see Appendix), which provides the weighting function for kriging. Regression kriging combines multivariate regression with the kriging of the regression residuals, and is used in the present study as the quality measure of spatial simulated annealing (SSA). It should be noted here that since the semivariograms for both precipitation hydrogen and oxygen stable isotope values were strikingly similar (Fig. 3b, c), except for their degree of variance, network optimization was performed only for the more frequently assessed variable, δ^{18}O, and the results may be taken as relevant to both parameters (δ^{2}H and δ^{18}O).

3.3. Removal of geographical governing effects

Trend-like tendencies in amount weighted annual mean δ_p over large distances which are determined by geographical factors (e.g. 'altitude' and the 'continental' effects) are known to drive the isotopic composition of precipitation globally (Rozanski et al. 1993) and regionally (Kern et al., 2020a). The complex physical mechanisms explaining these relationships can and should be explored; in the present study, however, their superimposed effect on precipitation stable isotope values at given locations should be removed, because these may mask the finer scale spatial autocorrelation patterns which the optimization of the monitoring network relies on. Thus, as in other, similar studies (e.g. Hatvani et al. (2020); Hatvani et al. (2017); Kern et al. (2014)) the effect linked to the geographical factors was minimized by computing best-fit multiple regression models (δ_p vs. latitude, longitude, elevation and planar distance from the Adriatic) and determining their residuals in a stepwise procedure suggested by O'Brien (2007).

3.4. Spatial simulated annealing (SSA)

Spatial simulated annealing (SSA) was used to identify areas un(der) represented or with redundant coverage by the precipitation stable isotope monitoring network across Slovenia and Hungary for 2018. SSA is an iterative, combinatorial, model-based network optimization algorithm in which a sequence of network designs is generated by deriving a new design by slightly and randomly changing the previous design (van Groenigen et al. 1999). When a new design is generated, the quality measure is computed and compared with the quality measure value of the previous design. The Metropolis criterion (Metropolis et al. 1953) defines the probability that either accepts the new design as a basis for further computation or rejects it, in which case the previous design stays as a basis for further designs (van Groenigen et al. 1999). In this study, SSA and its inverse application were used for (i) optimally adding new sites to areas which are underrepresented by the existing monitoring networks, and for (ii) the optimal removal of those monitoring sites which have a lower information content according to the quality measure (Heuvelink et al. 2012; Szatmári et al. 2019), respectively.

The appropriate selection of the quality measure is a crucial step, since the whole optimization process rests on it. In the course of network optimization, spatially averaged kriging variance was applied as a quality measure for quantifying the geographical coverage of the monitoring network, which is a widely accepted measure for the problem in hand (e.g. Bogárdi et al. 1985; van Groenigen 2000).

The kriging variance can be readily computed if the variogram is known, that is

$$\sigma^2(x_0) = 2\sum\nolimits_{i=1}^{N}\lambda_i\gamma(x_i, x_0) - \sum\nolimits_{i=1}^{N}\sum\nolimits_{j=1}^{N}\lambda_i\lambda_j\gamma\left(x_i, x_j\right) \tag{1}$$

where $\gamma(x_i, x_j)$ is the semivariance between the data points x_i and x_j, $\gamma(x_i,$

Table 1
Summary of the scenarios set in this study.

Scenario	1	2	3	4	5	6	7	8	9
Network is optimized for	HU	SI	SI	HU	SI	SI	HU	HU-SI	HU-SI
Are the SI sites involved?	no	yes	yes	yes	yes	yes	yes	yes	yes
Fix sites in SI?	–	no	Ljubljana, Portorož	–	no	Ljubljana, Portorož	Ljubljana, Portorož	no	Ljubljana, Portorož
Are HU sites involved?	yes	no	no	yes	yes	yes	yes	yes	yes
Are sites from Austria and Croatia considered?	no	no	no	no	no	no	no	yes	yes
Section	4.1.1			4.1.2				4.2	

x_0) is the semivariance between the i^{th} data point and the prediction point x_0 and λ is the kriging weights that can also be computed if the variogram is known (Webster and Oliver 2008).

The advantage of kriging variance as the quality measure for SSA is that according to its definition, the kriging variance is independent of the data values, and that makes it an unconditional variance (Chilès and Delfiner 2012). Furthermore, it is a variogram model dependent ranking of data configuration (Journel and Rossi 1989), and can, therefore, be highly useful in ranking and optimizing a monitoring network (Szatmári et al., 2015).

In the study, maps of regression kriging variance (RKV) are used for evaluating, comparing, ranking and optimizing the monitoring network (see e.g. Szatmári et al. 2019) via scenarios of different sets of monitoring stations (Table 1). Regression kriging is equal to the procedure discussed above, and is conducted on the residuals of a multivariate regression – for details see Sections 3.3 and 4. These RKV maps provide information on the geographical coverage of different geometric data configurations and can therefore be used for selecting the configuration which provides the best spatial coverage for the domain of interest. If the possibility arises to add one or more sites to an existing monitoring network, the location of the new sites is optimized by maximizing the decrease of the spatial mean of RKV, which yields a more even spatial coverage. In this study, a sequential optimization approach is conducted, that is, the optimal placement of only one new site at a time is targeted, taking the configuration of the existing monitoring network and the location of the previously added new site (or sites) into account. The higher the RKV, the higher the uncertainty of the geostatistical estimation: thus, it can be considered the error of the estimation as well. These can be used for the visualization of the geographical coverage of a given geometric data configuration in order to identify and delimit areas which are either overrepresented or underrepresented by monitoring stations (Szatmári et al., 2015). These kriging maps can be interpreted with the assumption of normality and homoscedasticity as the uncertainties of the geostatistical modeling for δ_p data.

