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Date: 19.05.2020
Subject: **Submission of revised MS**

Dear Prof. Klump,

Please find enclosed our revised manuscript now entitled: '*Isoscape of precipitation amount-weighted annual mean tritium (^{3}H) activity from 1976 to 2017 for the Adriatic-Pannonian region - AP^{3}H_v1 database*.

Please note that during the revision an additional colleague has contributed to the study (Miklós Süveges) and has been added as an author to the title page and the 'Author contribution' section as well. In adiition, considering the contribution to the revision the author-order has been changed as well.

On behalf of the team of authors I state here that the material in the manuscript is new work none of the submitted material has been published or is under consideration elsewhere, including the Internet, and will not be submitted to any other journal while the present manuscript is under consideration at ESSD.

If you have any questions regarding the enclosed materials, please do not hesitate to contact me.

Thank you for your editorial guidance.

Yours sincerely,

István Gábor Hatvani

https://www.earth-syst-sci-data-discuss.net/essd-2019-244/#discussion

Anonymous Referee #1

Received and published: 15 March 2020

The manuscript describes a high-resolution gridded dataset for tritium in precipitation across the Adriatic-Pannonia region in Europe. I am not familiar with the applications of tritium for hydrology, my expertise is on geostatistical methods for hydrological sciences.

The objective of the work is clearly stated in the abstract and in the introduction. Material and methods are described in detail in section 2. The data sources used are properly reported. Standard statistical techniques, such as ordinary kriging, have been used for spatial analysis. Pros and cons of the applied statistical methods are discussed in detail, especially in connection with the scarcity of data available. Section 3 described the gridded dataset. Section 4 contains the evaluation: the regional dataset has been compared with global ones, the benefits are clearly highlighted; a validation against independent measurements is also included (see Fig.5). All the presented results support the conclusions of the authors presented in section 5.

The contribution of this study for regional hydrological applications is valuable, since the uniqueness of such a reference and up-to-date dataset. Given the limited amount of stations available, the creation of a gridded dataset is totally justified and can provide useful data where no direct measurements are available. The statistical analysis is, as far as I can judge, without major flaws. The presentation of the manuscript is clear and concise.

In conclusion, the study is valuable. My advice to the editor is to publish the manuscript after minor adjustments to the text.

We would like to thank Reviewer#1 for her/his positive opinion on our work and for the constructive comments.

Specific comments follow.

Comment-1: Why use such a high-resolution 1x1 km grid when the planar distances (Fig.3) are hundreds of kilometers? By using this grid, the authors implicitly persuade the users that the information is available on a very local scale. This is not the case. The authors need to (1) justify their choice of a 1x1 km grid; (2) explicitly state that their gridded dataset is suitable for the representation of variations in the field over much larger spatial scales than the grid spacing.

Response-1: Thank you for the suggestion. The 1×1 km grid resolution was chosen based on practical considerations, it does not aim to imply that there are such fine km-scale differences, yet help the users to delineate smaller outcrops (e.g. watersheds) more accurately. This explanation has been added to the MS, see the track changes version in lines 220-225.

C-2: The authors apply kriging without showing that the input data satisfies the prerequisites for a direct application of ordinary kriging. However, the validation shows that the output is useful and -in a sense- this justifies the application of kriging. My

question for you is: have you considered other statistical interpolation methods? What is the reason that made you choose kriging?

R-2: To further reinforce the Reviewer's opinion on that the verification employed in the study is convincing two additional stations have been included as out-of-sample verification (please see Fig. 5). In addition, the discussion has been extended and the MS reorganized.

An additional checking was performed on the amount weighted annual means using h-scattergrams (Bohling, 2005) which did not find any outliers that have been introduced by the weighting procedure confirming that the dataset satisfies certain prerequisites of kriging. This explanation has been added to the track changes version of the revised manuscript in lines 160-164.

The deviance from normal distribution in the case of the ^3H values was found negligible; thus kriging can be applied confidently to the data. Moreover, despite kriging is known to smooth the data, thus decrease the range of the actual values, in the present case the extremities (positive or negative) were not the main subject of the analysis, rather the mid 60% of the data. This was one of the main reasons for choosing kriging to investigate the large-scale patterns, which kriging is highly applicable for (e.g. Wackernagel, 2003 or Chilès and Delfiner, 2012).

C-3: Figure 3. This is perhaps the core result of the paper and I like very much the way the authors present it. However, the blue shades in the colour scale are by far not optimal in representing the fields. Please present your main results in a way that the readers can fully appreciate them.

R-3: Fig. 3 has been substantially changed in the revised version. We hope that the more complex color scale (white-blue-red-yellow) sufficiently improves the contrast in the map series. See Fig. 2 in line 250.

https://www.earth-syst-sci-data-discuss.net/essd-2019-244/#discussion

Anonymous Referee #2
The authors have found a good way to fill in gaps in the existing information by creating a statistical model which uses the 3H data collected from Austria and the northern Balkans. The manuscript presents a time-based 3H precipitation isoscape of North-Balkan. The text is well-structured and easy to read. However, I do have some comments.
We would like to thank Reviewer#2 for her/his positive opinion on our work and for the constructive comments.

Comment-1: Key words: why do the authors only mention Slovenia and Hungary and not the northern Balkans as the title suggests?
Response-1: Our intention was to not repeat the words in the title among the key words, but mentioning the countries covered entirely by the developed database.

C-2: The purpose of this work is not only to create a database, but also to analyse and draw conclusions. I would recommend to slightly expand the purpose in the introduction.
R-2: The introduction (lines 82-85) and the discussion have been extended and latter restructured during the revision, please see track changes version.

C-3: The authors need to explain why they have used the 1×1 km grids. This seems an unreasonable accuracy compared to the size of the study area.
R-3: The 1×1 km grid resolution was chosen based on practical considerations, it does not aim to imply that there are such fine km-scale differences, yet help the users to delineate smaller outcrops (e.g. watersheds) more accurately. This explanation has been added to the MS, see the track changes version in lines 220-225.

C-4: "..the largest shallow freshwater lake in Central Europe". Wouldn't the 'largest lake in Central Europe' already be enough?
R-4: We accept that "freshwater" can be omitted however we would like to keep "shallow". We think this information is useful because the 2 to 6 yrs residence time for the water mentioned in the next sentence is understandable if the lake is shallow however might be weird if someone image a deep lake based on the shortened term "largest lake in Central Europe".

C-5: It is hard to follow the isoscape in Figure 3: I would recommend to use red-blue instead of the current green-blue combination to improve the contrast.
R-5: Accepted. This figure has been revised substantially. We hope the applied more complex color scale (white-blue-red-yellow) does improve the contrast in the map series sufficiently. See Fig. 2 in line 250.

C-6: The used data base (https://doi.pangaea.de/10.1594/PANGAEA.896938) is presented in a less used format (my computer required additional software to read it). Wouldn't it

be possible to present it in the HTML format to make it more usable? An example: https://doi.pangaea.de/10.1594/PANGAEA.911474?format=html#download .

R-6: The database can be considered as temporal sequence of 3D data quite similar to meteorological data. The chosen netCDF format is a common and popular file format in meteorology. The suggested example (https://doi.pangaea.de/10.1594/PANGAEA.911474) is a time series from a single station so, we think, it is not an applicable analogue to the presented dataset. To facilitate the usage of the data set an R-script was already provided in the supplement. In addition, we have expanded the "Data format and availability" section with a sentence in which we provide a link to the freeware tool of NASA (Panoply: https://www.giss.nasa.gov/tools/panoply/) with which interested readers can visualize and inspect of the netCDF files of annual grids (line 483)

C-7: There seems to be some confusion with the parentheses. I've highlighted these in the attached file.

R-7: We have carefully formatted the citations in the main text to correct the superfluous parentheses. The missing spaces between values and dimensions are also corrected at each highlighted place.

