

A European map of groundwater pH and calcium

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

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

76

77 **Copyright statement**

78 No copyright statement is needed.

79

80 **1. Introduction**

81 The Earth system is currently undergoing unprecedented changes in climate, global
82 biogeochemical cycles, and land use, resulting in biodiversity loss (Ceballos et al. 2017, Song et
83 al. 2018, Blowes et al. 2019, Brondizio et al. 2019). Freshwater systems belong to the most

84 endangered habitats (Cantonati et al. 2020a, Tickner et al. 2020) and, among them, groundwater-
85 dependent ecosystems, such as fens and springs, hold primacy (Janssen et al. 2016, Chytrý et al.
86 2019, Hájek et al. 2020, Stevens et al. 2020). Species composition and richness of spring systems
87 are generally governed by water pH and calcium concentration (Ca^{2+}), which are highly variable
88 at different spatial scales (Malmer 1986; Rydin and Jeglum 2013; Peterka et al. 2017; Horsáková
89 et al. 2018; Cantonati et al. 2020a,b). Therefore, understanding the spatial patterns in
90 groundwater pH and Ca^{2+} is important not only for general geochemical knowledge and for water
91 resource management, but to the same extent for the conservation of freshwater systems and
92 associated biodiversity.

93 In Earth and biodiversity sciences, ecological modelling is a widely used tool for
94 understanding the distribution and diversity patterns of ecosystems and habitats, and for
95 predicting their future development under global change. Ecological models usually incorporate
96 environmental or historical predictors extracted from thematic maps (Jiménez-Alfaro et al.
97 2018a, Večeřa et al. 2019, Divíšek et al. 2020), including soil properties for terrestrial
98 ecosystems (Hengl et al. 2017). However, soil parameters as soil pH contribute negligibly to the
99 models for groundwater-dependent habitats, even for those strongly controlled by pH and Ca^{2+} ,
100 such as base-rich fens (Jiménez-Alfaro et al. 2018b). This is due to a poor correlation between
101 groundwater chemistry and pH or Ca^{2+} in soil, disrupted mainly by mineral leaching or
102 accumulation of organic matter in soil. For this reason, there is a strong need to produce maps for
103 groundwater pH and Ca^{2+} concentration at the European scale that would allow producing the
104 continental-scale ecological models useful for enforcing conservation strategies in groundwater-
105 dependent habitats. Ideally, such models should include lithology as a dominant factor

106 determining groundwater pH and Ca²⁺ concentration (Hem 1985, Chapelle 2003, Tahvanainen
107 2004, Stevens et al. 2020).

108 In spite of important mapping efforts of groundwater (Duscher et al. 2015) and karst
109 aquifers (Chen et al. 2017) at the European and global level, the only available European-scale
110 maps of groundwater pH and Ca²⁺ concentration are those included in the FOREGS Geochemical
111 Atlas of Europe (Salminen et al. 2006). These maps are based on 808 stream-water
112 measurements distributed relatively equally across Europe. However, they show a large gap in
113 Eastern and Southeastern Europe (Romania, Bulgaria, Belarus, Russian Federation, Ukraine,
114 Moldova, Serbia, Kosovo, Montenegro, Bosnia and Herzegovina, Northern Macedonia). In
115 addition, those maps are based on insufficient data density in some areas rich in groundwater-
116 dependent ecosystem types, but heterogeneous in terms of lithology (the Alps, the Carpathians,
117 Bohemian Massif, the Cantabrian Mountains and the Pyrenees, and some regions of
118 Fennoscandia). We therefore aimed at substantial improvement of the existing data by creating a
119 database with field data measurements across the entire European continent, and at creating a
120 model-based map representing major patterns of groundwater pH and Ca²⁺ concentration at local
121 and continental scales. Our data will allow better understanding of the patterns and causes of
122 groundwater conditions in freshwater systems, strongly improving the spatial information
123 suitable for European-scale modelling of biodiversity in groundwater-dependent and related
124 ecosystems.

