A European map of groundwater pH and calcium

Michal Hájek[1*], Borja Jiménez-Alfaro[1,2*], Ondřej Hájek[1], Lisa Brancaleoni[3], Marco Cantonati[4], Michele Carbognani[5], Anita Dedić[6], Daniel Dítě[1,7], Renato Gerdol[3], Petra Hájková[1], Veronika Horsáková[1], Florian Jansen[8], Jasmina Kamberović[9], Jutta Kapfer[10], Tiina H. M. Kolari[11], Mariusz Lamentowicz[12], Predrag Lazarević[13], Ermin Mašić[14], Jesper Erenskjold Moeslund[15], Aaron Pérez-Haase[16,17], Tomáš Peterka[1], Alessandro Petraglia[5], Eulàlia Pladevall-Izard[17], Zuzana Plesková[1], Stefano Segadelli[18], Yuliya Semeniuk[19], Patrícia Singh[1], Anna Šímová[1], Eva Šmerdová[1], Teemu Tahvanainen[11], Marcello Tomaselli[5], Yuliya Vystavna[20,21], Claudia Biţă-Nicolae[22], Michal Horsák[1]

*the authors contributed equally to the study

[1] Department of Botany and Zoology, Faculty of Science, Masaryk University, Kotlářská 2, 61137 Brno, Czech Republic

[2] Research Unit of Biodiversity (CSIC/UO/PA), University of Oviedo, Spain

[3] Department of Life Sciences and Biotechnology, University of Ferrara, Corso Ercole I d'Este 32, 44121 Ferrara, Italy

[4] MUSE – Museo delle Scienze, Limnology & Phycology Section, Corso del Lavoro e della Scienza 3, Trento, 38123, Italy

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[5] Department of Chemistry, Life Sciences and Environmental Sustainability, University of Parma, Parco Area delle Scienze 11/A, 43124 Parma, Italy

[6] Faculty of Science and Education, University of Mostar, Rodoč bb, 88 000 Mostar, Bosnia and Herzegovina

[7] Plant Science and Biodiversity Center, Slovak Academy of Sciences, Dúbravská cesta 9, 84523 Bratislava, Slovak Republic

[8] Faculty of Agricultural and Environmental Sciences, University of Rostock, Justus-von-Liebig-Weg 6, 18059 Rostock, Germany

[9] Faculty of Natural Sciences and Mathematics, University of Tuzla, 75 000, Tuzla, Bosnia and Herzegovina

[10] Department of Landscape Monitoring, Norwegian Institute of Bioeconomy Research, Holtvegen 66, 9016 Tromsø, Norway

[11] Department of Environmental and Biological Sciences, University of Eastern Finland, Joensuu campus, Yliopistokatu 7, 80101 Joensuu, Finland

[12] Climate Change Ecology Research Unit, Faculty of Geographical and Geological Sciences, Adam Mickiewicz University in Poznan, Bogumiła Krygowskiego 10, 61-680 Poznaň, Poland

[13] lnstitute of Botany and Botanical Garden "Jevremovac", Faculty of Biology, University of Belgrade, Takovska 43, 11000 Belgrade, Serbia

[14] Department of Biology, Faculty of Science, University of Sarajevo, Zmaja od Bosne 33-35, 71000 Sarajevo, Bosnia and Herzegovina

[15] Section of Biodiversity, Department of Bioscience, Aarhus University, Grenaavej 14, 8410 Roende, Denmark

[16] Department of Biosciences, Faculty of Sciences and Technology, University of Vic - Central University of Catalonia, Vic, Spain, EU

[17] Department of Evolutionary Biology, Ecology and Environmental Sciences, Faculty of Biology, University of Barcelona, Barcelona, Spain

[18] Geological, Seismic and Soil Service, Emilia-Romagna Region, Bologna, Italy

[19] Stanislaw Leszczycki Institute of Geography and Spatial Organization, Polish Academy of Science, Warsaw, Poland

[20] Institute of Hydrobiology, Biology Centre CAS, Na Sadkach 7, 37005, Ceske Budejovice, Czech Republic

[21] Isotope Hydrology Section, International Atomic Energy Agency, Vienna International Centre, Vienna 1400, Vienna, Austria

[22] Institute of Biology Bucharest, Romanian Academy, Bucharest, Romania

Abstract. Water resources and associated ecosystems are becoming highly endangered due to ongoing global environmental changes. Spatial ecological modelling is a ~~widely used tool~~promising toolbox for understanding the past, present and future distribution and diversity patterns in groundwater-dependent ecosystems, such as fens, springs, streams, reed beds or wet grasslands. Still, the lack of detailed water chemistry maps prevents ~~their~~the use of reasonable ~~use~~ models to be applied on continental and global scales. Being major determinants of

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biological composition and diversity of groundwater-dependent ecosystems, groundwater pH and calcium are of utmost importance. Here we developed the up-to-date European map of groundwater pH and Ca, based on 7,577 measurements of near-surface groundwater pH and calcium distributed across Europe. In comparison to the existing European groundwater maps, we included a several times larger number of sites, especially in the regions rich in spring and fen habitats, and filled the apparent gaps in Eastern and Southeastern Europe. We used Random Forest models and regression kriging to create continuous maps of water pH and calcium at the continental scale, which is freely available also as a raster map (Hájek et al. 2020; 10.5281/zenodo.4139912). Lithology had higher importance than climate for both pH and calcium. The previously recognised latitudinal and altitudinal gradients were rediscovered with much refined regional patterns, as associated with bedrock variation. For ecological models of distribution and diversity of ~~groundwater-dependent, but also other~~many terrestrial~~,~~ ecosystems, ~~the~~our new map based on field ground water measurements is more suitable than ~~previously used~~ maps of soil pH, ~~unlike~~ which ~~it mirrors~~mirror not only bedrock chemistry ~~more than~~, but also vegetation-dependent soil processes.