Isoscape of precipitation amount-weighted annual mean tritium (^{3}H) activity from 1976 to 2017 for the Adriatic-Pannonian region - AP^{3}H_v1 database

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Abstract. Tritium (^{3}H) as a constituent of the water molecule is an important natural tracer in hydrological sciences. The anthropogenic tritium introduced into the atmosphere became unintentionally an excellent tracer of processes on the time scale of up to ~~a~~ 100 years. A prerequisite for tritium applications is to know the distribution of tritium activity in precipitation. Here we present ~~the spatially continuous gridded~~ a database ~~(of~~ isoscapes~~)~~ derived from 41 stations for amount-weighted annual mean tritium activity in precipitation for the period 1976 to 2017 on spatially continuous interpolated 1×1 km grids for the Adriatic-Pannonian Region (~~using 39 stations~~called AP^{3}H_v1 database), with a special focus on post-2010 years which are not represented by existing global models. ~~Three~~Five stations were used ~~to check~~for out-of-sample evaluation of the model performance independently confirming its capability to reproducing the spatiotemporal tritium variability in the region. ~~This 'Regional model'~~The AP^{3}H database is capable of providing reliable spatiotemporal input ~~data~~ for hydrogeological application at any place within Slovenia, Hungary and its surroundings. Results also show a decrease in the average spatial representativity of the stations regarding tritium activity in precipitation from ~~~600~~440 km in 1970s when bomb-tritium was still prevailing in precipitation, to ~~~300~~235 km in the 2010s. The post-2010 isoscapes can serve as benchmarks for background tritium activity for the region, helping to determine potential future local increases of technogenic tritium from these backgrounds. The gridded tritium isoscape is available in NetCDF-4 at doi: 10.1594/PANGAEA.896938 (Kern et al., 2019).

Keywords: precipitation, Hungary, Slovenia, geospatial tritium model, tritium isoscape

1. Introduction

Tritium (^3H) is a radioactive isotope of hydrogen (Alvarez and Cornog, 1939) with a half-life of 12.32 years (~~4500~~4,500 ± 8 days, (Lucas and Unterweger, 2000)). Natural tritium is formed mainly by spallation reactions of protons and neutrons of primary and secondary cosmic radiation with atmospheric nuclei, mainly by the interaction of fast neutrons with atmospheric nitrogen (Lal and Peters, 1967). Tritium emission by thermonuclear tests between the 1950s and 1980 enormously exceeded the natural production (Araguas-Araguas et al., 1996; Palcsu et al., 2018). Since that time, tritium emission to the atmosphere from anthropogenic sources (e.g. nuclear industry, medical applications, luminizing industry) corresponds to ~10% of the natural production and influences ^3H content in precipitation mainly at local to regional scales (Araguas-Araguas et al., 1996). Starting from the 1980s, the technogenic tritium became the prevailing anthropogenic atmospheric tritium input signal over the bomb tritium in Central Europe (Hebert, 1990).

Tritium is introduced into the hydrological cycle following oxidation to tritiated water ($^3H^1HO$). Tritium is an excellent tracer for determining time scales for the mixing and flow of waters, and is ideal for studying processes that occur on a time scale of less than 100 years (Kendall and McDonnell, 2012). It proved to be a powerful tool in various applications in hydrological researches (Jasechko, 2019) such as estimating mean residence time for surface water and groundwater (Michel, 1992; Stewart and Morgenstern, 2016; Zuber et al., 2001); dating cave drip waters (Kluge et al., 2010); understanding water circulation/mixing in geothermal (Ansari et al., 2017; Chatterjee et al., 2019) or permafrost settings (Gibson et al., 2016) and many other fields (Eyrolle et al., 2018)~~.~~.

A prerequisite for such applications is either a measured or modelled reference of precipitation tritium activity (Stewart and Morgenstern, 2016). Long-term measurements for precipitation tritium activity are worldwide rare, and even the longest time series are usually intermitted by gaps. In the absence of on-site measurements, either remote monitoring data have to be used as references (Huang and Pang, 2010; Thatcher et al., 1961), or estimations are required. There are several methods to reconstruct precipitation tritium time series for geographical locations (Li et al., 2019). The prediction of the first global model for tritium distribution in precipitation from 1960 to 1986 (Doney et al., 1992) was improved and provided a higher accuracy estimate for precipitation ^3H variations (Zhang et al., 2011) extending up to 2005 called, 'Modified global model of tritium in precipitation (MGMTP)'. Unfortunately, the key parameters of MGMTP only available as isoline maps (Zhang et al., 2011), from which the model's coefficients can be extracted with high uncertainty in a manual way, which leads them to be ambiguous. In addition, the quality of the estimated precipitation tritium activity values by MGMTP after1990 become quite poor (Zhang et al., 2011); for instance in the studied region it produced uninterpretable, negative values (Sect. 4). The most recent global model for precipitation tritium activity covering the period 1955-2010 (Jasechko and Taylor, 2015), used inverse distance weighting for interpolation and its output is available in gridded format. However, this is based only on precipitation

^{3}H activity concentration records of the stations of the Global Network of Isotopes in Precipitation (Rozanski et al., 1991) and it does not represent the most recent decade.

Although, global models are available, due to the differences in tritium activities around the globe, it is beneficial to define local precipitation ^{3}H input curves (Stewart and Morgenstern, 2016). In the northern part of the Balkan region, for instance, it was shown that ^{3}H content in precipitation deviated considerably after 1980 from the Vienna record (Miljević et al., 1992) which is popularly used as remote reference station in hydrological modeling/calculations in the Adriatic-Pannonian region. The quality of such curves is vital for the reliability of a hydrological model outputs when employed as input signal/data in hydrological modeling/calculations (Koeniger et al., 2008; Miljević et al., 1992). Indeed, it has recently been found that the (in)accuracy of the used precipitation tritium time series is the key uncertainty factor for groundwater recharge estimations (Li et al., 2019).

Measurements of precipitation tritium activity in the Adriatic-Pannonian region began in Vienna Hohe Warte in 1961, which is the longest continuously operating station in the world, and in Central Europe (IAEA, 2019). Additional stations started operation in the past ~50 years with frequent interruption in data collection (Araguas-Araguas et al., 1996; Krajcar Bronić et al., 1998; Rozanski et al., 1991; Vreča et al., 2008). The demand in long-term ~~tritium~~precipitation ^{3}H reference time-series in various hydrological/hydrogeological applications across the Adriatic-Pannonian region called forth the use of remote stations (e.g. Gessert et al. (2019); Kanduč et al. (2014); Kanduč et al. (2012)) and/or motivated the derivation of case specific "composite" tritium reference curves, e.g. (Kern et al., 2009); Krajcar Bronić et al. (1992); Ozyurt et al. (2014); Szucs et al. (2015). Derivation of these ad hoc "composite" ^{3}H reference curves usually applied different imputation methods to the 'gappy' time series and/or employed different interpolation techniques. Differences in the absolute values due to methodological differences might seem marginal if the peak concentration of the mid-1960s is used as a time-marker, however if data is used as input in tritium mass balance models in the post-bomb period differences become highly important (Li et al., 2019).

The aim of this study was to create a spatially continuous gridded database for tritium (isoscape) in precipitation across the Adriatic-Pannonian Realm for the decades around the turn of the 21st century with a special focus on the post-2010 which is not covered by the existing global models.

2. Materials and Methods

2.1. Used ^{3}H and precipitation data

An initial dataset was collected with ~~8053~~8450 monthly precipitation tritium activity values from ~~45~~46 stations ~~(GNIP (), ANIP , current project)~~ covering the period from Jan 1961 to Dec 2017 obtained from GNIP (IAEA, 2019), ANIP (Umweltbundesamt, 2019), SLONIP (SLONIP, 2020; Vreča and Malenšek, 2016)~~. To maximize the spatiotemporal density of the~~, and published data (Fórizs et al., 2020; Krajcar Bronić et al., 2020; Mandić et al., 2008; Palcsu et al., 2018)~~set not only the Adriatic-Pannonian region, but the bordering areas were included in the analyses as well. The availability of ^{3}H data varied in the investigated time period. Three time horizons were outlined with a relatively high abundance of data: early 1980s~~

(number of annual data (n_a)≈15), early 2000s (n_a ≈14) and the early 2010s (n_a ≈21) (Fig. .1a). Until 1973 tritium activity data was only available from Austria. Monitoring of isotopes in precipitation on a larger scale in the region began in the mid-1970s in Belgrade (RS), Zagreb (HR) and Budapest (HU) as well. Following the initiation of these measurements becomes the network suitable - specifically from 1976 - for the spatiotemporal analysis of the large-scale variability of precipitation tritium activity in the region. To maximize the spatiotemporal density of the data set not only the Adriatic-Pannonian region, but the bordering areas were included in the analyses as well. The availability of ^{3}H data varied in the investigated period. The relatively abundance of data increased in the early 1980s, early 2000s and from around 2010 onward (Fig. 1a). Between 2003 and 2005, the number of stations dropped (<9, Fig. 1a) due to a halt in the data collection of the Austrian stations. This was the lowest number of active stations in the investigated period. For the purpose of further calculations, the geographical coordinates of the stations were converted from latitude and longitude (EPSG: 4326, WGS84 projection) to metric coordinate system (EPSG:3857, WGS 84 / Pseudo-Mercator projection), since interpolation (variography see Sect. 2.3) has to be done on a metric scale. To be able to derive amount weighted annual tritium activity averages, monthly precipitation amounts were used from the GPCC's (1.0° × 1.0°) Full Data Monthly Product Version 2018 (Schneider et al., 2018).