125

126 **2. Methods**

127 *Data collection*

128 We assembled the data set of pH and Ca²⁺ (or electrical conductivity in $\mu\text{S}\cdot\text{cm}^{-1}$ at 20 °C;
129 hereinafter abbreviated as EC) measurements in groundwater, covering the whole of Europe,
130 with a greater density in the regions rich in endangered groundwater-dependent ecosystems such
131 as springs and fens. We excluded most of Ukraine and European part of Russian Federation,
132 because of large data gaps in these areas. We considered all types of shallow groundwater
133 systems, especially spring, spring-fen, and stream water. The core of our data set is formed by
134 unpublished pH and Ca²⁺ or EC data sets of co-authors (3,618 sites); some of them processed in
135 ecological papers without presenting original pH and Ca²⁺ data (Hájková et al. 2006, 2008; Hájek
136 et al. 2008; Sekulová et al. 2013; Plesková et al. 2016; Horsáková et al. 2018; Šímová et al.
137 2019). The second most important source were vegetation databases registered in GIVD
138 (Dengler et al. 2011; Table 1) and EVA (Chytrý et al. 2016), from where 1,160 measurements
139 from freshwater habitats were obtained. Both unpublished data and data from vegetation
140 databases were filtered using original information or metadata of the sources in a way that only
141 data from spring-fed fens and springs were considered. The data from ombrotrophic bogs and
142 clearly topogenic fens (mainly terrestrialised lakes) were omitted because their water chemistry
143 is governed by the decomposition of organic matter, atmospheric humidity and deposition, algal
144 photosynthesis (Kann and Smith 1999), and biotic processes such as cation exchange capacity of
145 mosses (Clymo 1963, Soudzilovskaia et al. 2010, Vicherová et al. 2015), rather than by bedrock
146 chemistry. We also obtained data from public data sets stored in national environmental and
147 nature conservation agencies of Germany, Slovenia and Bulgaria (1081 sites; see Table 1), data
148 from FOREGS Geochemical Atlas of Europe (Salminen et al. 2006; 808 sites) and literature data
149 based on our gap-oriented excerption (883 sites; Table 1); most data came from Hinterlang

150 (1992), Tanneberger et al. (2011), Eades et al. (2018), Kadūnas et al. (2017) and Savić et al.
151 (2017).

152

153 **Table 1.** Data sources. The name abbreviations are explained in the team list or
154 acknowledgements.

I. Vegetation databases.

<i>name</i>	<i>n</i>	<i>GIVD code</i>	<i>custodian</i>
European Mire Vegetation Database (EMVD)	510	GIVD EU-00-022	T.P.
National Vegetation Database of Denmark	373	EU-DK-002	J.E.M.
Britain_nvcd	224	GIVD EU-GB-001	J.R.
Balkan Vegetation Database	27	GIVD EU-00-019	K.V.
Germany_vegmv	19	GIVD EU-DE-001	F.J.
Basque country	7	EU-00-011	I.B.

II. Unpublished data sets

<i>Regions</i>	<i>n</i>	<i>co-authors of the data set</i>
Central and Eastern Europe	1405	Z.P., M.Há., P.H., T.P., D.D., L.S., M.L., J.N., P.P., P.S., A.Š., Y.S., C.B.-N.
Spain	645	A.P-H., E.P-I., B.J-A.
Bulgaria	428	P.H., M.Há., M.Hor.
Fennoscandia	392	M.Há., T.P., D.D., M.Hor., V.H., P.H., T.K., T.T., J.Kap., D.-I.Ø.
Apennines	285	M.T., M.Can., M.Car., S.S., A.P., L.B., R.G.
Europe (cross-taxon research)	281	M.Há., P.H., D.D., M.Hor., V.H.
Balkans except Bulgaria	134	A.D., E.M., J.Kam., P.L., T.P., M.Há., P.H.

III. Public data sets

<i>area and agency</i>	<i>n</i>	<i>provided via</i>
North Rhine-Westphalia (LANUV agency; D)	463	Dr. Dirk Hinterlang, Dr. Sabine Bergmann
Ministry of Environment and Water (BG)	442	Mrs. Rossitza Gorova
Ministry of the Environment (SI)	176	http://www.arso.gov.si/ , assessed 26 February 2019

IV. Geochemical Atlas of Europe

<i>area</i>	<i>n</i>	<i>reference</i>
Europe	808	Salminen et al. 2006

V. Other literature data (gap-oriented excerption)

<i>region and context</i>	<i>n</i>	<i>reference</i>
West-Central European springs	340	Hinterlang 1992
Lithuanian springs	194	Kadūnas et al. 2017
NE England	111	Eades et al. 2018
N Germany	58	Tanneberger et al. 2011
Central Bosnia	50	Savić et al. 2017
Western Bohemian mineral springs (CZ)	28	Laburdová and Hájek 2014
Scotland	24	Gorham 1957
Eastern Bosnia	20	Kamberović et al. 2019

Switzerland (mires)	14	Lamentowicz et al. 2010
British and French travertines	13	Pentecost and Zhaohui 2002
NW Poland (mires)	8	Lamentowicz and Mitchell 2005
Western Balkans	6	Ridl et al. 2018
Kosovo	5	Kelmendi et al. 2018
SE Croatia	4	Terzić et al. 2014
Kosovo (Rugova)	3	Lajçi et al. 2017
Serbia	3	Ćirić et al. 2018
NE Croatia	1	Špoljar et al. 2011
SE Croatia (Krčić)	1	Kolda et al. 2019

155

156 In total, we collected 7,577 samples (Table 1). Some of these samples are repeated
157 measurements conducted in the same site, especially in public data sets, while other samples
158 (from vegetation databases, or literature data) share the same coordinates and site name or code,
159 suggesting repeated measurements as well. We therefore averaged repeated measurements from
160 the same sampling sites. We further deleted samples whose coordinates were obviously
161 erroneous, such as those in oceans. These steps reduced the number of samples to 6,561, out of
162 which 6,459 samples contained information on water pH value and 5,927 samples contained
163 information about EC of water or Ca²⁺ concentration. Out of these 5,927 samples, 2,988 had
164 directly measured both Ca²⁺ and EC ($\mu\text{S}\cdot\text{cm}^{-1}$ at 20 °C), and for the remaining 2,939 samples we
165 estimated Ca²⁺ concentration by EC of water.