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1. Introduction

The Earth system is currently undergoing unprecedented changes in climate, global biogeochemical cycles, and land use, resulting in biodiversity loss (Ceballos et al. 2017, Song et al. 2018, Blowes et al. 2019, Brondizio et al. 2019). Freshwater systems belong to the most endangered habitats (Cantonati et al. 2020a, Tickner et al. 2020) and, among them, groundwater-dependent ecosystems, such as fens and springs, hold primacy (Janssen et al. 2016, Chytrý et al. 2019, Hájek et al. 2020, Stevens et al. 2020). Species composition and richness of spring systems are generally governed by water pH and calcium concentration (Ca^{2+}), which are highly variable at different spatial scales (Malmer 1986; Rydin and Jeglum 2013; Peterka et al. 2017; Horsáková et al. 2018; Cantonati et al. 2020a,b). Therefore, understanding the spatial patterns in groundwater pH and Ca^{2+} is important not only for general geochemical knowledge and for water resource management, but to the same extent for the conservation of freshwater systems and associated biodiversity.

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In Earth and biodiversity sciences, ecological modelling is a widely used tool for understanding the distribution and diversity patterns of ecosystems and habitats, and for predicting their future development under global change. Ecological models usually incorporate environmental or historical predictors extracted from thematic maps (Jiménez-Alfaro et al. 2018a, Večeřa et al. 2019, Divíšek et al. 2020), including soil properties for terrestrial ecosystems (Hengl et al. 2017). However, soil parameters as soil pH contribute negligibly to the models for groundwater-dependent habitats, even for those strongly controlled by pH and Ca^{2+}, such as base-rich fens (Jiménez-Alfaro et al. 2018b). This is due to a poor correlation between groundwater chemistry and pH or Ca^{2+} in soil, disrupted mainly by mineral leaching or accumulation of organic matter in soil. For this reason, there is a strong need to produce maps for groundwater pH and Ca^{2+} concentration at the European scale that would allow producing the continental-scale ecological models useful for enforcing conservation strategies in groundwater-dependent habitats. Ideally, such models should include lithology as a dominant factor determining groundwater pH and Ca^{2+} concentration (Hem 1985, Chapelle 2003, Tahvanainen 2004, Stevens et al. 2020).

In spite of important mapping efforts of groundwater (Duscher et al. 2015) and karst aquifers (Chen et al. 2017) at the European and global level, the only available European-scale maps of groundwater pH and Ca^{2+} concentration are those included in the FOREGS Geochemical Atlas of Europe (Salminen et al. 2006). These maps are based on 808 stream-water measurements distributed relatively equally across Europe. However, they show a large gap in Eastern and Southeastern Europe (Romania, Bulgaria, Belarus, Russian Federation, Ukraine, Moldova, Serbia, Kosovo, Montenegro, Bosnia and Herzegovina, Northern Macedonia). In addition, ~~the~~those maps are ~~constructed~~ based on insufficient data density in some areas rich in~~)~~ ~~different~~ groundwater-dependent ~~ecosystems~~ecosystem types, but heterogeneous in terms of lithology (the Alps, the Carpathians, Bohemian Massif, the Cantabrian Mountains and the Pyrenees, and some regions of Fennoscandia). We therefore aimed at substantial improvement of the existing data by creating a database with field data measurements across the entire European continent, and at creating a model-based map representing major patterns of groundwater pH and Ca^{2+} concentration at local and continental scales. Our data will allow better understanding of the patterns and causes of groundwater conditions in freshwater systems, ~~and~~ strongly ~~improve~~improving the ~~databases~~spatial information suitable for European-scale modelling of ~~the~~ biodiversity in groundwater-dependent and related ecosystems.

2. Methods

Data collection

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We assembled the data set of pH and Ca^{2+} (or electrical conductivity in µS.cm^{-1} at 20 °C; hereinafter abbreviated as EC) measurements in groundwater, covering the whole of Europe, with a greater density in the regions rich in endangered groundwater-dependent ecosystems such as springs and fens. We excluded most of Ukraine and European part of Russian Federation, because of large data gaps in these areas. We considered all types of shallow groundwater systems, especially spring, spring-fen, and stream water. The core of our data set is formed by unpublished pH and Ca^{2+} or EC data sets of co-authors (3,618 sites); some of them processed in ecological papers without presenting original pH and Ca^{2+} data (Hájková et al. 2006, 2008; Hájek et al. 2008; Sekulová et al. 2013; Plesková et al. 2016; Horsáková et al. 2018; Šímová et al. 2019). The second most important source were vegetation databases registered in GIVD (Dengler et al. 2011; Table 1) and EVA (Chytrý et al. 2016), from where 1,160 measurements from freshwater habitats were obtained. Both unpublished data and data from vegetation databases were filtered using original information or metadata of the sources in a way that only data from spring-fed fens and springs were considered. The data from ombrotrophic bogs and clearly topogenic fens (mainly terrestrialised lakes) were omitted because their water chemistry is governed by the decomposition of organic matter, atmospheric humidity and deposition, algal photosynthesis (Kann and Smith 1999), and biotic processes such as cation exchange capacity of mosses (Clymo 1963, Soudzilovskaia et al. 2010, Vicherová et al. 2015), rather than by bedrock chemistry. We also obtained data from public data sets ~~from~~stored in national environmental and nature conservation agencies of Germany, Slovenia and Bulgaria (1081 sites; see Table 1), data from FOREGS Geochemical Atlas of Europe (Salminen et al. 2006; 808 sites) and literature data based on our gap-oriented excerption (883 sites; Table 1); most data came from Hinterlang

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(1992), Tanneberger et al. (2011), Eades et al. (2018), Kadūnas et al. (2017) and Savić et al. (2017).