3.5. Scenarios with different sets of the precipitation stable isotope monitoring stations considered across Slovenia and Hungary

Using SSA, nine scenarios were investigated (Table 1) to provide a picture of how efficient the precipitation stable isotope monitoring

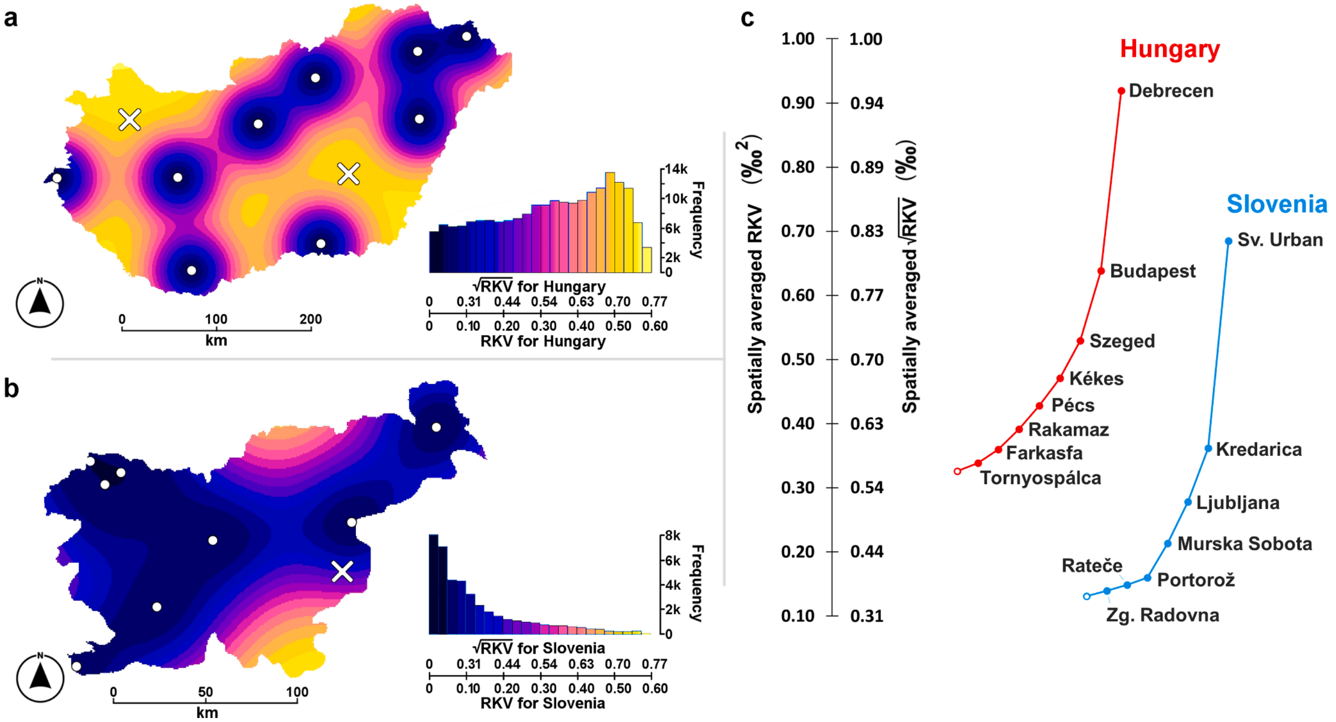

Fig. 4. The regression kriging prediction-error variance (RKV) of the active precipitation stable isotope monitoring network in 2018 across Hungary (a; Scenario 1) and Slovenia (b; Scenario 2). The values are reported as RKV (‰^{2}) and $\sqrt{RKV}$ (‰) in all figures. The inset histograms show the frequency distribution of RKV across the corresponding countries. The graphs in panel (c) show the increase in spatially averaged RKV with the omission of given sites for Hungary (red) and Slovenia (blue). The RKV of the full national network (the so-called ‘zero state’), corresponding to the maps in panels (a) and (b), is indicated by an empty circle. The countries in this case are considered separately without any information exchange between their precipitation stable isotope monitoring networks. The white crosses represent the optimal location if new sites are to be set up, for details see Section 5. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

networks of HU and SI are functioning. These were as follows; the networks are

- operating in a standalone way, with no information exchange between the countries, and with the condition that any monitoring site can be discarded (Scenarios 1 & 2), or that Ljubljana and Portorož cannot be discarded (Scenario 3), given their long and uninterrupted period of operation since 1981 and 2000, respectively, and historical participation in the GNIP network (Vreča et al. 2015; Vreča et al. 2014).
- operating in a standalone way with information exchange between the countries and the condition any monitoring site can be discarded (Scenarios 4 & 5), and, as above, the long-term operation of the stations at Ljubljana and Portorož cannot be discarded (Scenarios 6 & 7).
- being managed together with the information gathered from neighboring countries being taken into consideration (Scenarios 8 & 9).

4. Results and discussion

The first step after preprocessing the dataset of δ_p for the Adriatic Pannonian Region of 2018 was the removal of the large-scale governing effect of the spatial trend using multivariate regression. Latitude (Y) and elevation (ELE) were found to explain the effect of geographical factors influencing the δ_p records (Eqs. (2) and (3)) significantly ($p < 0.01$; corrected $r^2 > 0.44$) with negligible variance inflation (VIF $= 1.003$). The regression coefficients found for the 'elevation' parameter are justified, since these match the estimated isotopic 'altitude' effect for modern precipitation for the region for both isotopic species (Fórizs et al. 2011; Kern et al. 2020a; Malík and Michalko, 2010).

$$\delta^{18}\widehat{O} = -(5.18 \pm 6.361) + \left(-2.21[\pm 1.07] \times 10^{-6}\right) \times Y + \left(-1.33[\pm 0.25] \times 10^{-3}\right) \times ELE \tag{2}$$

$$\delta^{2}\widehat{H} = 87.77 \pm 49.62 + \left(-2.45[\pm 0.83] \times 10^{-6}\right) \times Y + \left(-7.7[\pm 0.19] \times 10^{-3}\right) \times ELE \tag{3}$$

The assessment of the spatial autocorrelation structure (see Appendix) of the regression residuals of the then (2018) currently operating precipitation stable isotope monitoring network in Slovenia and Hungary indicated an average ~ 150 km isotropic spatial range, suggesting a complete spatial coverage of Slovenia and Hungary (Fig. 3), and this was even if the active monitoring sites in the neighboring countries were not considered (Fig. 3: white dots). However, the degree of the coverage increases as one moves in a southwesterly direction. As indicated by the numerous overlapping range ellipses (Fig. 3), the (south)western part of the region is much more, even over-represented (Figs. 1 and 3). Moreover, as one moves toward the eastern parts of the study area, the quality of the coverage decreases.