To be able to derive amount weighted annual tritium activity averages, (0.5° × 0.5°) monthly precipitation amounts were used from the GPCC database , derived as precipitation anomalies at stations interpolated and then superimposed on the GPCC Climatology V2011 .

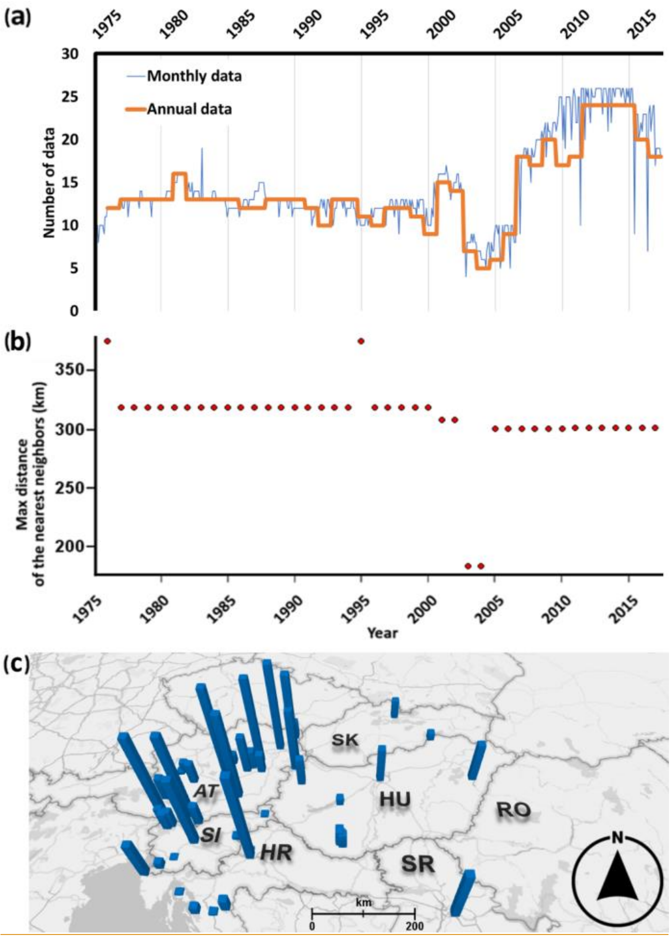

Figure 1:. Temporal and spatial characteristics of the dataset. Number of data from precipitation stations producing measurements of ^{3}H (1975-2017) A). **(a)** The thick orange line represents the number of stations applicable for computing precipitation amount weighted annual averages later used in the interpolation (1976-2017). **(b)** The largest distance between the neighboring active stations of the studied ^{3}H network in each year for 1976-2017 B). The. **(c)** Spatial distribution of the monitoring sites C),, where the height of the blue columns is proportional to the number of monthly data available between 1976 and 2017 at a given station; max= = 479 data at Podersdorf Austria. The

country codes follow the ISO-3166-1 ALPHA-2. The basemap was taken from Bing maps, HERE Technologies 2019; accessed on 27.09.2019.

2.2. Data preprocessing

A sequential univariate outlier detection procedure (Ben-Gal, 2005) was applied to the data to find possible outlying values, which deviate to a high extent from the other observations (Barnett and Lewis, 1974; Hawkins, 1980). During the procedure, the time series of the stations were pairwise compared for each year. The approach is similar to the relative homogeneity test applied to meteorological data, in which e.g. a candidate station's time series is compared to its neighboring stations'; e.g. (Alexandersson, 1986; Lindau and Venema, 2019; Sugahara et al., 2012).

To avoid comparing a station with all the others from the network, including distant ones recording different environmental conditions (e.g. Alpine region vs. Great Hungarian Plain), the comparison was done only within a given search radius. The network was screened for each station's distance to its nearest neighbor for each year. Then out of all the years, the most frequently occurring largest nearest neighbor (~320 km) was chosen (Fig. 1b) to serve as the search radius for the sequential univariate outlier detection. There were ~~15~~only two years when a station – specifically Belgrade - did not have a pair to compare it with~~. In 1976, 1993-2000 and 2003 it was Belgrade , while between 2013 and 2017 it was Debrecen~~ due to ~~their~~its relatively isolated location from the others in the network~~. These are the southeasternmost and northeasternmost stations~~ (Fig. 1c).

Pairwise differences of ^{3}H data in monthly steps were calculated for each station with its neighbors within the ~320 km search radius. These pairwise differences were then averaged per month and the values belonging to the same calendar year were handled together. Due to the decrease in atmospheric concentration in tritium (Palcsu et al., 2018; Rozanski et al., 1991), the difference values were not comparable between the years, so the outliers were identified annually. The monthly average difference values were annually standardized.

It was found that the standardized mean differences were mostly within the ±1 interval (~~82 %; Fig. 2)~~89%) suggesting the usually small difference between neighboring records. In rare occasions (n~~=6~~ = 11 occurrences; 0.~~09~~15%) the difference value was outside the ±7 interval. These deviations were considered as threshold, determining the set of possibly erroneous data (outlier (Ben-Gal, 2005)), which were investigated one-by-one, if possible~~ by consulting the data providers. For example, in Dec 1994 at Zagreb, the standardized differences indicated a possible error (d= -9.33), which coincided with experimental research in the nearby facility in which technogenic tritium was used , thus the sample was excluded from the analysis (~~, by consulting the data providers. Only two monthly values were left out from further calculations both much higher than measured ones compared to their surroundings. ~~Fig. 2).~~

~~**Figure 2**: Histogram representing the distribution of the standardized difference values between the precipitation stations within a ~320 km search radius (1976-2017). The grey shaded background highlights the ±1 standardized difference interval. The standardized difference of -9 (in a red rectangle) corresponds to an outlier measured at Zagreb (Dec 1994); it is shown on the inset map along with the ^{3}H records from its neighbors within the search radius. For further details see text.~~

Annual amount-weighted means were only calculated if at least 85% of the fallen precipitation was analyzed for ^{3}H., these years are herein referred to as "complete-year". If more than 15% of the fallen precipitation was not analyzed for ^{3}H, the year in question will be referred to as an "incomplete-year". This required completeness is stricter criterion than the GNIP protocol (70%; (IAEA, 1992). These amount-weighted annual averages served as the input values for deriving the isoscapes with variography. An additional checking was performed on the amount weighted annual means using h-scattergrams (Bohling, 2005) which did not find any outliers that have been introduced by the weighting procedure confirming that the dataset satisfies certain prerequisites of kriging.

A robust hemispheric-scale pattern is a poleward increasing trend of precipitation ^{3}H (Rozanski et al., 1991). Regression analysis between geographical latitude (using the metric coordinates in EPSG: 3857) and amount weighted-annual precipitation ^{3}H activity concentration mostly yielded insignificant linear relationships or contradictory to what was expected (i.e. poleward decreasing values e.g. 1987). The limited latitudinal extent of the study area (°(5)°) might explain the failure to detect the expected relationship. However, due to the lack of a clear spatial trend statistical trend removal was not conducted on the amount-weighted annual mean ^{3}H activities instead they were used for regional isoscape modeling.

2.3. Derivation of precipitation amount-weighted annual mean tritium activity isoscapes

Semivariograms (Webster and Oliver, 2008) were used as the weighting function in kriging (Cressie, 1990) to explore the spatial variance of precipitation amount-weighted annual mean ^{3}H activity for the stations of the Adriatic-Pannonian region. The empirical semivariogram may be calculated using the Matheron algorithm (Matheron, 1965), where $\gamma(h)$ is the semivariogram and $Z(x)$ and $Z(x+h)$ are the values of a parameter sampled at a planar distance $|h|$ from each other

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_i + h)]^2 \qquad (1)$$

$N(h)$ is the number of lag-h differences, i.e. $n \times (n-1)/2$ and n corresponds to the number of sampling locations at a distance h. The most important properties of the semivariogram are the nugget, quantifying the variance at the sampling location (including information regarding the error of the sampling), the sill that is, the level at which the variogram stabilizes, which is the sum of the nugget (c_0) and the reduced sill (c), and 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). If the semivariogram does not have a rising part and the points of the empirical semivariogram align parallel to the abscissa, a nugget-type variogram is obtained. In this case, the sampling frequency is insufficient to estimate the sampling range using variography (Hatvani et al., 2017).