Table 1. Data sources. The name abbreviations are explained in the team list or acknowledgements: **Table did no change.**

In total, we collected 7,577 samples (Table 1). ~~A part~~Some of ~~the~~these samples~~,~~ ~~however, represented~~ are repeated measurements conducted in the same site, especially in public data sets~~. Some~~ , while other samples (from ~~other data sets (~~vegetation databases, or literature data) ~~shared~~share the same coordinates and site name or code, suggesting repeated measurements as well. We therefore averaged repeated measurements from the same sampling ~~spots~~sites. We further deleted samples whose coordinates were obviously erroneous, such as those in oceans. These steps reduced the number of samples to 6,561, out of which 6,459 samples contained information on water pH value and 5,927 samples contained information about EC of water or Ca^{2+} concentration. Out of these 5,927 samples, 2,988 ~~samples~~ had directly measured both Ca^{2+} and EC ($\mu S.cm^{-1}$ at 20 °C), and for the remaining 2,939 samples we estimated Ca^{2+} concentration by EC of water.

Imputation of missing Ca^{2+} values by EC of water

For imputation of Ca^{2+} values based on EC, we first aimed at constructing a simple imputation equation based on the well-known correlation between EC and Ca^{2+} concentration in springs and

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fens (Hem 1985, Sjörs & Gunnarsson 2002, Plesková et al. 2016). In our data set of 2,988 samples, as well as in its regional subsets, this relationship was strongly governed by EC values above ca 1,000 µS.cm^{-1}, although they formed only a small part of the data set (4.7% of the data set; 139 samples). In the EC range 1,000-10,000 µS.cm^{-1} (an outlier of 17,000 µS.cm^{-1} was omitted), the correlation between water EC and Ca^{2+} concentration was not statistically significant (r = 0.15, P = 0.07). The problem of high EC values governing the regression model was the most apparent in the public data sets. In the data set of Bulgarian Ministry of Environment, weak correlation between EC and Ca^{2+} persisted even when EC values above 1000 were omitted (Supplementary Figure 1). This database further contains many samples which are not near-surface samples that were measured in other datasets. We therefore finally decided (1) not to include the database of Bulgarian Ministry of Environment into the imputation model, and (2) limit the gradient of EC to 1,000 µS.cm^{-1}. We further omitted a few samples from ophiolite (Kamberović et al. 2019) where high EC occurred despite low Ca^{2+}. The resulting data set of 2,319 samples nevertheless still showed some samples with suspiciously high or low Ca^{2+} concentration relative to EC (Supplementary Figure 2), suggesting either the effect of other ions than Ca^{2+} or inconsistent analytical methodology. Because our aim was to create the most accurate imputation model rather than testing the relationship, we removed these outliers. Therefore, we calculated the EC:Ca and Ca:EC ratios and removed outliers, i.e., all points outside the 1.5 x interquartile range. The final imputation model was hence based on 2,062 sites. We performed a null-intercept linear regression (Figure 1) with Ca^{2+} as dependent variable (y) and EC as predictor (x); the resulting equation y = 0.153x was obtained (R^2 = 0.84). Such relationship between Ca2+ and EC is similar as that found in the abovementioned studies (Hem 1985, Sjörs & Gunnarsson 2002, Plesková et al. 2016). Based on this equation, we imputed Ca^{2+}

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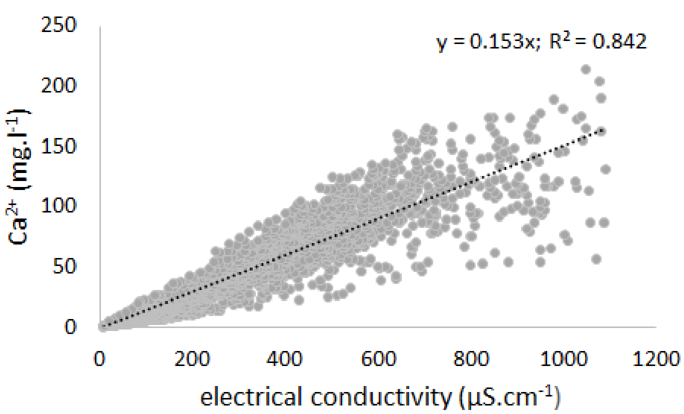
concentrations to all samples where only EC was measured. The imputed Ca^{2+} values show a somewhat narrower range (Supplementary Figure 3) than originally measured values. Both subsets show minimum Ca^{2+} value below 1 mg.l^{-1}, but imputed data show lower non-outlier maximum (125.5 mg.l^{-1}) than measured data (197.2 mg.l^{-1}). Absolute maximum value was also lower for the subset with imputed values. Imputation of Ca^{2+} values to all samples, including high-EC ones (> 1,000 µS.cm^{-1}), hence did not skew the imputed data to higher values.

Figure 1: The final regression model to impute Ca^{2+} values based on electrical conductivity (EC; µS.cm^{-1}; n = 2,062



Geographical modelling and selection of the predictors

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We used our dataset ~~with 6,561 sites~~ with measured groundwater pH and either measured or imputed Ca^{2+} concentrations to model expected values across non-sampled areas. ~~Groundwater~~Our aim was to produce continuous maps for groundwater-dependent pH (GW-pH) ~~was modeled with~~using 6,459 samples (pH min = 2.20; max = 11.32; mean = 6.69); and groundwater-dependent Ca^{2+} (GW-Ca) ~~with~~using 5,927 samples (min = 0.15; max = 3567.41; mean = 48.73 mg.l^{-1}). Ca^{2+} values were ln-transformed. All field samples had geographic coordinates assigned from GPS or georeferenced with an accuracy between ca 10 m (precise field measurements) and 500 m (~~some database data~~from georeferenced sites in databases). We kept the pH outliers; ten values below 3.5 and nine values above 8.8. Even if these values may be suspicious, they largely come from published sources (FOREGS Geochemical Atlas of Europe, British vegetation database). Apart from measurement error, they may be explained by the influence of mineral waters from deep hydrological circulations (e.g., in a spring in the Apennines, very high pH value 11.2 was due to enrichment with sodium and chloride associated with low temperature reaction between meteoric water and ultramafic rocks; Boschetti and Toscani 2008, Boschetti et al. 2013, Segadelli et al. 2017, Cantonati et al. 2020c). These values form only a minor part of the data set and have negligible effect on the results.