166

167 *Imputation of missing Ca²⁺ values by EC of water*

168 For imputation of Ca²⁺ values based on EC, we first aimed at constructing a simple imputation
169 equation based on the well-known correlation between EC and Ca²⁺ concentration in springs and
170 fens (Hem 1985, Sjörs & Gunnarsson 2002, Plesková et al. 2016). In our data set of 2,988
171 samples, as well as in its regional subsets, this relationship was strongly governed by EC values

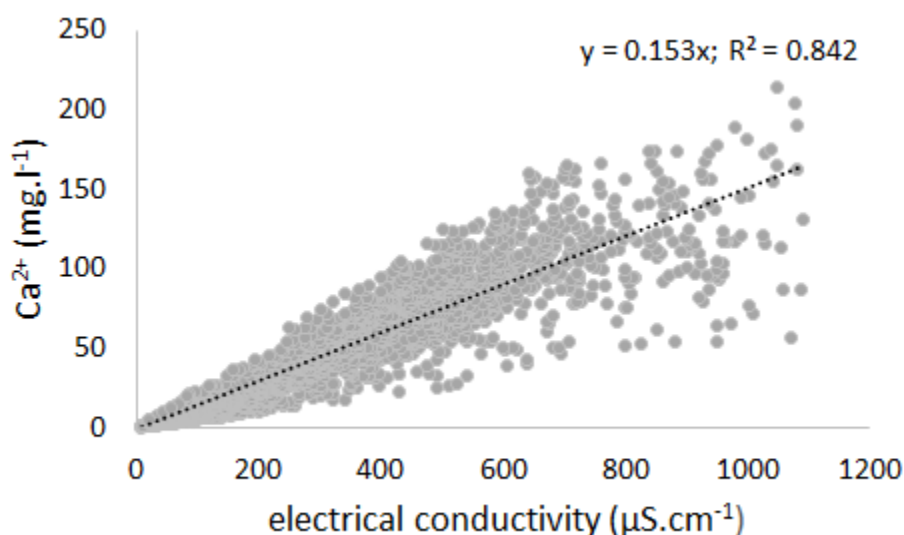
172 above ca 1,000 $\mu\text{S}\cdot\text{cm}^{-1}$, although they formed only a small part of the data set (4.7% of the data
173 set; 139 samples). In the EC range 1,000-10,000 $\mu\text{S}\cdot\text{cm}^{-1}$ (an outlier of 17,000 $\mu\text{S}\cdot\text{cm}^{-1}$ was
174 omitted), the correlation between water EC and Ca^{2+} concentration was not statistically
175 significant ($r = 0.15$, $P = 0.07$). The problem of high EC values governing the regression model
176 was the most apparent in the public data sets. In the data set of Bulgarian Ministry of
177 Environment, weak correlation between EC and Ca^{2+} persisted even when EC values above 1000
178 were omitted (Supplementary Figure 1). This database further contains many samples which are
179 not near-surface samples that were measured in other datasets. We therefore finally decided (1)
180 not to include the database of Bulgarian Ministry of Environment into the imputation model, and
181 (2) limit the gradient of EC to 1,000 $\mu\text{S}\cdot\text{cm}^{-1}$. We further omitted a few samples from ophiolite
182 (Kamberović et al. 2019) where high EC occurred despite low Ca^{2+} . The resulting data set of
183 2,319 samples nevertheless still showed some samples with suspiciously high or low Ca^{2+}
184 concentration relative to EC (Supplementary Figure 2), suggesting either the effect of other ions
185 than Ca^{2+} or inconsistent analytical methodology. Because our aim was to create the most
186 accurate imputation model rather than testing the relationship, we removed these outliers.
187 Therefore, we calculated the EC:Ca and Ca:EC ratios and removed outliers, i.e., all points
188 outside the 1.5 x interquartile range. The final imputation model was hence based on 2,062 sites.
189 We performed a null-intercept linear regression (Figure 1) with Ca^{2+} as dependent variable (y)
190 and EC as predictor (x); the resulting equation $y = 0.153x$ was obtained ($R^2 = 0.84$). Such
191 relationship between Ca^{2+} and EC is similar as that found in the abovementioned studies (Hem
192 1985, Sjörs & Gunnarsson 2002, Plesková et al. 2016). Based on this equation, we imputed Ca^{2+}
193 concentrations to all samples where only EC was measured. The imputed Ca^{2+} values show a
194 somewhat narrower range (Supplementary Figure 3) than originally measured values. Both

195 subsets show minimum Ca^{2+} value below 1 mg.l^{-1} , but imputed data show lower non-outlier
196 maximum (125.5 mg.l^{-1}) than measured data (197.2 mg.l^{-1}). Absolute maximum value was also
197 lower for the subset with imputed values. Imputation of Ca^{2+} values to all samples, including
198 high-EC ones ($> 1,000 \mu\text{S.cm}^{-1}$), hence did not skew the imputed data to higher values.