For geostatistical modeling (e.g. kriging), theoretical semivariograms have to be used to approximate the empirical ones

(Cressie, 1990). Gaussian semivariograms were obtained with a maximum lag distance of 400 km and 11 uniform bins (steps) in order to achieve the most balanced number of station pairs per bin in the analysis. The effective (or even called practical) 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) were determined and used to evaluate the spatial representativity of the network. In the case of Gaussian semivariograms $a_e = \sqrt{3} \times a$)(Wackernagel, 2003). The reported ranges in the study area are planar distances in km; conversion to geodetic distance in the region: d_{planar} × ~~×~~0.678 ≈ $d_{geodetic}$.

~~In a preliminary screening it was found that semivariograms had to have at least 3 station pairs in the first bin and more than 14 pairs in the first 3 bins to be applicable for interpolation; these were the minimum requirements for kriging.~~ Semivariograms ~~perfectly~~ applicable for interpolation were obtained from years 1977, 1982, ~~2007~~1983, 2001, 2010, ~~2011~~ and from 2012~~. The number of active stations in these years varied between 13 and 24. These years where~~ to 2015 which, unsurprisingly, belonged to the periods with a relatively higher abundance of data (Fig. 1a). Common characteristics of the variograms from these years (further on used as the *reference years*~~.~~) were that (i) the number of stations behind the variograms from these years varied between 13 and 24 and (ii) the variograms had at least 11 pairs in the first 3 bins without any empty ones. This seemed a minimum requirement in the present database to derive theoretical variograms suitable for kriging.

The years with a reduced number of available stations (Fig. 1a) produced semivariograms not applicable for kriging (for technical explanation see Appendix 1), because the data were sporadically spread in space and/or none of the stations provided continuous measurements in time. Both types of data gaps can be classified as missing at random (MAR) (Little and Rubin, 2002). Because most modern data-imputation-methods start by assuming the missing data is MAR, imputation tools could have been applied in years with insufficient data density for proper interpolation. However, in every case, no method can provide an 'automatic' solution to the problem of missing data, and any approach must be used with caution considering the context of the problem (Kenward and Carpenter, 2007); for instance, the accuracy of the imputed value will not be optimal and the spatial correlation and intra-variable relationships will be corrupted (Barnett and Deutsch, 2015).

Thus, in these – so called - "intermediate" years, the semivariogram of the reference years having the most overlap with regard to its station distribution, was used as the weight for kriging. To do so, it was investigated for each intermediate year, how many sites are commonly active in its temporally neighboring reference year. The following requirements were also considered:

- the maximum number of sites active in a given intermediate year which are not active in the reference year can be 3
- if the difference in the number of active stations between an intermediate year and the "neighboring" two reference years is the same, then the semivariogram of the reference year with the greater number of active stations was used rendering that variogram more robust,
- and the one closest to the intermediate year in time.

Finally, ~~42~~46 stations were considered for further evaluation out of which ~~39~~41 stations were used for tritium isoscape derivation~~ while three~~. The amount weighted annual tritium activities from these 41 stations were ~~excluded from interpolation and~~ used to derive the gridded (1 × 1 km) annual precipitation ^{3}H isoscapes across Slovenia and Hungary for 1976-2017, called the AP^{3}H_v1 database, using ordinary point kriging with the assigned variograms derived as discussed above. Note here, that the grid resolution was chosen based on practical consideration, it does not aim to imply that there are such fine km-scale differences, yet help the users to delineate smaller outcrops of e.g. watershed areas more accurately.

For out-of-sample verification two stations were withheld from Hungary and Slovenia each: Nick (HU; active: 1990-2004) and Zgornja Radovna (SI; active: 2010-2017) to validate that the interpolated product is useful and -in a sense- justifies the application of kriging in the region. Two additional stations recorded precipitation ^{3}H for multiple, but exclusively 'incomplete-years': Jósvafő (HU; active: 1988-1996) and Malinska (HR; active: 2000-2001) that were not applicable for variography, yet were used to further test the performance of the ~~interpolated products (Table 1): Zgornja Radovna (active: 2010-2017) from Slovenia,~~ geospatial model of precipitation ^{3}H and the critical limit of ratio of annual precipitation amount with missing ^{3}H data. Lastly, an additional short record from Siófok (HU; active: 2013-2016) ~~from Hungary, and Malinska (active: 2000-2001) from Croatia..~~was also excluded from the spatial model to compare it with the interpolated data; see Sect. 4.

All computations were performed ~~using~~with Golden Software Surfer 15, ArcGIS 10, GS+ 10 and R (R Core Team, 2019) ~~GS+ 10.~~using the script in Supplement. For certain visualizations of the results, Gimp 2.8 and MS Excel 365 were used.

Table 1: Sampling sites with basic geographical information used in the study arranged ~~by country alphabetically.~~alphabetically by country codes (ISO-3166-1 ALPHA-2) and station names. The number of monthly precipitation ^{3}H activity concentration data between 1976 and 2017 used in this study is indicated as well as the number of monthly data available in the Global Network of Isotopes in Precipitation in the same period (column title GNIP). The stations below the dashed line were used for model performance testing; for details see Sect. 4.

Country~~Name~~	Station	Latitude	Longitude	Elevation	~~Country~~	No. of ~~monthly~~ data (1976-2017)	GNIP
AT	Apetlon	47.741	16.831	119	~~AT~~	321	
AT	Bad Aussee	47.600	13.783	640	~~AT~~	15	
AT	Eisenkappl	46.489	14.584	550	~~AT~~	106	
AT	Gloggnitz	47.675	15.943	440	~~AT~~	96	
AT	Gößl	47.640	13.901	710	~~AT~~	59	
AT	Graz Universität	47.078	15.450	366	~~AT~~	447	321
AT	Gutenstein	47.875	15.886	475	~~AT~~	447	
AT	Karlgraben	47.678	15.560	775	~~AT~~	193	
AT	Klagenfurt	46.643	14.320	447		~~AT~~445	~~446~~318
AT	Lackenhof	47.870	15.142	882	~~AT~~	51	
AT	Nasswald	47.764	15.688	774	~~AT~~	95	

AT	Planneralm	47.403	14.200	1605	~~AT~~	106	
AT	Podersdorf	47.855	16.835	120	~~AT~~	467	
AT	St. Peter im Katschtal	47.027	13.596	1220	~~AT~~	121	
AT	Villacher Alpe	46.603	13.672	2164	~~AT~~	451	334
AT	Wien Hohe Warte	48.249	16.356	203	~~AT~~	444	421
AT	Wildalpen	47.664	14.978	610	~~AT~~	447	
AT	Zistersdorf	48.544	16.750	201	~~AT~~	88	
HR	Plitvice	44.881	15.619	580	~~HR~~	65	27
HR	Puntijarka	45.908	15.968	988		9	
HR	Tonkovića	44.790	15.368	432		13	
HR	Zagreb[1]	45.817	15.983	157		~~HR~~473	~~133~~325
HR	Zavižan	44.815	14.976	1594	~~HR~~	39	38
HU	B	46.070	18.111	177		104	
HU	Boda	46.087	18.047	233		135	
HU	Budapest	47.464	19.073	101		~~HU~~287	181
HU	Debrecen	47.475	21.494	110	~~HU~~	202	
	~~Met B~~	~~46.070~~	~~18.111~~	~~177~~		~~HU~~	~~41~~
	~~Met Boda~~	~~46.087~~	~~18.047~~	~~233~~		~~HU~~	~~82~~
HU	~~Met~~ Het	46.125	18.047	165		~~HU~~136	~~82~~
HU	~~Met~~ II~~. üz~~	46.100	18.093	332		~~HU~~79	~~27~~
HU	~~Met~~ V~~. üz~~	46.122	18.092	330		~~HU~~60	~~58~~
HU	~~Met~~ Z	46.037	18.125	117		~~HU~~137	~~75~~
RS	Belgrade	44.783	20.533	243		~~RS~~283	283
SI	Kozina	45.604	13.932	486		~~SI~~35	35
SI	Kredarica	46.379	13.849	2514	~~SI~~	91	
SI	Ljubljana[2]	46.095	14.597	282	~~SI~~	371	291
SI	Murska Sobota	46.652	16.191	186	~~SI~~	11	
SI	Portorož	45.467	13.617	2	~~SI~~	196	70
SI	Postojna	45.766	14.198	533	~~SI~~	5	
SI	Rateče	46.497	13.713	864	~~SI~~	88	
SI	Sv. Urban	46.184	15.591	283	~~SI~~	16	
SK	Liptovský Mikuláš	49.098	19.590	570		~~SK~~96	96

Beszúrt cellák
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	Siófok	46.911	18.041	108	HU	39
HR	Malinska	45.121	14.526	1	HR10	10
HU	Jósvafő	48.484	20.551	211	39	
HU	Nick	47.385	17.036	139	142	
HU	Siófok	46.911	18.041	108	39	
SI	Zgornja Radovna	46.428	13.943	750 SI	89	

1: In the investigated period two stations were conducting measurements in Zagreb in a non-overlapping way (Krajcar Bronić et al., 2020)

2: In the investigated period three stations were conducting measurements in Ljubljana in a non-overlapping way (Vreča et al., 2014; Vreča et al., 2008)

Beszúrt cellák

Beszúrt cellák

Törölt cellák

Beszúrt cellák

3. Tritium isoscapes (from 1976- to 2017)

According to The obtained regional gridded precipitation amount-weighted annual mean ^3H activity time series for the Adriatic-Pannonian region (AP^{3}H_v1 database) capture the well-known decrease in precipitation ^3H activity (e.g. Fig. referred to hereinafter as Regional model) the monitoring network provides a proper representativity of the study area (e.g. Fig. 3, in-set maps2).