For each site, we obtained environmental predictors from thematic GIS maps (see below). We focused on the predictors that may causally affect the groundwater pH and calcium richness. Aquifer chemistry is of prime importance (Hem 1985, Fairchild et al. 1994, Frei et al. 2000, Chapelle et al. 2003, Tahvanainen 2004, Stevens et al. 2020), but no such thematic map exists, at least at the scale needed for computing our spatial predictions. We therefore included the lithological groups from the Hydrogeological Map of Europe (Duscher et al. 2015), together with soil pH maps (see below), for which we anticipated a certain correlation with bedrock chemistry.

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Apart from aquifer chemistry, residence time may also affect groundwater chemistry by impacting dissolution rates. Precipitation amount and frequency affect not only flow paths activity and redistribution of groundwater, but also its residence time in the aquifer, impacting carbonate dissolution and precipitation rates (Hem 1985, Crossman et al. 2011, Lewandowski et al. 2015, Vystavna et al. 2020). Groundwater with a short transit time (1–3 years) or 'young water' (Soulsby et al. 2015) can be particularly sensitive to changes in precipitation amount and frequency. ~~For each site, we obtained environmental predictors from the thematic GIS maps (see below). For some sites, important predictors were missing in the maps (e.g., sites at far north in the arctic zone, or close to sea or water bodies) and these sites were therefore not included in the final models.~~

We therefore also considered climatic parameters associated with precipitation in the models (see below). Although there are some other potential predictors of minor importance that may affect groundwater chemistry (Hem 1985, Stevens et al. 2020), no corresponding thematic map is available to be included into our models. For some sites, the selected predictors were missing in the maps (e.g., sites at far north in the arctic zone, or close to sea or water bodies) and these sites were therefore not included in the final models. We finally collected topographic data to test the potential effect of elevation and slope as indirect factors potentially influencing groundwater chemistry.

Numerical analyses

Numerical analyses were done in R version 3.6.3 (R Core Team, 2020), with the support of ArcGIS 10.2 (ESRI, Redlands, CA) for geoprocessing and map production. We first conducted

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exploratory analyses to test the Ca and pH prediction ability of different GIS layers related to soil bedrock, climate, and topography. on the variation of both Ca and pH. We focused on layers with a complete coverage of Europe, with an eastern border from the Black Sea in Turkey to the White Sea in Russian Federation, thus including the regions with a relatively good cover of field measurements (Figure 2). We performed Linear Models for individual variables to select those providing significant relationships and > 1% of explained variance. A variable for soil pH (measured in water solution) at 15 cm depth for a 250 m grid resolution provided by the soilgrids project (www.soilgrids.org) had the highest explanatory power for GW-pH ($R^2 = 0.22$) and GW-Ca ($R^2 = 0.16$). The same results were obtained when using the same variable for 5 or 10 cm depth. We also tested soil estimates from Ballabio et al. (2019), but they provided weaker relationships for both GW-pH ($R^2 = 0.14$ using soil pH as a predictor) and GW-Ca ($R^2 = 0.01$ using soil pH, $R^2 = 0.01$ using soil $CaCO_3$). To account for lithology, we used the lithological groups (litho3 level) included in the polygon layer of the Hydrogeological map of Europe (Duscher et al. 2015) as a categorical variable. We also selected annual precipitation (Bio12) as provided in CHELSA (Karger et al. 2017) to account for precipitation gradients which are expected to influence groundwater regimes. Other CHELSA variables related to precipitation were highly correlated with annual precipitation (Pearson r > 0.75) and omitted. Slope and elevation showed negligible effects on both GW-pH and GW-Ca ((linear regression, adjR2 < 0.05, P < 0.001). Since preliminary models showed no differences with these variables were included, they were discharged.

The variables of lithology and soil pH were aggregated to the same grid extent of CHELSA at 1 km resolution, using the dominant unit and a bilinear interpolation, respectivelyas the most appropriate scale to balance the original scales of both layers. This grid extent is also the most

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suitable spatial scale to be used in the context of further ecological modeling, which is in many cases combined with climatic data from e.g. CHELSA or WorldClim (www.worldclim.org) for making temporal climatic projections. The lithological map (originally at 1:1,500,000 scale, which corresponds to a raster resolution of c. 1 km) was converted to a grid resolution using the dominant unit. Soil pH was converted from the original 250 m to 1 km grid resolution using a bilinear interpolation to create a smooth surface based on the weighted average of the four nearest cells.

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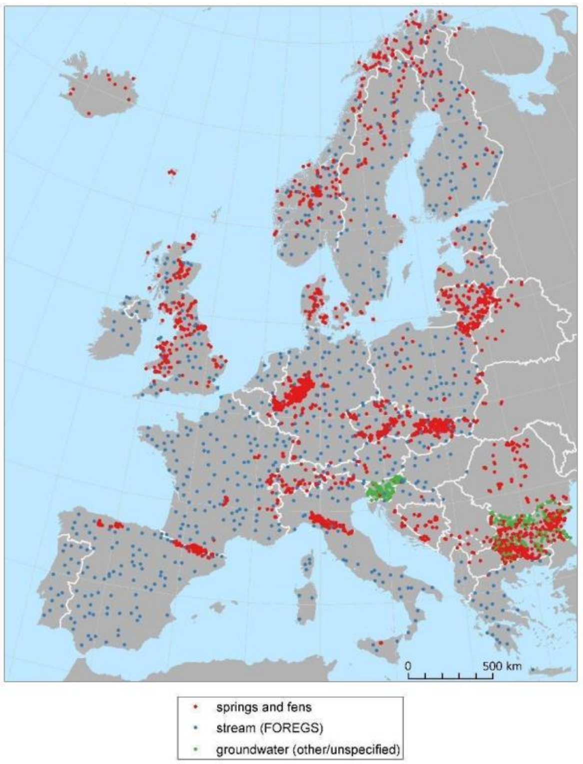
Figure 2. Spatial distribution of the three groups of calibration data collected for modelling groundwater pH and Ca^{2+} in European fens (original and literature data from springs and fens; data from streams from FOREGS Geochemical Atlas of Europe; other data). Other data include public data from national groundwater monitoring of Bulgaria and Slovenia. For separate maps of pH and Ca^{2+} see Supplementary Figure 5.