4.1. Exploration of the national precipitation stable isotope monitoring networks of Slovenia and Hungary

4.1.1. Considering the national networks as standalone ones with no information exchange between them (Scenarios 1–3)

It becomes clear from the maps of the kriging variances that although both countries are entirely covered by the currently active precipitation stable isotope monitoring network, in Hungary, the predictability of δ^{18}O values is lower than in Slovenia (Fig. 4a, b). It should be noted that on the maps RKV is reported, which is a statistical measure in ‰^{2}, the square-root of which ($\sqrt{RKV}$) is the per mil value (‰). The latter is of interest when model uncertainties are compared to actual analytical uncertainties. In Hungary, the $\sqrt{RKV}$ is relatively high (min = 0.56‰), while in Slovenia the minimum is below 0.36‰ (Fig. 4c). Specifically, the error of the present prediction is below 3 st. dev. of the typical analytical precision of the currently most frequently used method for stable isotope analysis of δ^{18}O (1 st. dev. = 0.1‰ (Lis et al. 2008)) in ~52% and 11% of the territory of Slovenia and Hungary, respectively (Fig. 4a,b inset histograms). The areas where the estimation uncertainty exceeds a value three times that of the analytical error ($\sqrt{RKV} > 0.3$‰) lie along the central south-north axis of Slovenia and most of Hungary, except for a zone within ~25 km of the stations. If the setting up of additional monitoring sites is planned, under the current circumstances, these should be the primary subject areas to the increase quality of the representativity of the region, e.g. NW Hungary. If, however, the opposite occurs, and a monitoring station must cease its activity, the current network will suffer the least information loss if that station is removed which changes the RKV the least (Fig. 4c). In the case of Hungary, discarding Tornyospálca would result in the smallest information loss (Fig. 3c, and illustrated in Fig. A1a), while in the case of Slovenia discarding Zg. Radovna and Rateče would result in negligible information loss (Fig. 3c and A1b).

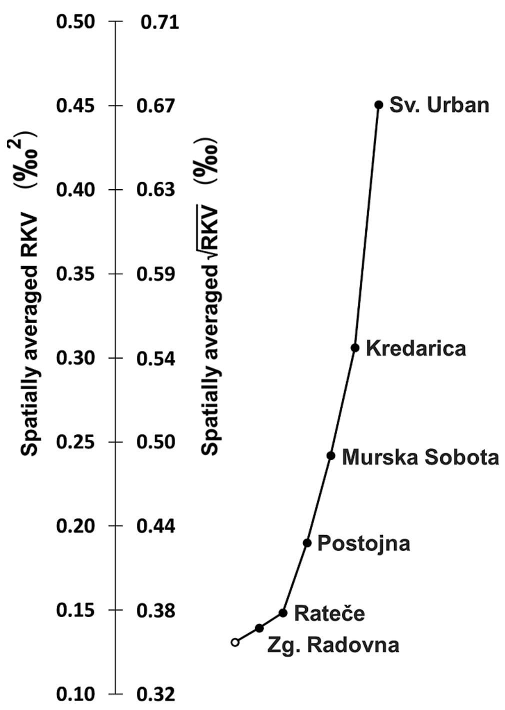

Fig. 5. Increase in RKV (‰^{2}) depending on which station is discarded from the active precipitation δ^{18}O monitoring network in 2018 across Slovenia, assuming that the Ljubljana and Portorož stations have to remain continuously in operation. The zero state is indicated by an empty circle.

In any monitoring network, there might be stations whose importance is enhanced/increased due to those providing long-term integrated environmental monitoring e.g. Beard et al. (1999), baseline information e.g. Bruns et al. (1991), or essential information concerning a specific environment (see Section 1). In the Slovenian network of isotopes in precipitation (SLONIP), the Ljubljana and Portorož stations are such stations because of their long and uninterrupted operation and participation in the global network (Vreča and Malenšek 2016). Thus, in various scenarios (Scenarios 3, 6, 7 and 9), the question of which stations could be first removed from the network if the previous two must continue operation was addressed (Table 1). In Scenario 3 the picture did not change compared to Scenario 2 (Fig. 4b,c), but discarding Zg. Radovna and Rateče would result in practically negligible information loss in either case. However, the average RKV increases, since in both cases, these two stations do provide some measure of information to the network (Fig. 5).