199

200 **Figure 1:** The final regression model to impute Ca^{2+} values based on electrical conductivity (EC;
201 $\mu\text{S.cm}^{-1}$; $n = 2,062$

202



203

204 *Geographical modelling and selection of the predictors*

205 We used our dataset with measured groundwater pH and either measured or imputed Ca^{2+}
206 concentrations to model expected values across non-sampled areas. Our aim was to produce
207 continuous maps for groundwater-dependent pH (GW-pH) using 6,459 samples (pH min = 2.20;
208 max = 11.32; mean = 6.69); and groundwater-dependent Ca^{2+} (GW-Ca) using 5,927 samples
209 (min = 0.15; max = 3567.41; mean = 48.73 mg.l^{-1}). Ca^{2+} values were ln-transformed. All field

210 samples had geographic coordinates assigned from GPS or georeferenced with an accuracy
211 between ca 10 m (precise field measurements) and 500 m (from georeferenced sites in
212 databases). We kept the pH outliers; ten values below 3.5 and nine values above 8.8. Even if
213 these values may be suspicious, they largely come from published sources (FOREGS
214 Geochemical Atlas of Europe, British vegetation database). Apart from measurement error, they
215 may be explained by the influence of mineral waters from deep hydrological circulations (e.g., in
216 a spring in the Apennines, very high pH value 11.2 was due to enrichment with sodium and
217 chloride associated with low temperature reaction between meteoric water and ultramafic rocks;
218 Boschetti and Toscani 2008, Boschetti et al. 2013, Segadelli et al. 2017, Cantonati et al. 2020c).
219 These values form only a minor part of the data set and have negligible effect on the results.

220 For each site, we obtained environmental predictors from thematic GIS maps (see below).
221 We focused on the predictors that may causally affect the groundwater pH and calcium richness.
222 Aquifer chemistry is of prime importance (Hem 1985, Fairchild et al. 1994, Frei et al. 2000,
223 Chapelle et al. 2003, Tahvanainen 2004, Stevens et al. 2020), but no such thematic map exists, at
224 least at the scale needed for computing our spatial predictions. We therefore included the
225 lithological groups from the Hydrogeological Map of Europe (Duscher et al. 2015), together with
226 soil pH maps (see below), for which we anticipated a certain correlation with bedrock chemistry.
227 Apart from aquifer chemistry, residence time may also affect groundwater chemistry by
228 impacting dissolution rates. Precipitation amount and frequency affect not only flow paths
229 activity and redistribution of groundwater, but also its residence time in the aquifer, impacting
230 carbonate dissolution and precipitation rates (Hem 1985, Crossman et al. 2011, Lewandowski et
231 al. 2015, Vystavna et al. 2020). Groundwater with a short transit time (1–3 years) or ‘young
232 water’ (Soulsby et al. 2015) can be particularly sensitive to changes in precipitation amount and

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

241

242 *Numerical analyses*

243 Numerical analyses were done in R version 3.6.3 (R Core Team, 2020), with the support of
244 ArcGIS 10.2 (ESRI, Redlands, CA) for geoprocessing and map production. We first conducted
245 exploratory analyses to test the prediction ability of GIS layers related to soil bedrock, climate,
246 and topography on the variation of both Ca and pH. We focused on layers with a complete
247 coverage of Europe, with an eastern border from the Black Sea in Turkey to the White Sea in
248 Russian Federation, thus including the regions with a relatively good cover of field
249 measurements (Figure 2). We performed Linear Models for individual variables to select those
250 providing significant relationships and $> 1\%$ of explained variance. A variable for soil pH
251 (measured in water solution) at 15 cm depth for a 250 m grid resolution provided by the soilgrids
252 project (www.soilgrids.org) had the highest explanatory power for GW-pH ($R^2 = 0.22$) and GW-
253 Ca ($R^2 = 0.16$). The same results were obtained when using the same variable for 5 or 10 cm
254 depth. We also tested soil estimates from Ballabio et al. (2019), but they provided weaker
255 relationships for both GW-pH ($R^2 = 0.14$ using soil pH as a predictor) and GW-Ca ($R^2 = 0.01$

256 using soil pH, $R^2 = 0.01$ using soil CaCO_3). To account for lithology, we used the lithological
257 groups (litho3 level) included in the polygon layer of the Hydrogeological map of Europe
258 (Duscher et al. 2015) as a categorical variable. We also selected annual precipitation (Bio12) as
259 provided in CHELSA (Karger et al. 2017) to account for precipitation gradients which are
260 expected to influence groundwater regimes. Other CHELSA variables related to precipitation
261 were highly correlated with annual precipitation (Pearson $r > 0.75$) and omitted. Slope and
262 elevation showed negligible effects on both GW-pH and GW-Ca ((linear regression, $\text{adj}R^2 <$
263 0.05 , $P < 0.001$). Since preliminary models showed no differences with these variables were
264 included, they were discharged.