~~Spatial predictions were~~



Because even the combination of the selected variables might not lead to precise fine-scale indication of aquifer chemistry, we further employed a kriging approach to data analysis, assuming an effect of spatial correlation to estimate the values close to the original samples. The final spatial predictions were therefore based on regression kriging (RK), a technique that combines a regression model based on explanatory variables with the interpolation of model residuals with ordinary kriging (Hengl et al. 2007; Meng et al. 2013). RK is especially appropriate for modeling soil attributes at medium and large scales, combining the spatial autocorrelation of soil variables with the explanatory power of auxiliary variables (Keskin and

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Grunwald 2018). We implemented RK with the GSIF R package (Hengl 2020). As the regression component, we computed Random Forests since a preliminary analysis with our data showed better performance than linear models, generalised linear models, or generalised additive models. Random Forests are ensemble learning methods based on decision trees and an internal correction of overfitting, which provide high interpretability and good performance when compared with other algorithms used in soil spatial modeling (Wiesmeier et al. 2011). Another advantage of Random Forests is that they have no requirements for considering the probability distribution of soil variables, fitting complex non-linear relationships for spatial extrapolation (Hengl et al. ~~2015).~~2015) that ultimately improve the spatial predictions. We fitted the Random Forests model and the residual variogram for groundwater pH and Ca^{2+} separately using the function fit.gstatModel() in GSIF package. Effect plots for the predictors were created for the same models using partial() function in pdp package (Greenwell 2017). Spatial predictions were then computed with the predict() function using the model object generated previously and a 5-fold cross-validation. Model evaluation was based on the calculation of the Mean Error (ME) and the Root Mean Squared Error (RMSE) as the differences between predicted and observed values (Keskin and Grunwald 2018; Pham et al. 2019). We compared the relationships between the models produced for both groundwater pH and Ca^{2+} by using a random sampling of 5,000 points to extract cell values and computing a Pearson correlation. To assess regional differences, we correlated values grouped in 25 neighboring cells of each single cell using the rasterCorrelation() function in the ~~package~~ spatialEco~~.~~ R package (Evans 2020).

3. Results

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In measured data, ranges and medians of pH and Ca^{2+} concentration ~~was~~were similar across Europe (Supplementary Figure 4), with the lowest pH values found in the Atlantic and Iberian regions, and the highest pH values found in southern Europe except Iberian Peninsula. ~~Lowest~~The lowest Ca^{2+} values were found in boreal Europe, while the highest in Central and Southern Europe. The Random Forest models computed with the lithology, soil pH, and precipitation explained 40% and 55% of the variance for GW-pH and GW-Ca, respectively. Lithology was the variable with the highest importance in both models (Figure 3), although its effect was higher in the model computed for Ca^{2+} than for pH. ~~Conversely, soil~~These effects were mainly associated with the lithological units reflecting calcareous bedrocks, followed by categories with coarse and fine sediments such as flysch (Figure 4). Soil pH had higher relative importance in GW-pH than GW-Ca, ~~while~~although in both cases the variable had a similar positive effect. Finally, annual precipitation had the lowest contributions in ~~both~~the two models (Figure 3), with both Ca^{2+} and pH dropping suddenly after the threshold of annual precipitation of ca 1800 mm (Figure 4), although the highest pH values occur under the lowest precipitation and tend to decrease towards high-precipitation areas.

Figure 3. Variable importance of Random Forest models computed for groundwater pH and Ca^{2+}. MSE = Mean Standard Error.

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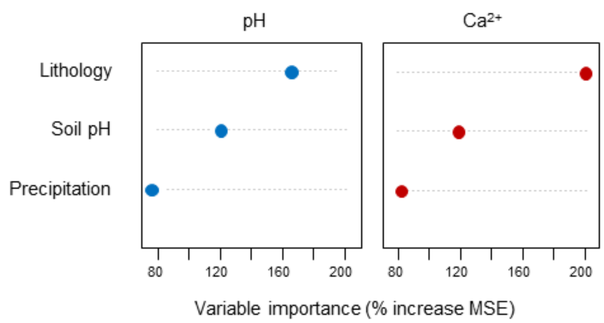
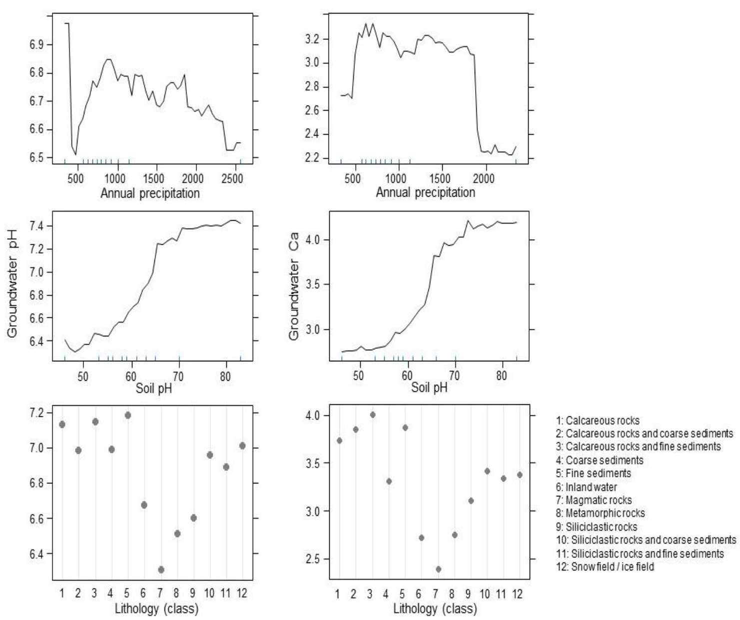


Figure 4. Partial dependence plots showing the effects of the variables used in the Random Forest models computed for model groundwater pH and Ca^{2+} in Europe.