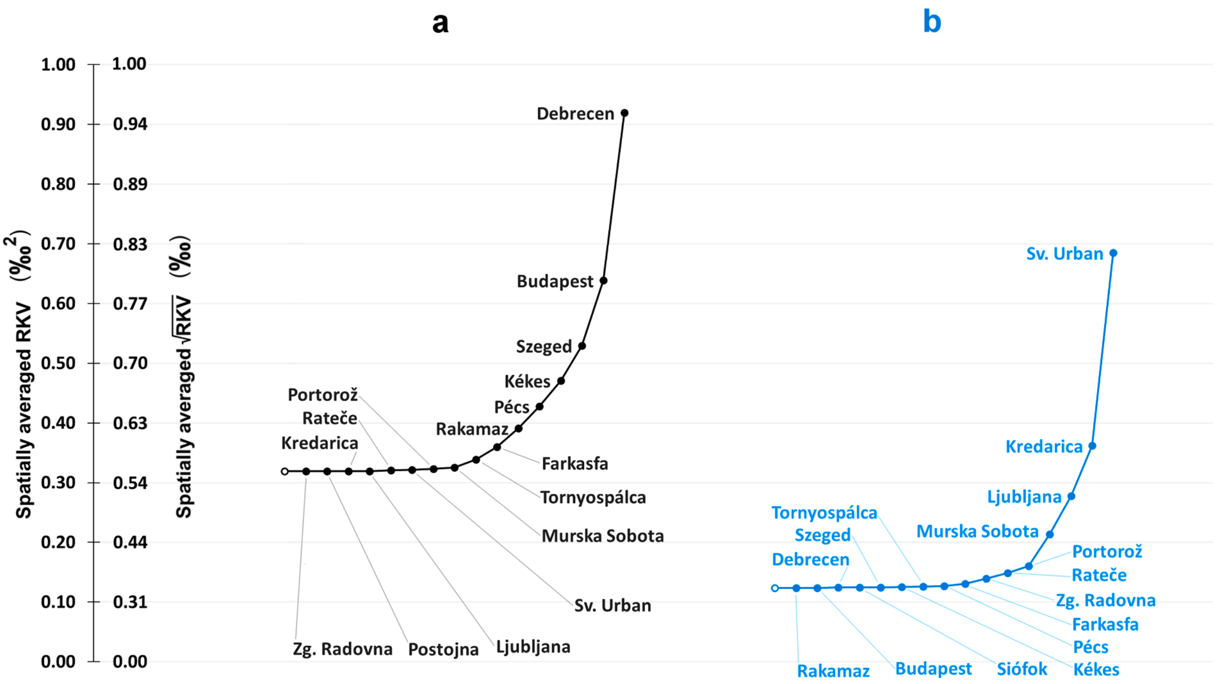

Fig. 6. Increase in RKV depending on which station is discarded from the active precipitation stable isotope monitoring network in 2018 across Hungary (a; black; Scenario 4), and Slovenia: if any station can be discarded (b; blue; Scenario 5). The countries in this case are considered separately but information exchange between them is taken into account.

4.1.2. Considering the national precipitation stable isotope networks of Slovenia and Hungary assuming information exchange between them (Scenarios 4–7)

In reality, the information provided by the precipitation stable isotope monitoring networks of Slovenia and Hungary extends across the national borders (Fig. 3), and it can be exchanged by the operating research institutes. Thus, any change in either may result in a change in the degree to which the other country is "covered" by its own network, especially in the case of a very small, geomorphologically and climatologically diverse countries like Slovenia. The most pronounced effect is obviously expected near the national borders.

The target of the SSA in this case was again the separate countries, considering the information provided by the other country's precipitation stable isotope monitoring network, nonetheless only taking Slovenia and Hungary into account, i.e. evaluating the RKV relative to the two countries' areas separately. In the case of the Hungarian network, if those Slovenian sites are discarded, the removal of the Murska Sobota precipitation monitoring station, i.e. the one closest to the Hungarian border from Slovenia (<20 km), has a noticeable effect on the precision ($\Delta\sqrt{RKV} = 0.114‰$) of δ^{18}O estimation in Hungary (Fig. 6a; Scenario 4). If the Slovenian network is in the focus, only the removal of the Farkasfa station located ~4 km from the Hungarian border with Slovenia has a similarly noticeable effect ($\Delta\sqrt{RKV} = 0.1‰$) on the RKV of precipitation δ^{18}O estimations for Slovenia (Fig. 6b; Scenario 5), as seen above for Hungary (Scenario 4: Murska Sobota).

If a scenario is considered in which specific stations cannot be discarded (Ljubljana and Portorož; Table 1; Scenarios 6 & 7), there is no change in the RKV of δ^{18}O estimation observable across Hungary (in Scenario 7) as compared to a scenario (Scenario 4) in which these stations could be removed. Although the order in which the removal of the stations is suggested from the Slovenian network does not change as a result of keeping these particular stations, the maximum $\sqrt{RKV}$ across Slovenia is kept below 0.67‰ even if these are the only two stations functioning (this scenario is not shown in a figure).

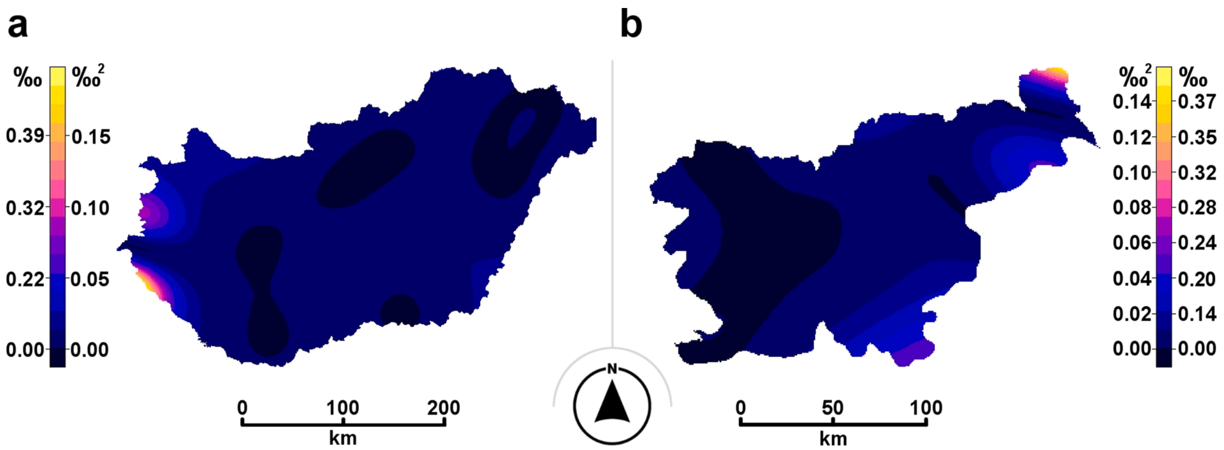

Fig. 7. Difference maps of RKV from between the scenarios when neither (Scenarios 1 & 2) and both the Hungarian and Slovenian precipitation stable isotope monitoring networks have information exchange (Scenarios 4 & 5). The map showing the difference between Scenario 4 and Scenario 1 **(a)**, and that for Scenario 5 and Scenario 2 **(b)**. The countries in this case are considered separately but information exchange between them is taken into account.