265 The variables of lithology and soil pH were aggregated to the same grid extent of CHELSA at 1
266 km resolution, as the most appropriate scale to balance the original scales of both layers. This
267 grid extent is also the most suitable spatial scale to be used in the context of further ecological
268 modeling, which is in many cases combined with climatic data from e.g. CHELSA or
269 WorldClim (www.worldclim.org) for making temporal climatic projections. The lithological
270 map (originally at 1:1,500,000 scale, which corresponds to a raster resolution of c. 1 km) was
271 converted to a grid resolution using the dominant unit. Soil pH was converted from the original
272 250 m to 1 km grid resolution using a bilinear interpolation to create a smooth surface based on
273 the weighted average of the four nearest cells.

274

275 **Figure 2.** Spatial distribution of the three groups of calibration data collected for modelling
276 groundwater pH and Ca^{2+} in European fens (original and literature data from springs and fens;
277 data from streams from FOREGS Geochemical Atlas of Europe; other data). Other data include

278 public data from national groundwater monitoring of Bulgaria and Slovenia. For separate maps
279 of pH and Ca²⁺ see Supplementary Figure 5.

280



281

282

283 Because even the combination of the selected variables might not lead to precise fine-scale
284 indication of aquifer chemistry, we further employed a kriging approach to data analysis,
285 assuming an effect of spatial correlation to estimate the values close to the original samples. The
286 final spatial predictions were therefore based on regression kriging (RK), a technique that

287 combines a regression model based on explanatory variables with the interpolation of model
288 residuals with ordinary kriging (Hengl et al. 2007; Meng et al. 2013). RK is especially
289 appropriate for modeling soil attributes at medium and large scales, combining the spatial
290 autocorrelation of soil variables with the explanatory power of auxiliary variables (Keskin and
291 Grunwald 2018). We implemented RK with the GSIF R package (Hengl 2020). As the regression
292 component, we computed Random Forests since a preliminary analysis with our data showed
293 better performance than linear models, generalised linear models, or generalised additive models.
294 Random Forests are ensemble learning methods based on decision trees and an internal
295 correction of overfitting, which provide high interpretability and good performance when
296 compared with other algorithms used in soil spatial modeling (Wiesmeier et al. 2011). Another
297 advantage of Random Forests is that they have no requirements for considering the probability
298 distribution of soil variables, fitting complex non-linear relationships for spatial extrapolation
299 (Hengl et al. 2015) that ultimately improve the spatial predictions. We fitted the Random Forests
300 model and the residual variogram for groundwater pH and Ca^{2+} separately using the function
301 `fit.gstatModel()` in GSIF package. Effect plots for the predictors were created for the same
302 models using `partial()` function in `pdp` package (Greenwell 2017). Spatial predictions were then
303 computed with the `predict()` function using the model object generated previously and a 5-fold
304 cross-validation. Model evaluation was based on the calculation of the Mean Error (ME) and the
305 Root Mean Squared Error (RMSE) as the differences between predicted and observed values
306 (Keskin and Grunwald 2018; Pham et al. 2019). We compared the relationships between the
307 models produced for both groundwater pH and Ca^{2+} by using a random sampling of 5,000 points
308 to extract cell values and computing a Pearson correlation. To assess regional differences, we

309 correlated values grouped in 25 neighboring cells of each single cell using the rasterCorrelation()
310 function in the spatialEco R package (Evans 2020).