When adding the kriging component, model predictions reached 65% and 74% of explained variance for GW-pH and GW-Ca, respectively. The mean values of standard errors (SE; 0.00058 for pH; -0.0009 for Ca) and Root Mean Squared Errors (RMSE; 0.588 for pH; 0.690 for Ca) were higher in the models for pH, but in both cases showed low values ~~and~~that suggest accurate predictions, in agreement with their total explained variance.

Model predictions for ~~groundwater~~ GW-pH reflected the lowest values in Scandinavia, Iceland, northern UK, and some regions of Central and Eastern Europe (Figure ~~4~~5). The highest values were predicted in eastern Iberia and many regions of Central and Eastern Europe, although a big part of the study area was dominated by neutral pH values (6 to 7). The spatial patterns for GW-Ca (Figure ~~4~~5) were rather similar to pH. The overall correlation between the two models was 0.83 (Pearson r, P < 0.001), but they showed differences in some regions. This was supported by the spatial correlation computed for each cell (Figure ~~5~~6), reflecting different magnitudes of correlation across the study area, especially in the eastern Iberian Peninsula and Southeastern Europe.

Figure ~~4~~5. Model predictions based on Regression Kriging. Note the Ca^{2+} concentration is on ln-scale.

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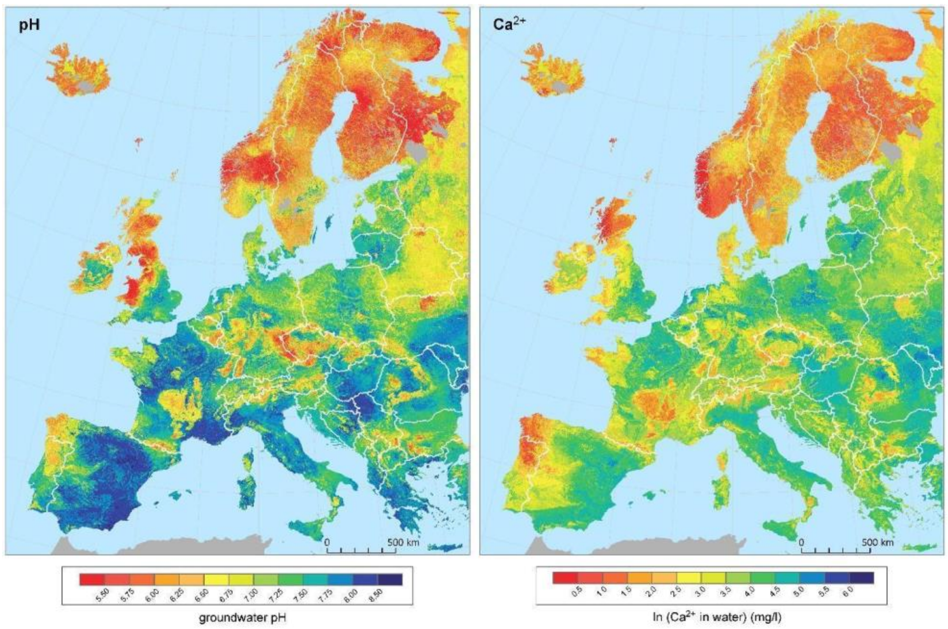
pH
Ca2+
0 500 km
5.50 5.75 6.00 6.25 6.50 6.75 7.00 7.25 7.50 7.75 8.00 8.50
groundwater pH
0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0 5.5 6.0
ln (Ca2+ in water) (mg/l)

Figure ~~5~~6. Spatial correlation between the models computed for groundwater pH and Ca^{2+}. Values show Pearson correlation coefficient computed over every single cell by using a sampling of 25 neighboring cells.

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Discussion

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3.1. *Spatial patterns in groundwater pH and Ca^{2+} concentration in Europe*

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As expected, the ~~recent~~ values of water pH and Ca^{2+} concentration are largely shaped by lithology in groundwater-dependent habitats across Europe. Indeed, it has been recognised by regional studies that the distribution of major spring and fen habitats, of which the species composition largely depends on pH and Ca^{2+}, is well determined by bedrock type (Hájek et al. 2002, Tahvanainen 2004, Hinterlang 2017, Peterka et al. 2017, Cantonati et al. 2020c). Since the European-scale geological map we used here is not precise enough to capture differences in bedrock chemistry within the major ~~geological~~lithological units that are defined largely by ~~age~~geological stratification, the contribution of soil pH in the model probably also reflected lithological variation, as soil pH generally correlates with regional bedrock chemistry (Chadwick and Chorover 2001). On the other hand, soil pH is also affected by climate-dependent pedogenesis, which incorporates a climate-zonal geographical component in this effect (Duchaufour 2012, Maxbauer et al. 2017). ~~Precipitation is another important determinant of groundwater chemistry. Precipitation amount and frequency affect not only flow paths activity and redistribution of groundwater, but also its residence time in the aquifer, impacting carbonate dissolution and precipitation rates (Crossman et al.~~

Precipitation is another determinant of groundwater chemistry in our study. High annual precipitation above ca 1800 mm obviously reduced~~2011, Lewandowski et al. 2015, Vystavna et al. 2020). Groundwater with a short transit time (1–3 years) or ‘young water’ (Soulsby et al. 2015) can be particularly sensitive to changes in precipitation amount and frequency. Here, intensive precipitation may reduce~~ an interaction time of groundwater with Ca^{2+} and carbonates deposited in rocks (Fairchild et al. 1994, Segadelli et al. 2017, Cantonati et al. 2020b), resulting in lower Ca^{2+} concentration ~~of elements~~ in groundwater. ~~In snow-influenced ecosystems,~~This effect is more pronounced in snowy regions, where seasonal snowmelt ~~can also~~

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~~modulate~~modulates the recharge patterns of groundwater. ~~Particularly, the~~The duration of the snowmelt period can impact the occurrence and dynamic of preferential flow, and prolong or reduce the interaction of the seepage with soil and bedrock materials (Mohammed et al. 2019). We therefore suggest that fast hydrological pathways and short transit time driven by snowmelt and precipitation can explain the lowest Ca^{2+} in hyper-oceanic cold regions of SW Norway or W Scotland. It may further explain lower pH and Ca^{2+} values on windward slopes of high mountains, even if bedrock is moderately calcium-rich. Nevertheless, understanding the complementary effects of precipitation and slope will need to account for more accurate models based on GPS data, better precipitation data, and high-resolution (<250 m) topographic predictors.