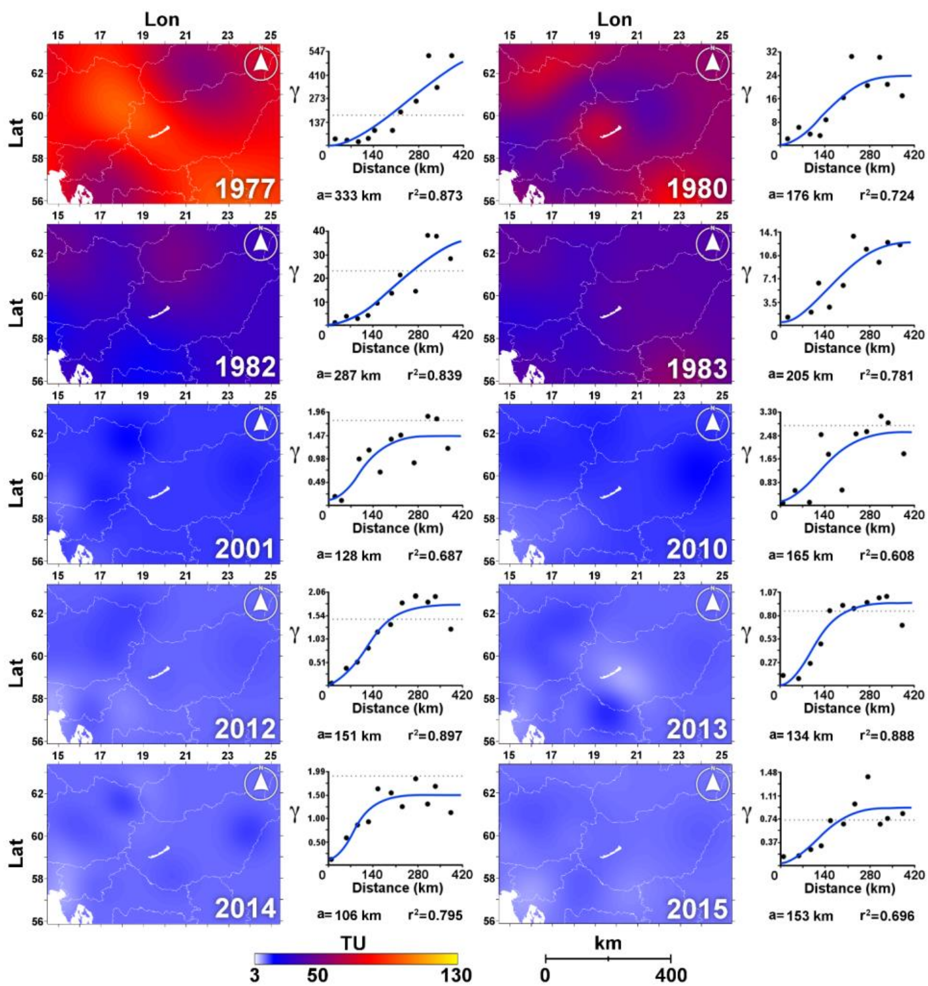

Figure 2 Isoscapes of amount-weighted annual mean ^3H activity (TU) in precipitation and semivariograms for the reference years in the Adriatic-Pannonian Region. The Adriatic Sea and Lake Balaton are marked in white. Isoscape grid resolution: 1 × 1 km. The figures on the right side of the isoscapes show the empirical (black dots) and theoretical semivariograms (blue line) used for kriging along with the obtained ranges (a planar distances in km) and the fit (r^2) of the theoretical semivariograms. The dotted horizontal line indicates the average variance.

~~The~~Besides this most striking long-term temporal pattern ~~(decrease in precipitation ^{3}H activity; Fig. 3)~~ prevailing in the whole region seen from the isoscapes ~~is also reflected in time series of distant locations (Fig. 4). Moreover,~~ ~~The distinctive interannual fluctuation of amount-weighted annual mean ^{3}H activity at Budapest and Ljubljana (Fig. 4) also indicate that the Regional model produced differing sub-regional variability over the modelled time. For instance, the maxima of the modelled~~subregional differences in precipitation ^{3}H activity ~~occurs in 1979 and 1976, while a local minima from the early '90s in 1990 and 1991 at Budapest (Fig. 4a) and Ljubljana (Fig. 4b) are observed, respectively.~~

within the region is also reconstructed. Although no significant relationship was documented between latitude and/or continentality, still increasing precipitation ^{3}H activity was observable inland with the lowest values documented along the Slovenian and northern Croatian coast in all years (see e.g. 1982, 2010, 2014; Fig. ~~3~~2). This pattern can be related to the generally observed lower activity at maritime coastal stations due to the higher contribution of primary marine evaporation practically free from ^{3}H (Eastoe et al., 2012; Rozanski et al., 1991; Tadros et al., 2014; Vreča et al., 2006) and higher contribution of recycled modern meteoric water over the continent. For instance, moisture originating from continental Europe and the Atlantic Ocean was found to be distinct regarding tritium concentrations (8.8 TU and ~0 TU, respectively) (Juhlke et al., 2020)~~.~~.

~~Results~~Two of the longest records from Slovenia (Ljubljana) and Hungary (Budapest) illustrate the performance of the estimations and their potential in mitigating lack of data. Budapest and Ljubljana ^{3}H records – 'complete-years' were used both in the variograms of the "anchor years" and in the interpolation - were compared to the interpolated product's time series of the nearest grid cell (Fig. 3). In the 'complete-years', when the measured values were used in interpolation, there is an expected perfect match between the measured and modelled values. It becomes clear that the estimated records are more than capable in filling the gaps of the measured time series, when there were no measurements (e.g. Ljubljana: 1985 and 1996; Fig. 3b). and usually provide a higher mean annual precipitation ^{3}H in the case of 'incomplete-years'. The magnitude of differences between the measured and modeled data in the 'incomplete-years' varied between ~16.5 and 1 TU for Budapest and Ljubljana, with a general tendency of obtaining higher differences with a higher ratio of precipitation not represented by tritium measurements.

The distinctive interannual fluctuation of amount-weighted annual mean ^{3}H activity at Budapest and Ljubljana (Fig. 3) also indicate that the AP^{3}H database produced differing sub-regional variability over the modelled time. In the whole investigated period (1976-2017) the coefficient of variation of the annual differences between the precipitation ^{3}H values of the 'complete-years' in Budapest and Ljubljana was 62%, calling for the need for spatially representative estimates, such as the ones presented from the AP^{3}H database. In the period when both stations were parallel active (1981-2003, excluding the 'incomplete-years') the precipitation ^{3}H activity in Budapest was higher by 5.5 TU than in Ljubljana on average, and this difference is quite well reflected by the AP^{3}H data which were 4.57 TU higher at Budapest on average. Besides these overall differences, distinct interannual variability can be observed at the Ljubljana and Budapest stations. For example, the maxima of the modelled precipitation ^{3}H activity occurred in 1976 at the Ljubljana- and in 1978 at the Budapest station, and a minor peak seen in 1988 at Budapest (Fig. 3a) was without a counterpart in the Ljubljana records (Fig. 3b).