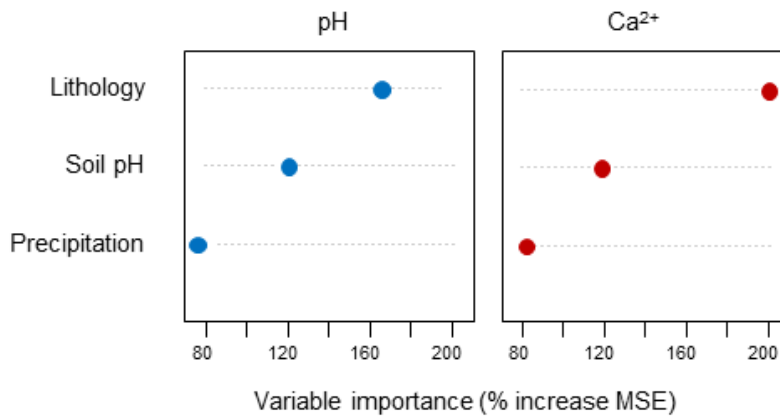
311

312 **3. Results**

313 In measured data, ranges and medians of pH and Ca²⁺ concentration were similar across
314 Europe (Supplementary Figure 4), with the lowest pH values found in the Atlantic and Iberian
315 regions; and the highest pH values found in southern Europe except Iberian Peninsula. The
316 lowest Ca²⁺ values were found in boreal Europe, while the highest in Central and Southern
317 Europe. The Random Forest models computed with the lithology, soil pH, and precipitation
318 explained 40% and 55% of the variance for GW-pH and GW-Ca, respectively. Lithology was the
319 variable with the highest importance in both models (Figure 3), although its effect was higher in
320 the model computed for Ca²⁺ than for pH. These effects were mainly associated with the
321 lithological units reflecting calcareous bedrocks, followed by categories with coarse and fine
322 sediments such as flysch (Figure 4). Soil pH had higher relative importance in GW-pH than GW-
323 Ca, although in both cases the variable had a similar positive effect. Finally, annual precipitation
324 had the lowest contributions in the two models (Figure 3), with both Ca²⁺ and pH dropping
325 suddenly after the threshold of annual precipitation of ca 1800 mm (Figure 4), although the
326 highest pH values occur under the lowest precipitation and tend to decrease towards high-
327 precipitation areas.

328

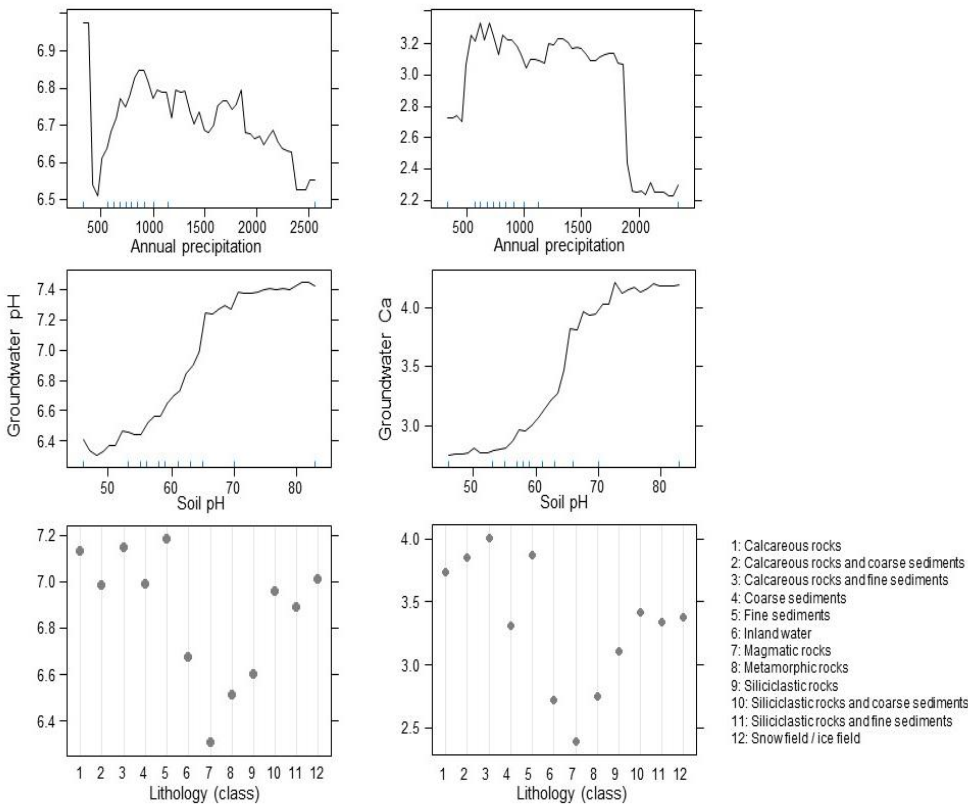
329 **Figure 3.** Variable importance of Random Forest models computed for groundwater pH and
330 Ca²⁺. MSE = Mean Standard Error.



331

332 **Figure 4.** Partial dependence plots showing the effects of the variables used in the Random