The resulting pattern at the European scale is governed by the strong latitudinal and altitudinal gradients, i.e. decreasing pH and Ca^{2+} northwards, and regionally also towards mountain regions. This pattern largely follows bedrock chemistry, with crystalline rocks prevailing, and most carbonate rocks being eroded by glaciers, in high latitudes and altitudes. The excess of precipitation over evaporation, and theoretically also slower weathering rates in colder regions (White & Blum 1995), contribute as well. Although this pattern is well known (Økland et al. 2001, Hájek et al. 2006, Hinterlang 2017, Peterka et al. 2017) and has been captured also by the FOREGS Geochemical Atlas of Europe (Salminen et al. 2006), our improved model provides much finer regional patterns. In Southern Europe, low pH and Ca^{2+} values were modelled in the Pyrenees, the Balkans, SW Corse, and Calabria, i.e. the regions where boreal or endemic types of fen communities occur as relicts (Chytrý et al. 2020). The Alps, the Apennines, the Carpathians, and the Baltic region show a fine-scaled mosaic of alkaline (calcium-rich) and acidic (calcium-poor) groundwater that contributes to the high

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diversity and conservation value of groundwater-dependent ecosystems, such as fens (Cantonati et al. 2009, 2011, Gerdol et al. 2011, Joosten et al. 2017, Horsáková et al. 2018). The most apparent “acidic island” in Central Europe is located in the SW part of the Bohemian Massif (Czech Republic, Germany), where acidic types of springs and fens are quite frequent and some studies further document anthropogenic acidification on siliceous bedrock in 1970–80s, being re-emerged recently because of extreme climatic events (Kapfer et al. 2012, Schweiger et al. 2015). It is, however, possible that particularly this acidic island is picked out mainly because of the high amount of available data.

Clearly, most of Fennoscandia is markedly acidic and calcium-poor mainly due to glacial history. Yet, the model identified small alkaline and calcium-enriched islands in NE and Central Sweden, and NW Norway, which are associated with rare types of calcareous fen and spring communities (Dierssen 1982, Vorren et al. 1999, Udd et al. 2015, Miller et al. 2020). More localised pockets of calcareous habitats are however known from most parts of Fennoscandia that are not recognised with the grain of our European-wide analysis. With our results, the future modelling of diversity and distribution of individual habitat types of groundwater-dependent wetlands will be more reliable~~, regionally~~. Regionally rare habitat conditions ~~can~~will be ~~better~~ recognised better, and the disentangling of ~~the~~ climate and pH effects will be more~~ easily~~ feasible.

3.2. ~~*Persisting data*~~*Data gaps and further improvements*

Although being based on the hitherto most comprehensive field data set currently available, the presented map cannot be considered definitive. Surely there are many pH and EC

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or Ca^{2+} measurements conducted across Europe that we could not include into the data set because they are hardly accessible. Except for Russian Federation and Moldova, largest gaps still occur in the southern parts of the Pannonian plain (southeastern Hungary, northern Serbia, and western Romania), in SE Belarus, and eastern Ukraine. We ~~dispose of~~have available some data from the latter region (Vystavna et al. 2015; Supplementary Table 2), but a large gap in the rest of the data set prevented reliable geospatial modelling. These data might be used in future updates of the map once the gap in Central Ukraine is filled. The lack of data in the Pannonian plain has led to poor correlation between predicted pH and Ca^{2+} values (Fig. 4). Such a poor correlation and sometimes low density of data apply also for some other lowland regions, such as the Danube plain in S Romania, Po valley in Italy and valleys around the Duero, Ebro, and Tagus rivers in Spain. Apart from eutrophication, this ~~result~~pattern may be caused by the imbalanced distribution of groundwater-dependent habitat types in our data set. Unlike mountain regions, the data for these lowlands were largely taken from the FOREGS Geochemical Atlas of Europe (Salminen et al. 2006) and national groundwater databases, i.e. largely from stream water. Considering the major purpose of our map~~, the~~ (creating new data sources for ecological modelling of fens and springs~~,~~) these regions are ~~generally not as important~~less crucial for biodiversity modeling because~~ of the low number of existing target habitats, as~~ they have largely been transformed to arable land or they are too dry. On the other hand, caution is needed when interpreting the maps in an ecological sense. The extremely high pH (> 8) and Ca^{2+} (ln [Ca^{2+}] > 4; i.e., Ca^{2+} > 55 mg.l^{-1}) values that occur in lowlands visually govern the map, but for ecological differentiation of groundwater-dependent habitats in Europe the differences within the middle part of the gradient, i.e. between pH 5.5 and 7.0, are much more important (Malmer 1986, Wheeler & Proctor 2000, Hájek et al. 2006, Rydin and Jeglum 2013). ~~In any case, our~~