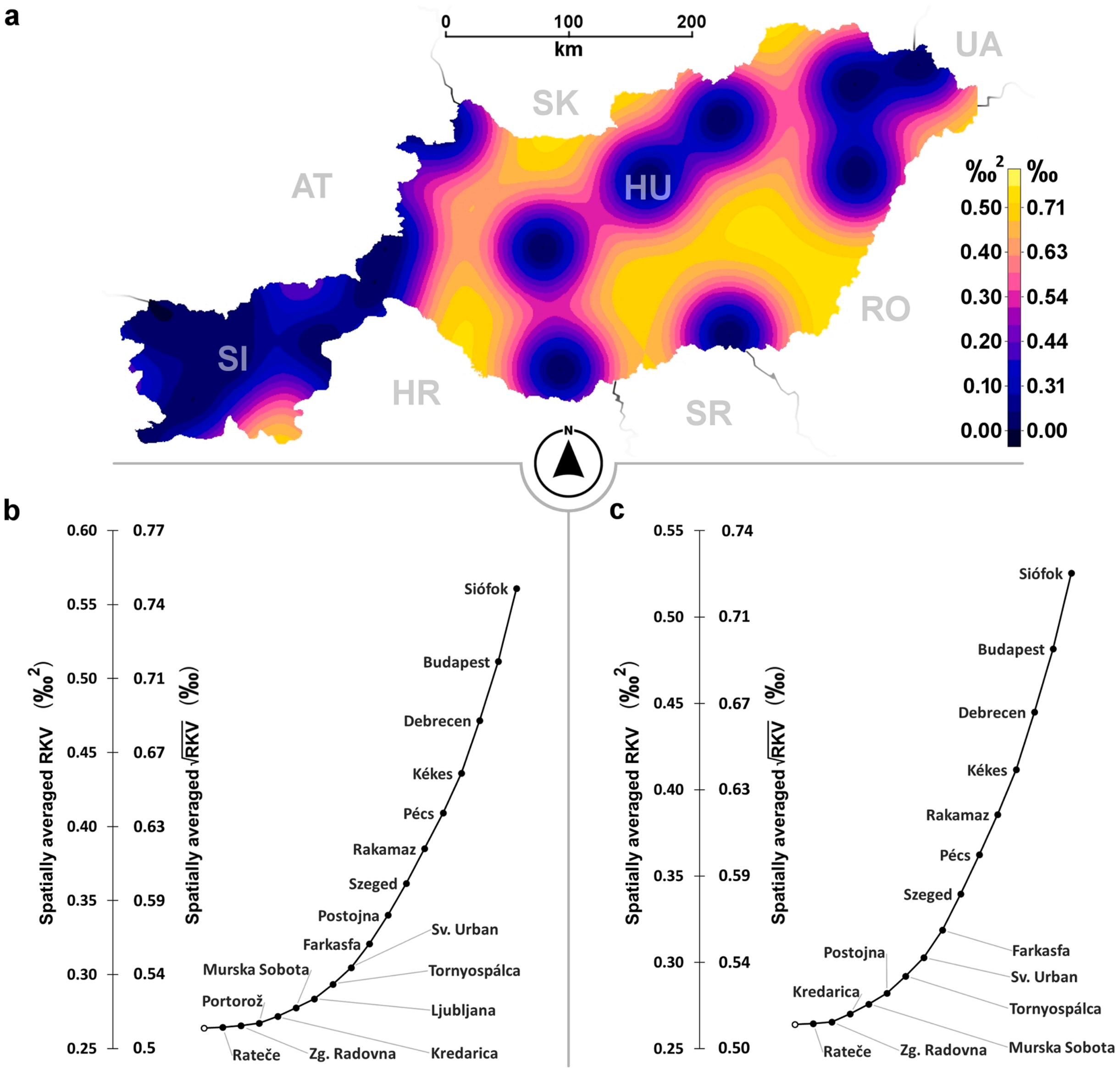

Fig. 8. RVK of the precipitation stable isotope monitoring network of Slovenia and Hungary if those were to be managed together and the neighboring countries' coverage is also considered. Map (a; Scenario 8) and corresponding change in RKV if any station were to be discarded (b; Scenario 8) and if the Ljubljana and Portorož stations must continue operation (c; Scenario 9). Slovenia and Hungary in this case are considered to be fully managed together.

The information exchange between the precipitation stable isotope monitoring networks of Hungary and Slovenia has a minor, but beneficial effect when the whole area of the countries is under consideration (average $\sqrt{RKV} = 0.083‰$; Fig. 7). However, a meaningful amelioration of δ^{18}O representativity ($\Delta\sqrt{RKV} > 0.3‰$) was observed mostly near their common border. In Hungary, an increase in the precision of prediction was in the main confined to a ~10 km band along the border with Slovenia (Fig. 7a) covering two 300 km^2 regions in SW and W Transdanubia, while in NE Slovenia (Prekmurje region) a 200 km^2 area was positively affected by the presence of the Hungarian network at a distance of ~15 km from the border (Fig. 7b). These changes indicate improvements of the prediction model (Hengl et al. 2007) for precipitation δ^{18}O estimations in the studied region.

4.2. Assessing the active precipitation stable isotope monitoring networks of Slovenia and Hungary together, with the influence of the station(s) from neighboring countries (Scenarios 8 & 9)

In geostatistics a traditional measure of the uncertainty of a pattern based on fixed sampling locations is RKV, which is highly dependent on the proper analysis and modeling of spatial correlation structures, and which can be unreliable in the case of insufficient data or a spatial structure which is unreliable (Kanevski et al. 2009). Moreover, it is suggested that, if possible, the information provided by neighboring sites/networks should be exploited, since these can reduce the need to set up new sites (Parr et al. 2002). In the following, an optimal scenario is considered, one in which the Slovenian and Hungarian national precipitation stable isotope monitoring networks are actively cooperating. In addition, the effect of the precipitation monitoring stations operating in the neighboring countries is also taken into consideration. The difference compared to the previous scenarios (Scenarios 4–7) is that in this case the RKVs are evaluated relative to the area of Slovenia and Hungary together (113 000 km^2).