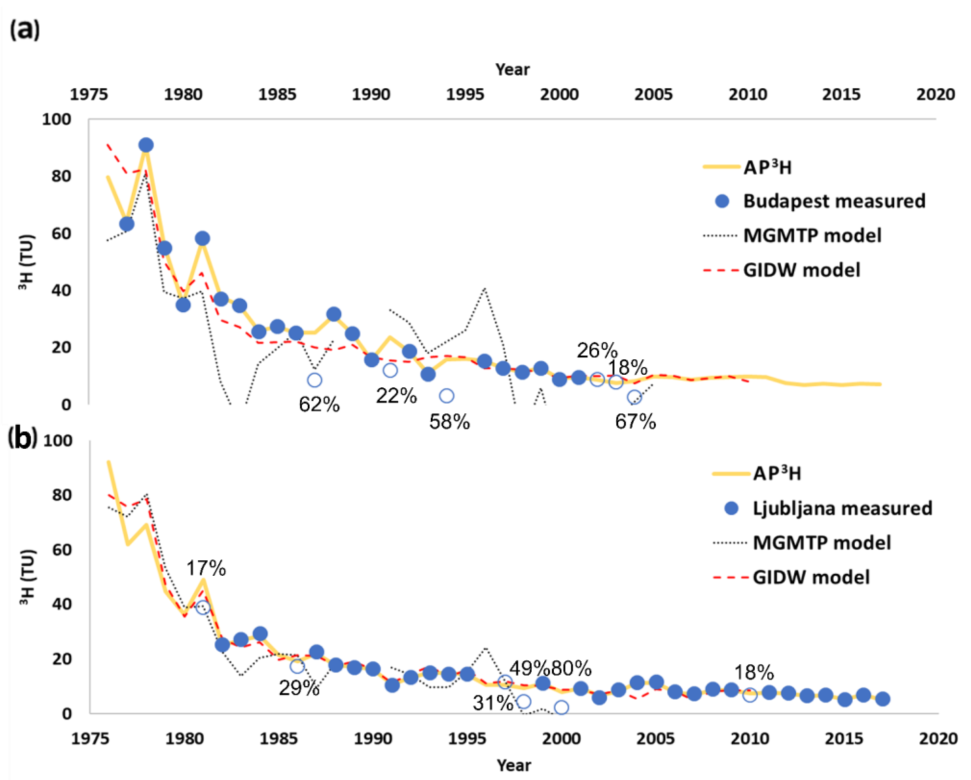

Figure 3 Measured and estimated ^{3}H values at **(a)** Budapest, Hungary and **(b)** Ljubljana, Slovenia between 1976 and 2017. The black dotted lines indicate the estimations of the 'modified global model of tritium in precipitation' (MGMTP; (Zhang et al., 2011)) and the red dashed ones indicate the Global inverse distance weighted model (GIDW; (Jasechko and Taylor, 2015)). Note here that uninterpretable negative estimates of MGMTP were not shown. The empty circles indicate an 'incomplete-year' in which the given ^{3}H value was not used for interpolation in the AP^{3}H database and the percentages next to these symbols indicate the ratio of fallen precipitation not analyzed for ^{3}H in a given year.

The estimated effective range show a decrease in the spatial autocorrelation of tritium activity concentration of precipitation from the 1970s to the 2010s (Fig. 3): ~600: ~440 km in the 1970s, ~450425 km in the 1980s, to ~300360 km and 235 km in the 2000s and 2010s. respectively (Fig. 4). This period (1970-2010) was characterized by the removal of bomb-tritium from the atmosphere (Araguas-Araguas et al., 1996; Palcsu et al., 2018). The overwhelming activity of bomb-produced ^{3}H was several orders of magnitude higher than the natural background (Rozanski et al., 1991), and largely masked the smaller-scale natural variability. During last 2-3recent decades the tritium activity in precipitation has declined globally and regionally, approaching the natural pre-bomb level andlevels indicating that the bomb-tritium is now barely present in modern precipitation. Since, in the Adriatic Pannonian region, the ^{3}H activity in precipitation approached natural levels by the early 1990s2000s (Krajcar Bronić et al., 2020; Palcsu et al., 2018; Vreča et al., 2008), it can be expected that the ~200-300 km range obtained for the 2010s reflects the range of similarity of natural ^{3}H variability in the study area (SE Europe and E Central Europe). Regarding spatial coverage, the northwestern part of the region was much more represented in all years, due to the expected denser station network along the Austrian border with Slovenia and Hungary (Fig. 1c; Fig 3).

Figure 3: Isoscapes of ^{3}H activity (TU) and semivariograms for the reference years (upper panels: 1977, 1982, 2007; lower panels: 2010, 2011, 2012) in the Adriatic-Pannonian Region. The areas outside the union of the range ellipses of a given year are dimmed and the Adriatic Sea marked in white. Isoscape grid resolution: 1 × 1 km. Easting and northing in 10^{5} km. The inset figures show the empirical (empty black squares) and theoretical semivariograms (blue line) used for kriging along with the obtained effective ranges (a_e planar distances in km) and the fit (r^2) of the theoretical semivariograms. The dotted horizontal line indicates the average variance.

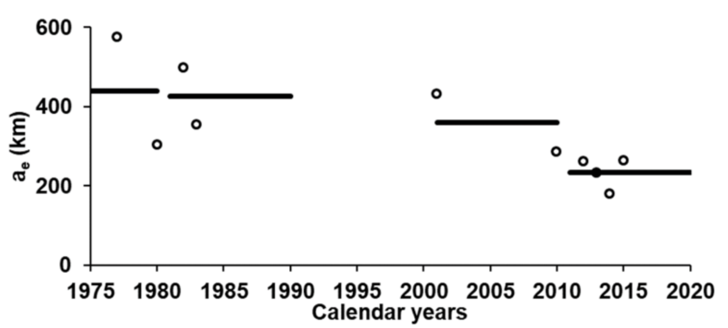

Figure 4 Effective ranges (a_e) of the semivariograms used for kriging. Annual ranges are indicated by empty circles, and the decadal averages by horizontal black lines.

4. Verification of goodness of interpolation

~~Two of the longest records from both Slovenia (Ljubljana) and Hungary (Budapest) illustrate the performance of the estimations and their potential in mitigating lack of data. Budapest- and Ljubljana ^{3}H records - used both in the variograms of the "anchor years" and in the interpolation - were compared to the interpolated product's time series of the nearest grid cell (Fig. 4). In the years~~The performance of the AP^{3}H database replicating measured precipitation ^{3}H activity was tested via out-of-sample verification. The actual amount-weighed annual mean precipitation ^{3}H activity time series at a Hungarian (Nick, Fig. 5a) and a Slovenian (Zgornja Radovna, Fig. 5b) station were compared to the AP^{3}H data of the grid cell closest to the specific stations. The annual mean precipitation ^{3}H activity estimates from the AP^{3}H database nicely fitted to the amount-weighed annual mean precipitation ^{3}H activity data for both independent records even if a small portion (<8%) was not represented with measured ^{3}H in the experimental data (Fig. 5a,b). The agreement between the independent mean annual precipitation ^{3}H record and the AP^{3}H estimates is superb for Zgornja Radovna where the difference between modeled and actual values ranged from 1 to 6% (mean: 3.2%). The difference between modeled and actual values in 'complete-years' for Nick ranged from 0 to 31% (mean: 8.3%). The relatively larger mean error for the Nick record is due to an overestimation of the model values in 1994 (Fig. 5a); however, the model data fit to the rest of the measured ones as good as seen for Zgornja Radovna. The pattern observed for the 'incomplete-years' strengthens previous impressions that (i) modeled mean annual precipitation ^{3}H is higher than the mean calculated from just a few monthly measurements and (ii) there is a tendency of obtaining higher differences with a higher ratio of precipitation not represented by tritium measurements.~~ when the measured values were used in interpolation, there is an expected perfect match between the measured and modelled values. It becomes clear that the estimated records are more than capable in filling the gaps of the measured time series, when there were no measurements (e.g. Ljubljana: 1985 and 1996; Fig.~~ The comparison between the modeled and actual values for the stations recorded precipitation ^{3}H for multiple, but exclusively 'incomplete-years', further strengthens these impressions.