333 Forest models computed for model groundwater pH and Ca²⁺ in Europe.



334

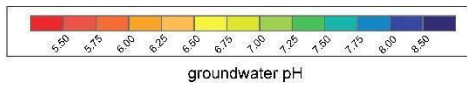
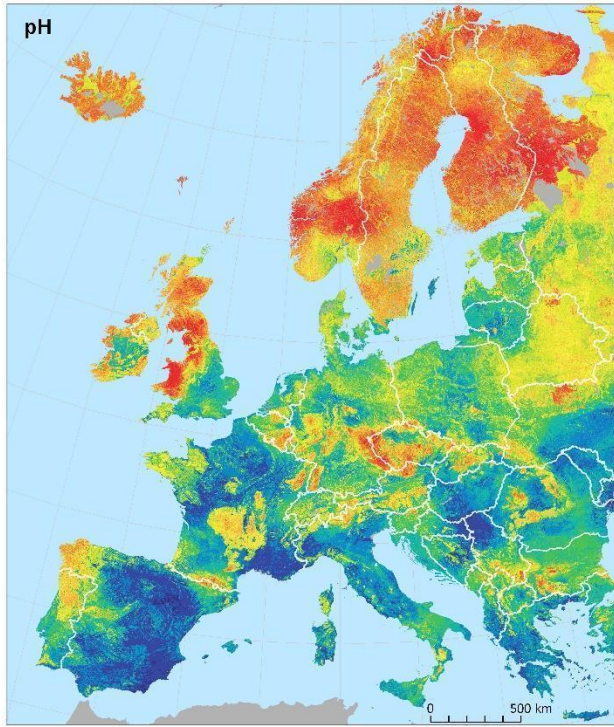
335 When adding the kriging component, model predictions reached 65% and 74% of explained
336 variance for GW-pH and GW-Ca, respectively. The mean values of standard errors (SE; 0.00058
337 for pH; -0.0009 for Ca) and Root Mean Squared Errors (RMSE; 0.588 for pH; 0.690 for Ca)
338 were higher in the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

340 Model predictions for GW-pH reflected the lowest values in Scandinavia, Iceland, northern UK,
341 and some regions of Central and Eastern Europe (Figure 5). The highest values were predicted in
342 eastern Iberia and many regions of Central and Eastern Europe, although a big part of the study
343 area was dominated by neutral pH values (6 to 7). The spatial patterns for GW-Ca (Figure 5)
344 were rather similar to pH. The overall correlation between the two models was 0.83 (Pearson r , P
345 < 0.001), but they showed differences in some regions. This was supported by the spatial
346 correlation computed for each cell (Figure 6), reflecting different magnitudes of correlation
347 across the study area, especially in the eastern Iberian Peninsula and Southeastern Europe.

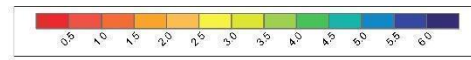
348

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

351



groundwater pH



ln (Ca²⁺ in water) (mg/l)

352

353

354 **Figure 6.** Spatial correlation between the models computed for groundwater pH and Ca²⁺. Values
355 show Pearson correlation coefficient computed over every single cell by using a sampling of 25
356 neighboring cells.



357

358 **Discussion**

359 3.1. *Spatial patterns in groundwater pH and Ca²⁺ concentration in Europe*

360 As expected, the values of water pH and Ca²⁺ concentration are largely shaped by
361 lithology in groundwater-dependent habitats across Europe. Indeed, it has been recognised by

362 regional studies that the distribution of major spring and fen habitats, of which the species
363 composition largely depends on pH and Ca^{2+} , is well determined by bedrock type (Hájek et al.
364 2002, Tahvanainen 2004, Hinterlang 2017, Peterka et al. 2017, Cantonati et al. 2020c). Since the
365 European-scale geological map we used here is not precise enough to capture differences in
366 bedrock chemistry within the major lithological units that are defined largely by geological
367 stratification, the contribution of soil pH in the model probably also reflected lithological
368 variation, as soil pH generally correlates with regional bedrock chemistry (Chadwick and
369 Chorover 2001). On the other hand, soil pH is also affected by climate-dependent pedogenesis,
370 which incorporates a climate-zonal geographical component in this effect (Duchaufour 2012,
371 Maxbauer et al. 2017).

372 Precipitation is another determinant of groundwater chemistry in our study. High annual
373 precipitation above ca 1800 mm obviously reduced an interaction time of groundwater with Ca^{2+}
374 and carbonates deposited in rocks (Fairchild et al. 1994, Segadelli et al. 2017, Cantonati et al.
375 2020b), resulting in lower Ca^{2+} concentration in groundwater. This effect is more pronounced in
376 snowy regions, where seasonal snowmelt modulates the recharge patterns of groundwater. The
377 duration of the snowmelt period can impact the occurrence and dynamic of preferential flow, and
378 prolong or reduce the interaction of the seepage with soil and bedrock materials (Mohammed et
379 al. 2019). We therefore suggest that fast hydrological pathways and short transit time driven by
380 snowmelt and precipitation can explain the lowest Ca^{2+} in hyper-oceanic cold regions of SW
381 Norway or W Scotland. It may further explain lower pH and Ca^{2+} values on windward slopes of
382 high mountains, even if bedrock is moderately calcium-rich. Nevertheless, understanding the
383 complementary effects of precipitation and slope will need to account for more accurate models

384 based on GPS data, better precipitation data, and high-resolution (<250 m) topographic
385 predictors.