From the RKV map showing the precision of prediction of δ^{18}O values it is clear that the inclusion of the ten eastern Austrian stations decreased the unrepresented areas in the western part of the region (W Hungary and N-NW Slovenia; Fig. 8a), compared to the case in which the countries were evaluated in isolation (Fig. 4). The improved coverage

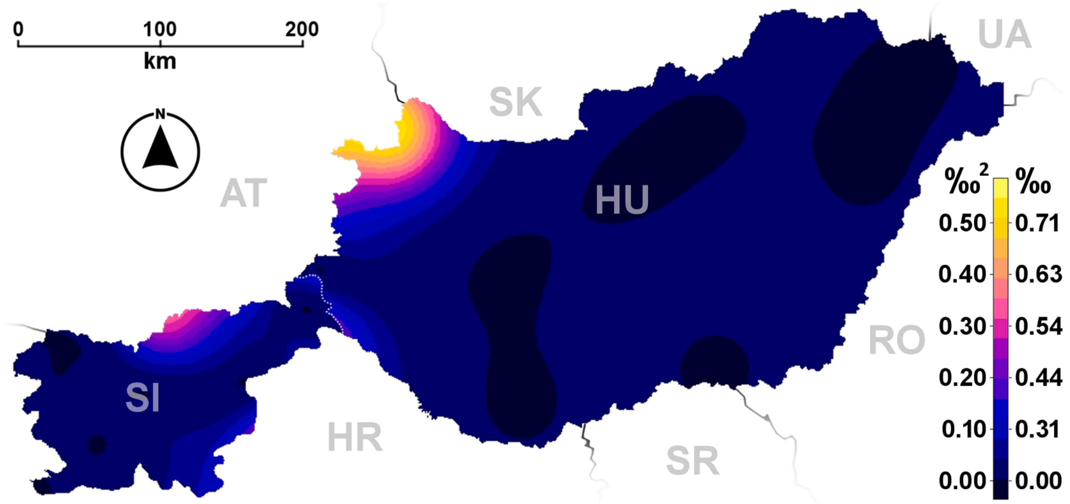

Fig. 9. Difference map of RKV when neither Hungary and Slovenia's precipitation stable isotope monitoring networks have information exchange (Scenarios 1 & 2), and the situation when these have information exchange between them and the neighboring countries (AT and HR), as well (Scenario 8). The map shows the difference in RKV between Scenario 8 and $\mathrm{Scenario1} \cup \mathrm{Scenario2}$. The border between Hungary and Slovenia is marked with a white dotted line. The break in the isolines seen at this border is a technical artefact, because when this difference map of RKV was derived, the maps of Scenario 1 and Scenario 2 were stitched together and compared to the map of Scenario 8.

when the Austrian stations are included (Fig. 3) has a beneficial effect on the Slovenian-Hungarian network. The positive effect of the Zagreb station from Croatia (Krajcar Bronić et al. 2020) on the representativity of the joint Slovenian-Hungarian network is minor compared to that of the Austrian ones, and mainly manifests itself in the southern parts (Fig. 8a).

The SSA results suggest that if (i) any station could be omitted from the network (Scenario 8), then the Rateče and Zgornja Radovna stations would be again the first candidates, since their removal would result in an almost unnoticeable change in the $\sqrt{RKV}$ of δ^{18}O estimations (<0.04‰; Fig. 8b), implying an insignificant information loss compared to the zero-state of the precipitation δ^{18}O monitoring network of Slovenia and Hungary taken together (Fig. 8a).

Prior to the removal of a station of any hydrological monitoring network, its importance should be investigated taking into account the key factors of the *post-hoc* stratification of any environmental monitoring network (see Section 1 and e.g. Parr et al. 2002). It should be taken into consideration that both Rateče (synoptic national meteorological station) and Zg. Radovna (national precipitation meteorological station) are located in different alpine valleys with variable climatic conditions. As such, they are of high climatological and environmental value (Beard et al. 1999; Ogrin and Kozamernik 2020; Torkar et al. 2016), and can also be considered long-term integrated monitoring stations that have been providing isotope in precipitation data since 2010 (Vreča and Malenšek 2016). Thus, their conservation is of great importance in terms of their providing valuable continuous information on the spatio-temporal variations of long-term precipitation (Ogrin and Kozamernik 2020; Parr et al. 2002), which is required for a variety of fields, e.g. water resources (Torkar et al. 2016), civil engineering, agriculture, forestry, hydrology (Frei and Schär 1998) and climatology (Widmann and Schär 1997). According to the change in RKV, the next station that could be removed is Kredarica. However, this is the highest meteorological station in the studied network (Table A1), and also the highest mountain synoptic station in Slovenia with long-term meteorological observations, having ongoing stable isotope monitoring since 2010 (Vreča and Malenšek 2016). It is particularly important to have such elevated stations to obtain a robust empirical estimate for the regional 'altitude' effect (Kern et al. 2020a). All three stations are very specific due to their location in the vulnerable karstic alpine catchment area, with precipitation amount of up to > 3200 mm and which has important influence on water resources (Ogrin and Kozamernik 2020).

If (ii) the Ljubljana and Portorož stations cannot be omitted, and the effect of the neighboring Austrian and Croatian stations is considered, then the order of the first couple of stations that could be removed does not change (from Rateče up to Murska Sobota; Fig. 8b,c). As a consequence of the preservation of the two Slovenian stations (Ljubljana and Portorož), the effect of the possible removal of the data provided by the Postojna ($\Delta\sqrt{RKV} = 0.24$‰) and Pécs ($\Delta\sqrt{RKV} = 0.216$‰) stations decreases relative to that of the other stations between Scenario 8 (Fig. 8b) and Scenario 9 (Fig. 8c).

4.3. Added value from the cooperation between neighboring countries' precipitation stable isotope monitoring networks vs. standalone operation

The areal average precision ($\sqrt{RKV}$) of estimating precipitation δ^{18}O values increased by ~0.2‰ for the study area when the two countries were considered together with neighboring stations. However, along the borders of Hungary and Slovenia with their neighboring countries (Scenario 8) and each other, the improvement in $\sqrt{RKV}$ was well above average (Fig. 9). In 7.1% of the total area, the precision of the estimation increased by 0.32‰. This was mostly restricted to the NW corner of Hungary (in the vicinity of the Seewinkel), and in a ~60 km band exceeded ~0.63‰ along the border with Austria. There were also parts of Slovenia where the precision of estimating precipitation δ^{18}O estimation was improved. There was improvement in the northern part (Koroška region; $\sqrt{RKV} > 0.55$‰) and a noticeable one in a narrower band at the SW border with Croatia (Dolenjska region; $\sqrt{RKV} > 0.32$‰) (Fig. 9). The larger improvement along the Austrian border was to be expected, since there are numerous Austrian monitoring stations situated closer than 15 km from the border of the study area, and are therefore closer than Zagreb, which is more than 20 km from the SW Slovenian border (Table A1).