Station records from Nick and Zgornja Radovna were not used in the interpolation to derive the AP^{3}H database, yet perfectly match the actual values when ^{3}H activity concentration was measured for the total volume of annual precipitation. This excellent agreement supports the assumption that the AP^{3}H database provides similarly accurate estimates as at other stations for the 'incomplete-years', as well. If so, then differences between the AP^{3}H data and the mean precipitation ^{3}H activity estimates accompanied with certain portion of precipitation amount lacking ^{3}H activity can represent a weighting bias due to the fragmented observation record. Comparing the differences between the estimated mean precipitation ^{3}H activity values from these fragmented records and the AP^{3}H estimates to the percentage of precipitation amount lacking ^{3}H activity data in the so called 'incomplete-years' for Zgornja Radovna, Nick, Jósvafő, and Malinska reveled the relation between these characteristics (Fig. 5e). If ^{3}H activity was lacking for less than 15% of the annual precipitation total the difference between the calculated mean precipitation ^{3}H activity values and the AP^{3}H estimates is practically negligible (Fig. 5e). The difference between modeled and calculated mean precipitation ^{3}H considerably increased (> 6 TU) if the proportion of the precipitation lacking ^{3}H activity was between 15% and 30% of the annual precipitation total (Fig. 5e). According to the GNIP protocol the required completeness is 70% of the precipitation total for calculating amount-weighted mean isotopic values for a certain period (IAEA, 1992). Above results (Fig. 5e) confirm that 30% missing data is indeed a critical limit because if exceeded, each calculated mean precipitation ^{3}H activity value of these scanty annual datasets showed great deviation from the modeled values. However, occasionally, a precipitation deficit over 15%, yet under 30%, causes a noticeable difference (Fig. 5e) suggesting that fewer than 30% missing data may be advisable for the calculation of a representative amount-weighted isotopic means. In addition, these reinforcing the decision made in the present study to only include values for which more than 85% fallen precipitation was analyzed for ^{3}H.

Although at Nick and Zgornja Radovna –as seen above– the AP^{3}H database provides a realistic estimation (Fig. 5a,b) one may ask why are the calculated annual mean precipitation ^{3}H activity values are systematically higher at Siófok (mean difference in the 'complete-years': 1.52 TU) than AP^{3}H data (Fig. 5d). It could be explained by the closeness of the largest shallow lake in Central Europe, Lake Balaton (Hatvani et al., 2020)4b) or in the case of "incomplete years", when the ratio of fallen precipitation not analyzed for ^{3}H in a given year was >15% (e.g. Budapest: 1987 and 1991, Fig. 4a; Ljubljana: 1986, 1997-1998, 2000 and 2010, Fig. 4b). In these particular years, when the measured ^{3}H values were not used for interpolation, the modelled values seem more capable of reproducing the actual ^{3}H variability using the neighboring stations' than from the fragmented ^{3}H data of the incomplete year.

The average differences between the Regional model and measured values were -0.03TU for Budapest, and 1.13TU for Ljubljana, excluding the years with not enough precipitation represented. In the meanwhile, the average difference in the so-called incomplete years was ~17 to 0 TU for Budapest and Ljubljana, with a general tendency of obtaining higher differences with a higher ratio of precipitation not represented by tritium measurements. It is noteworthy, that although the short-term intradecadal variability of atmospheric tritium is different at the two sites, their long-term decrease concurs even at a ~ 400 km distance, again indicating the goodness of the interpolation.

The presented Regional model. The mean residence time in the largest basin of the lake, Siófok Basin, was estimated to be between two and six years in the 1990s (Istvánovics et al., 2002), which presumably in the same range in the 2010s as well. Keeping in mind the gradual decrease of ^{3}H in meteoric waters (in the region e.g. Fig. 4), the local evaporation from this 'aged' reservoir can provide an isotopically detectable contribution to the atmospheric moisture which can be transferred to the falling rain droplets via molecular exchange mechanisms (Bolin, 1959), resulting in higher tritium activity values at the Siófok station than the ones from the AP^{3}H database(Fig. 5d). This is a simple example illustrating the potential of this new regional annual mean precipitation ^{3}H activity database (AP^{3}H_v1) in hydrological applications.

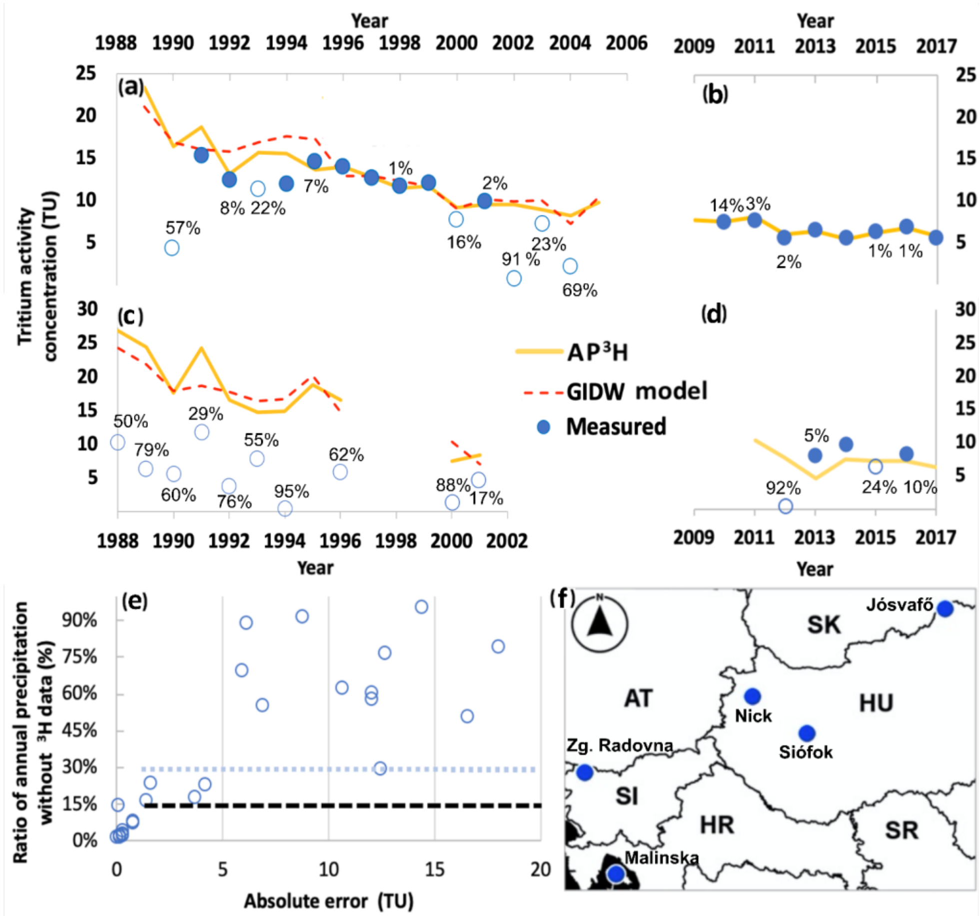

Figure 5 Amount-weighted annual mean ^{3}H activity concentration in precipitation measured at **(a)** Nick (1990-2004; HU), **(b)** Zgornja Radovna (2010-2017; SI) **(c)** Jósvafő (HU; 1988-1996) and Malinska (2000-2001; Krk Island; HR), and **(d)** Siófok (2009-2017; HU) stations along with the corresponding AP^{3}H esitmates. The percentages next to the symbols of the measured values indicate the ratio of fallen precipitation not analyzed for ^{3}H in a given year, if it was >0%. The red dashed line in panels **(a)** and **(c)** show the corresponding values of Global inverse distance weighted model (GIDW; (Jasechko and Taylor, 2015)). **(e)** Cross plot showing the relation between absolute error of estimated precipitation ^{3}H activity by the AP^{3}H and the percentage of precipitation without ^{3}H measurement at Nick, Zgornja Radovna, Jósvafő, and Malinska. Thick dashed line mark the limit of completeness (15%) applied in this study, while gray dotted line marks the limit

of completeness (30%) according to the GNIP protocol (IAEA, 1992). **(f)** The map shows the location of the five sites used for evaluation of the performance of the AP^{3}H database.

5. Comparison between global and regional modelled annual mean ^{3}H activity concentrations in precipitation across the Adriatic-Pannonian Region

The presented AP^{3}H_v1 database of tritium activity was compared with the spatially corresponding output of both currently available global precipitation tritium isoscapes: the Modified global model of tritium in precipitation (MGMTP (Zhang et al., 2011)) and the Global inverse distance weighted model (GIDW) at Budapest (Fig. ~~4a~~3a) and Ljubljana (Fig. ~~4b~~3b). Between 1975 and 1980 the ~~Regional model's~~estimates from the AP^{3}H database and the ~~MGMTP's estimates~~MGMTP are very similar and resemble the actual weighted annual mean precipitation ^{3}H at Budapest. However, only at Ljubljana is the MGMTP capable of steadily reproducing the actual measurements until the late-1990. Afterwards, it indicates solely negative values, which are uninterpretable, just as most of the MGMTP predicted values at Budapest after 1980. In the meanwhile, the ~~Regional model gave~~AP^{3}H database provided much more accurate and reliable results (Fig. ~~4~~3) as discussed above. Note here, that the weak estimation of the MGMTP can be attributed to the difficulties in reading the precipitation tritium activity values from the only available output (isoline map) of the model and the undocumented factors of the model in given years.