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Our data set is expected to be amended in the future, as more studies will be published and more data will be available., so further versions will be accessible in the open repository. New data will help to improve predictions for those regions with relatively lower sampling effort, and also those with a lithologically heterogeneous landscapelandscapes. Future updates of the model may also focus at finer spatial resolution (e.g., 100 to 250 m) but this will require to increase the spatial accuracy of the calibration data and the predictor variables, as in some Central-European areas (Le et al. 2019, Chuman et al. 2019). Although we tested several variables with potential predictive effect on groundwater pH and Ca, many of them had lower explanatory power (e.g. Ballabio et al. 2019) or they were redundant with the soil pH layer we used (i.e. previous versions of soil.grids using the same data sources). The low predictive value of other predictors with potential predictive value, like slope, elevation or other precipitation variables, is probably related to their broad spatial scale, with values averaged at 1 km grid resolution having little impact to discriminate groundwater variation at the landscape level (Jiménez-Alfaro et al. 2018b). This contrasts with the predictive value of the lithological layer, which is however based on a similar spatial (but originally vectorial) resolution. However, lithological bedrock is generally more homogeneous at the landscape scale, with the only exception of certain geological complex regions. These findings suggest that future improvements of our models will depend on the quality of new lithological (or related soil chemistry) variables with direct effect on groundwater pH and Ca. We also note that the lithological map we used here is simplified to large units, while many regional and national geological maps are being produced at finer details. The combination of such new predictors with accurate calibration data at the continental level and at fine scale resolution seems the most likely opportunity to produce significantly better

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models, since the influence of spatial-dependent (kriging) effects is rather limited by the distribution of sample points.

Conclusions

Here, we provide the first European map of groundwater pH and Ca^{2+} content. We collected as even as possible distributed field measurements of water pH and Ca^{2+} or EC from European groundwater-dependent habitats, ~~Despite these persisting gaps, our European map of near-surface groundwater pH and EC provides the best solution for modelling the biodiversity of groundwater-dependent ecosystems, especially at the continental or supra-regional scale. We even believe that this map could be more suitable also for ecological modelling of other than groundwater-dependent habitats. It may mirror the bedrock chemistry better than the map of soil pH, because soil pH is a resultant of pedogenetic processes, which are tightly associated with the character of the vegetation cover itself (Miles 1985, Duchaufour 2012).~~

~~**Conclusions**~~

~~Here, we provide the first European map of groundwater pH and Ca^{2+} content. We collected as even as possible distributed field measurements of water pH and Ca^{2+} or EC from European groundwater-dependent habitats, having high data density~~with relatively higher sampling effort in regions rich in endangered groundwater-dependent ecosystems (springs, fens~~),~~). Despite the general high accuracy of our models, we note that prediction uncertainties may affect the reliability of models computed with both Random Forests and kriging (Hengl et al. 2018; Szatmári & Pásztor 2019). Another source of prediction uncertainty is related to the quality of

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the original chemical measurements and used geospatial modelling. The model considered predominantlythe georeferentiation of their geographic position. Moreover, the predictor variables rely on spatial models (soil pH, precipitation) or broad geographic maps (lithology) which are based on their own uncertainties and assumptions. Future Improvements of groundwater pH and Ca estimates should therefore consider a more accurate set of response variables and fine-scale predictors, preferably including lithology and soil pH (i.e., variables surrogating bedrock chemistry) and precipitation sum (i.e., residence time of groundwater). Our results also Despite potential uncertainties and data gaps, this study uses an unprecedented combination of data to provide a freely accessible mapand realistic maps that can be used in any kind of spatial modelling, showing better resolution and fewer gaps than previously published maps. The character of our input data, which are also freely accessible, predetermines our map for being used in ecological modelling to address the distribution and diversity of groundwater-dependent ecosystems and associated species. The character of our input data, which are also freely accessible, predetermines our map for being used in ecological modelling to address the distribution and diversity of groundwater-dependent ecosystems and associated species. Moreover, we assume the map will represent the best choice also for other types of earth modeling, because unlike previous maps we included Eastern-European and Balkan countries and considered lithology in geospatial modelling.We even believe that our maps could be also suitable for ecological modelling of other than groundwater-dependent habitats. It may mirror the bedrock chemistry better than the map of soil pH, because soil pH is a resultant of pedogenetic processes, which are tightly associated with the character of the vegetation cover itself (Miles 1985, Duchaufour 2012). We conclude that our European maps of near-surface groundwater pH and EC provides the best solution currently available for modelling the

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biodiversity of groundwater-dependent ecosystems, especially at the continental or supra-regional scale.

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Data availability

The dataset of georeferenced pH and EC measurements and the resulting maps in GIS-compatible format (shapefile) are accessible at ~~www.zenodo.org~~www.zenodo.org; doi 10.5281/zenodo.4139912 (Hájek et al. 2020).

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Code availability

No original R code was used; the used codes are cited.

Sample availability

No geoscientific samples registered as International Geo Sample Number (IGSN) have been used for the manuscript.

Team list

Michal Hájek (M.Há.), Borja Jiménez-Alfaro (B.J.-A.), Ondřej Hájek, Lisa Brancaleoni (L.B.), Marco Cantonati (M.Can.), Michele Carbognani (M.Car.), Anita Dedić (A.D.), Daniel Dítě (D.D.), Renato Gerdol (R.G.), Petra Hájková (P.H.), Veronika Horsáková (V.H.), Florian Jansen (F.J.), Jasmina Kamberović (J.Kam.), Jutta Kapfer (J.Kap.), Tiina Kolari (T.K.), Mariusz

Lamentowicz (K.L.), Predrag Lazarević (P.L.), Ermin Mašić (E.M.), Jesper Erenskjold Moeslund (J.E.M.), Aaron Pérez-Haase (A.P.-H.), Tomáš Peterka (T.P.), Zuzana Plesková (Z.P.), Alessandro Petraglia (A.P.), Eulàlia Pladevall-Izard (E.P.-I.), Stefano Segadelli (S.S.), Yuliya Semeniuk (Y.S.), Patrícia Singh (P.S.), Anna Šímová (A.Š.), Eva Šmerdová (E.Š.), Teemu Tahvanainen (T.T.), Marcello Tomaselli (M.T.), Yuliya Vystavna (Y.V.), Claudia Biţă-Nicolae (C.B.-N.), Michal Horsák (M.Hor.).

Author contribution

M.H. and B.J-A. contributed equally to the paper. They conceived the research, collected data and led writing. B.J-A. designed and performed Random Forest and Regression Kriging models. O.H. prepared the input data and final map outputs. All authors provided unpublished data and commented on the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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