Many approaches of different complexity and design have been applied to optimize/extend the spatial functioning of precipitation monitoring networks, among them ones based on probabilistic GIS e.g. Shafiei et al. (2013), complex framework e.g. Chang et al. (2016); and geostatistical approaches (e.g. Agou et al. (2019); Barca et al. (2008); Goovaerts (2000), including spatial annealing (e.g. Feki et al. (2016); Pardo-Igúzquiza (1998)). The spatial simulated annealing algorithm itself is a straightforward technique for optimization problems, and has been widely used for finding the best sampling design or monitoring network (Baume et al. 2011; Romary et al. 2011; Szatmári et al. 2016; Szatmári and Pásztor 2019). The application of RKV as a pre-survey quality measure is highly recommended, since it can be derived without knowledge of the exact data values, i.e. it can be computed before the actual sampling takes place, making it a cost-effective pre-survey quality measure (Lark and Lapworth 2012). Besides, KV provides information on the geometric error of the sampling design or the monitoring network, which can be used to express uncertainty if it is reasonable to hold the assumption of normality and homoscedasticity (Szatmári and Pásztor 2019).

Environmental monitoring networks are primarily designed to detect variables of interest to human or ecosystem health and are primarily limited by budgetary constraints (Melles et al. 2011; Nunes et al. 2004). In the case of already functioning networks similar to the precipitation stable isotope monitoring networks in Slovenia and Hungary which were the subject of this study, their optimization consists of two main budgetary factors: investment, in the case of new sites, and exploration, that is, the continuous operation of ongoing sites. The latter is less costly, but both have to be considered during the actual optimization of the network(s).

Data from precipitation stable isotope monitoring can serve as a useful input in hydrological models (Sprenger et al. 2016), and the accuracy of the results yielded by the model strongly depends on the input data; this is why precipitation monitoring network optimization is of great importance (Feki et al. 2016). In the present study, despite the presence of some degree of redundancy (Fig. 3), professional considerations rule out any suggestion of the removal of any of the monitoring stations functioning in 2018 in Slovenia and Hungary. Indeed, if possible, new stations should be set up in the (S)E and NW parts of Hungary and the E part of Slovenia to improve the precision of estimating precipitation stable isotope values in the region yet further. The latter two would assume yet greater importance if the neighboring networks were for any reason to stop operating in future.

5. Where to set up additional precipitation stable isotope monitoring site(s) in Slovenia and Hungary?

The question of the optimal location for additional sampling site(s) to be set up if the possibility arises remains to be addressed. In the case of Hungary, if one new site is installed, it should be placed in the northwest part of the country (47.469°N, 17.355°E; Fig. 4a), while a second one in eastern Hungary (47.009°N, 20.498°E; Fig. 4a) would also be a valuable addition. The addition of one or two sites would increase the mean accuracy of the estimation of precipitation stable isotopes by 5% and 11%, respectively (areal average $\sqrt{RKV}$ for Hungary).

In Slovenia, the most appropriate location for an additional site would be in the south-eastern part of the country, close to its border with Croatia (45.941°N, 15.502°E; Fig. 4b), which would result in a 13% increase in the precision of modeling the variance of stable isotopes in precipitation across Slovenia. Indeed, new stations set up in the NW and (S)E parts of Hungary and the SE part of Slovenia would improve the precision of estimating precipitation stable isotopes in the region yet further.

6. Conclusions

The current research is the first example of the detailed geostatistical assessment of the spatial representativity of a precipitation stable isotope monitoring network. It provides essential information for present and future international co-operation in precipitation stable isotope monitoring network across Slovenia and Hungary by providing an objective overview of the current state of the representativity of the monitoring stations of stable isotopes in precipitation under various scenarios. With variography, the areas that are insufficiently represented, and those that are overrepresented by the current network have been determined. More importantly, the evaluation of the change in regression kriging variance by spatial simulated annealing (SSA) has pointed out that:

(i) the mutual information exchange of the precipitation stable isotope monitoring networks of Slovenia and Hungary increases the precision of estimating precipitation $\delta^{18}O$ values by ~0.3‰ in a 15–30 km band near the borders,
(ii) by taking into account data available from stations in other neighboring countries (Austria and Croatia), the real average precision of estimating precipitation $\delta^{18}O$ values increased by ~0.2‰ across Slovenia and Hungary, and
(iii) there are certain stations which could be discarded on the basis of their modest influence on the overall network: Rateče and Zg. Radovna in Slovenia and Tornyospálca in Hungary, although professional considerations have to be taken into account before any decision is made.

The current network provides an appropriate representation to monitor precipitation stable isotopes across Slovenia and Hungary; however, to increase the precision of estimating precipitation stable isotopes by at least 10%, one additional station is required in Slovenia, and two in Hungary.

On the basis of an extended regional network, the community would be able to establish more realistic isoscapes of precipitation for this part of Europe. Nevertheless, in the future, when new/additional data representing a longer timespan become available, the precipitation stable isotope monitoring network design can and should be re-evaluated.

CRediT authorship contribution statement

István Gábor Hatvani: Conceptualization, Investigation, Data curation, Formal analysis, Writing - original draft. **Gábor Szatmári:** Methodology, Investigation, Software, Writing - original draft. **Zoltán Kern:** Conceptualization, Funding acquisition, Supervision, Writing - original draft, Investigation, Project administration. **Dániel Erdélyi:** Data curation, Formal analysis, Visualization. **Polona Vreča:** Funding acquisition, Supervision, Project administration, Writing - review & editing. **Tjaša Kanduč:** Resources, Writing - review & editing. **György Czuppon:** Resources, Writing - review & editing. **Sonja Lojen:** Resources. **Balázs Kohán:** Methodology.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A

See Table A1 and Fig. A1.