The GIDW (Jasechko and Taylor, 2015) model was capable of reproducing the measured precipitation tritium values much more accurately at all locations than the MGMTP (Fig. ~~4~~3). Nevertheless, the GIDW model produced a striking overestimation at the beginning of the modelled period, for example, in 1977, when the measured values at Budapest were overestimated by >20 TU (Fig. ~~4a~~3a). On the contrary, the GIDW model underestimated the actual values from 1981 to 1991, except for one year (Fig. ~~4a). It should be noted, that the Regional model gave an even better regional estimate, then either of the global models.~~ 3a).

~~**Figure 4:** Measured and estimated ^{3}H values at Budapest, Hungary (A) and Ljubljana, Slovenia (B) between 1976 and 2017. The black dotted lines indicate the estimations of the 'modified global model of tritium in precipitation' (MGMTP; and the red dashed ones indicate the Global inverse distance weighted model (GIDW). Note here that uninterpretable negative estimates of MGMTP were not shown. The percentages next to the modelled values indicate the ratio of fallen precipitation not analyzed for ^{3}H in a given year, if it was >0%. The empty circles indicate an "incomplete year" in which the given ^{3}H value was not used for interpolation.~~

~~As an additional out-of-sample verification, the measured precipitation tritium records at stations Zgornja Radovna (2010-2017), Siófok (2013-2016) and Malinska (Dec 2000 and 2001) were compared to the Regional model's estimated ^{3}H time series of the grid closest to the stations. The average annual difference between the modelled and measured values was 3.8 TU in 2001 at Malinska (Fig. 5a), 0.1 TU at Zgornja Radovna (Fig. 5b) and -1.7 TU at Siófok stations (Fig. 5c), while the st. dev. of the differences was 0.3 and 1.6 TU for Zgornja Radovna and Siófok respectively. The Regional model estimated annual amount-weighted ^{3}H activity at Zgornja Radovna very accurately, while the somewhat higher difference at Siófok could be~~

explained by the closeness of the largest shallow freshwater lake in Central Europe, Lake Balaton . The mean residence time in the largest basin of the lake, Siófok Basin, was estimated to be between 2 and 6 yrs in the 1990s (Istvánovics et al., 2002), which presumably in the same range in the 2010s as well. Keeping in mind the gradual decrease of ^{3}H in meteoric waters (in the region e.g. Fig. 4), the evaporation from this 'aged' reservoir can provide an isotopically detectable contribution to the atmospheric moisture measured at Siófok station, resulting in higher tritium activity values than the modelled ones (Fig. 5c). The high difference (+3.8 TU) between the Regional model and the measured values at Malinska can be attributed to the high portion of precipitation (20%) not having corresponding tritium measurements in either year. Moreover, at Malinska, the Regional model provided more reliable estimates than the MGMTP, which The skills of the AP^{3}H database and the GIDW model could only be compared at Nick station because the record of Zgornja Radovna starts in the terminal year of the GIDW model's coverage. In the first 'complete-year' of the Nick record the GIDW estimation is closer to the actual measurements compared to the AP^{3}H estimations, however from 1992 to 1994 the GIDW estimations show much higher (mean error: 4.6 TU) annual mean ^{3}H activity compared to the values calculated from the monthly measurements. In the meanwhile the difference with the AP^{3}H estimations is smaller (mean error: 2.2 TU). Interannual variations of the estimated precipitation ^{3}H activity from the GIDW and the AP^{3}H database are in agreement at Jósvafő and the models are largely in agreement regarding the absolute values and the decreasing trend, too.

However, the MGMTP had practically no success in estimating the actual measured values. For instance, in both 2000 and 2001 the MGMTP produced negative - thus meaningless - values in the periodat Malinska, when direct measurements were available (Fig. 5a, while the present database provided more reliable estimates (Fig. 5c).

Figure 5: Tritium activity concentration values (measured and modelled by the Regional model) at stations Malinska (Krk Island) in 2000 and 2001 (A), Zgornja Radovna (B) and Siófok (C) between 2009 and 2017. The percentages next to the modelled values indicate the ratio of fallen precipitation not analyzed for ^{3}H in a given year, if it was >0%. The red empty circle indicates a single available monthly measured value for December 2000. Error bars show measurement uncertainties, although it is smaller than the marker in B and C. The inset map shows the location of the sites used for out-of-sample verification.

Taken all together, the AP^{3}H_v1 database gave better regional estimates, than either of the global models. This may be because i) the AP^{3}H database is based on more local station-data for the study area compared to the global models relying only on the GNIP records (Table 1) and ii) the improved performance of the database obviously benefitted from incorporating the spatial correlation structure of precipitation tritium activity into the presented geostatistical prediction.

5.6. Possibility of applications (outlook, conclusions)

Continuous long-term records of tritium in precipitation are scarcely available worldwide, thus estimations or modelling are necessary to exploit its potential in hydrological researches. In order to decrease the uncertainty of tritium activity in the hydrological models, the application of regional ^{3}H models have to be increased, since these are more capable of producing accurate estimations than global ones (Stewart and Morgenstern, 2016).

Instead of using remote station data or ad hoc composite curves, site specific time-series retrieved from the presented ~~Regional~~AP^{3}H_v1 database of precipitation amount-weighted annual mean ^{3}H isoscapes should be used. These isoscapes (Kern et al., 2019) can serve as a reference dataset for studies on infiltration dynamics, water transport through various compartments of the hydrological cycle, mixing processes, run-off modelling; e.g. to estimate mean residence time in surface waters and groundwater (Kanduč et al., 2014; Ozyurt et al., 2014; Szucs et al., 2015). As a specific type of hydrogeological application, the Regional model of ^{3}H time-series will serve as a benchmark in estimating the mean infiltration age of dripwater (Kluge et al., 2010) which can provide an additional tool for ongoing cave monitoring studies from the region (e.g. Czuppon et al. (2018); Czuppon et al. (2013); Fehér et al. (2016); Surić et al. (2010)) in a spatiotemporally accurate way.

The higher precipitation ^{3}H activity observed at a lakeshore station (Fig. 5c) likely reflects moisture recycling from the aged lake surface water via evaporation to the local precipitation. The observed deviation highlights the potential of the database to reveal sub-regional anomalous local sources in the hydrological cycle. As a special case the post-2010 isoscapes can serve as benchmarks for background tritium activity for the region, helping to determine local increases of technogenic tritium from these backgrounds.

~~Our Regional model~~The AP^{3}H database was able to provide better estimates than either of the currently available global models for the study area. Its values seem capable of reproducing the actual annual mean precipitation ^{3}H activity than from the fragmented ^{3}H data representing fewer than 85% of the annual sum of precipitation. Prior to 1975 we encourage the use of the GIDW model's estimations (Jasechko and Taylor, 2015) as a reference for studies dealing with precipitation tritium activity. The Regional model and the GIDW model should be spliced together at 1975 and can be used together in the need of a semi-centennial precipitation tritium activity dataset.

~~6.~~7. Data format and availability

The final product~~,~~, (AP3H_v1.nc), the spatially continuous annual (1976-2017) 1×1 km grids of precipitation amount-weighted annual mean tritium activity for the Adriatic-Pannonian Region is provided in a netCDF-4 (net-work common data form) format available at PANGAEA (https://doi.pangaea.de/10.1594/PANGAEA.896938) (Kern et al., 2019), compiled using the EPSG 3857 projection. For a solely visual inspection of the annual grids Panoply (https://www.giss.nasa.gov/tools/panoply/~~A~~) is recommended. An ~~R-~~script written to be able to browse the dataset and convert the projection to EPGS 4326 is provided in the supplement. This publication describes AP^{3}H_v1 database. As the database is updated with corrected or extended new data the new versions will be indexed with incrementing integers.

~~7.~~8. Author contribution

ZK designed the experiments. PV, MŠ, TK, LP, MS, GyC and IKB contributed data. IGH and DE developed the model code and performed the analyses. ZK, IGH, PV and DE prepared the manuscript with contributions from TK, MŠ, IF, and BK. The authors applied the FLAE approach for the sequence of authors. See https://doi.org/10.1371/journal.pbio.0050018 for further details. All authors took part in the manuscript preparation, and revision.

~~8.~~9. Competing interests:

The authors declare that they have no conflict of interest.

~~9.~~10. Acknowledgements

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