Table A1
Basic geographical information of precipitation stable isotope monitoring stations operating in Slovenia and Hungary and in the neighboring countries within a 150 km band with publicly available data for 2018, arranged by country and longitude.

Name	Lat (°)	Lon (°)	Elevation (m)	Country	Source
Farkasfa	46.91	16.309	312	HU	HU-SI cooperation SNN118205 and N1-0054 projects
Siófok	46.911	18.041	108	HU	
Pécs	45.994	18.235	198	HU	
Budapest	47.432	19.187	139	HU	
Kékes	47.872	20.013	1012	HU	
Szeged	46.256	20.09	81	HU	
Rakamaz	48.128	21.47	940	HU	
Debrecen	47.475	21.494	110	HU	
Tornyospálca	48.273	22.177	108	HU	
Portorož	45.475	13.616	2	SI	
Rateče	46.497	13.713	864	SI	
Kredarica	46.379	13.849	2514	SI	
Zg. Radovna	46.428	13.943	750	SI	
Postojna	45.766	14.193	533	SI	
Ljubljana	46.095	14.597	282	SI	
Sv. Urban	46.184	15.591	283	SI	
Murska Sobota	46.652	16.191	186	SI	
Villacher Alpe	46.603	13.672	2164	AT	ANIP; Umweltbundesamt (2019)
Klagenfurt	46.643	14.32	447	AT	
Eisenkappl	46.489	14.584	550	AT	
Graz Universität	47.078	15.45	366	AT	
Karlgraben	47.678	15.56	775	AT	
Gutenstein	47.875	15.886	475	AT	
Gloggnitz	47.675	15.943	440	AT	
Wien Hohe Warte	48.249	16.356	203	AT	
Moosbrunn	48.019	16.464	186	AT	
Podersdorf	47.855	16.835	120	AT	
Zagreb	45.817	15.983	157	HR	Krajcar Bronić et al. (2020)

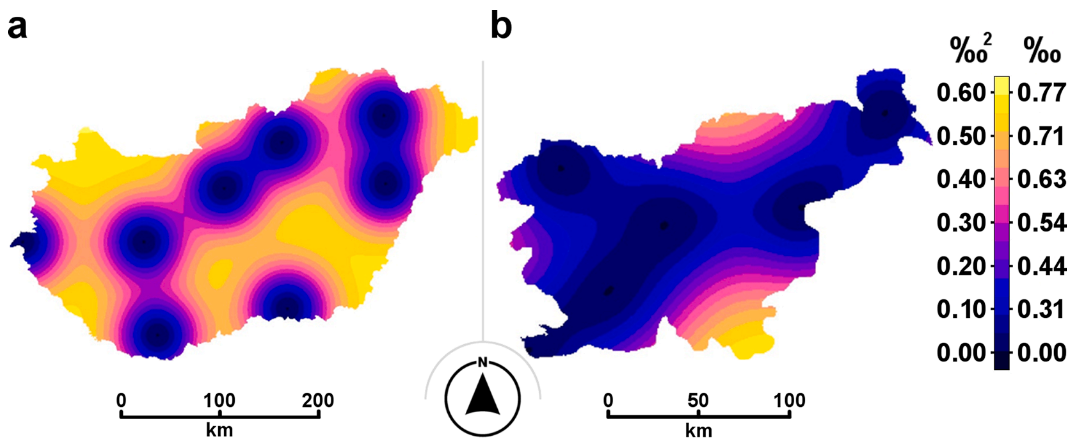

Fig. A1. RKV maps of Hungary **(a),** and Slovenia **(b)** for 2018 under Scenarios 1 and 2, with the omission of sites representing the least valuable information on precipitation δ^{18}O variance in the two countries separately. In the case of Hungary, the map represents the RKV without the information provided by the Tornyospálca station (a), while for Slovenia, the Zgornja Radovna and Rateče stations have been omitted. The countries in this case are considered separately without any information exchange between them.

Variography and kriging

The δ_p residuals obtained by multivariate regression (Section 2.3) were used as the input for the empirical semivariograms (Webster and Oliver 2008), which serve as the weighting function in kriging (Cressie 1990; Molnar 1985; Molnár et al. 2010). The empirical semivariogram may be estimated using Matheron's method-of-moments algorithm (Matheron 1965), where $\gamma(\mathbf{h})$ is the semivariogram and $Z(\mathbf{x})$ and $Z(\mathbf{x}+\mathbf{h})$ are the values of a parameter sampled at a planar distance $|\mathbf{h}|$ from each other

$$\gamma(\mathbf{h}) = \frac{1}{2N(\mathbf{h})}\sum_{i=1}^{N(\mathbf{h})}[Z(\mathbf{x}_i) - Z(\mathbf{x}_i+\mathbf{h})]^2 \tag{A2}$$

$N(\mathbf{h})$ is the number of lag-h differences, i.e. $n\times(n\text{-}1)/2$ and n corresponds to the number of sampling locations along a separation vector h. The most important properties of the semivariogram are (i) the nugget (c_0), a discontinuity at the origin of the semivariogram relating to measurement error and/or small-scale variation (Goovaerts 1997), (ii) the sill – that is, the level at which the semivariogram stabilizes, which is the sum of the nugget (c_0) and the partial sill (c), and (iii) the range (a), which is the distance within which the samples have an influence on each other and beyond which they are uncorrelated (Chilès and Delfiner 2012).

For geostatistical modeling (e.g. kriging), theoretical semivariograms have to be used to approximate the empirical ones (Cressie 1990). In this study, a Gaussian semivariogram was obtained with a maximum lag distance of 504 km and 14 uniform bins (steps) with a minimum number of pairs in a bin = 5 in order to achieve the most balanced number of station pairs per bin in the analysis. The effective range (a_e), which is the distance within which the samples have an influence on each other and beyond which they are uncorrelated (Chilès and Delfiner 2012), was determined and used to evaluate the spatial representativity of the network.

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