386 The resulting pattern at the European scale is governed by the strong latitudinal and
387 altitudinal gradients, i.e. decreasing pH and Ca²⁺ northwards, and regionally also towards
388 mountain regions. This pattern largely follows bedrock chemistry, with crystalline rocks
389 prevailing, and most carbonate rocks being eroded by glaciers, in high latitudes and altitudes.
390 The excess of precipitation over evaporation, and theoretically also slower weathering rates in
391 colder regions (White & Blum 1995), contribute as well. Although this pattern is well known
392 (Økland et al. 2001, Hájek et al. 2006, Hinterlang 2017, Peterka et al. 2017) and has been
393 captured also by the FOREGS Geochemical Atlas of Europe (Salminen et al. 2006), our
394 improved model provides much finer regional patterns. In Southern Europe, low pH and Ca²⁺
395 values were modelled in the Pyrenees, the Balkans, SW Corse, and Calabria, i.e. the regions
396 where boreal or endemic types of fen communities occur as relicts (Chytrý et al. 2020). The
397 Alps, the Apennines, the Carpathians, and the Baltic region show a fine-scaled mosaic of
398 alkaline (calcium-rich) and acidic (calcium-poor) groundwater that contributes to the high
399 diversity and conservation value of groundwater-dependent ecosystems, such as fens (Cantonati
400 et al. 2009, 2011, Gerdol et al. 2011, Joosten et al. 2017, Horsáková et al. 2018). The most
401 apparent “acidic island” in Central Europe is located in the SW part of the Bohemian Massif
402 (Czech Republic, Germany), where acidic types of springs and fens are quite frequent and some
403 studies further document anthropogenic acidification on siliceous bedrock in 1970–80s, being re-
404 emerged recently because of extreme climatic events (Kapfer et al. 2012, Schweiger et al. 2015).
405 It is, however, possible that particularly this acidic island is picked out mainly because of the
406 high amount of available data.

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

416

417 3.2. *Data gaps and further improvements*

418 Although being based on the hitherto most comprehensive field data set currently
419 available, the presented map cannot be considered definitive. Surely there are many pH and EC
420 or Ca²⁺ measurements conducted across Europe that we could not include into the data set
421 because they are hardly accessible. Except for Russian Federation and Moldova, largest gaps still
422 occur in the southern parts of the Pannonian plain (southeastern Hungary, northern Serbia, and
423 western Romania), in SE Belarus, and eastern Ukraine. We have available some data from the
424 latter region (Vystavna et al. 2015; Supplementary Table 2), but a large gap in the rest of the data
425 set prevented reliable geospatial modelling. These data might be used in future updates of the
426 map once the gap in Central Ukraine is filled. The lack of data in the Pannonian plain has led to
427 poor correlation between predicted pH and Ca²⁺ values (Fig. 4). Such a poor correlation and
428 sometimes low density of data apply also for some other lowland regions, such as the Danube
429 plain in S Romania, Po valley in Italy and valleys around the Duero, Ebro, and Tagus rivers in

430 Spain. Apart from eutrophication, this pattern may be caused by the imbalanced distribution of
431 groundwater-dependent habitat types in our data set. Unlike mountain regions, the data for these
432 lowlands were largely taken from the FOREGS Geochemical Atlas of Europe (Salminen et al.
433 2006) and national groundwater databases, i.e. largely from stream water. Considering the major
434 purpose of our map (creating new data sources for ecological modelling of fens and springs)
435 these regions are less crucial for biodiversity modeling because they have largely been
436 transformed to arable land or they are too dry. On the other hand, caution is needed when
437 interpreting the maps in an ecological sense. The extremely high pH (> 8) and Ca^{2+} ($\ln [\text{Ca}^{2+}] > 4$;
438 i.e., $\text{Ca}^{2+} > 55 \text{ mg.l}^{-1}$) values that occur in lowlands visually govern the map, but for ecological
439 differentiation of groundwater-dependent habitats in Europe the differences within the middle
440 part of the gradient, i.e. between pH 5.5 and 7.0, are much more important (Malmer 1986,
441 Wheeler & Proctor 2000, Hájek et al. 2006, Rydin and Jeglum 2013).

442 Our data set is expected to be amended in the future, as more studies will be published and more
443 data will be available, so further versions will be accessible in the open repository. New data will
444 help to improve predictions for those regions with relatively lower sampling effort, and also
445 those with lithologically heterogeneous landscapes. Future updates of the model may also focus
446 at finer spatial resolution (e.g., 100 to 250 m) but this will require to increase the spatial accuracy
447 of the calibration data and the predictor variables, as in some Central-European areas (Le et al.
448 2019, Chuman et al. 2019). Although we tested several variables with potential predictive effect
449 on groundwater pH and Ca, many of them had lower explanatory power (e.g. Ballabio et al.
450 2019) or they were redundant with the soil pH layer we used (i.e. previous versions of soil.grids
451 using the same data sources). The low predictive value of other predictors with potential
452 predictive value, like slope, elevation or other precipitation variables, is probably related to their

453 broad spatial scale, with values averaged at 1 km grid resolution having little impact to
454 discriminate groundwater variation at the landscape level (Jiménez-Alfaro et al. 2018b). This
455 contrasts with the predictive value of the lithological layer, which is however based on a similar
456 spatial (but originally vectorial) resolution. However, lithological bedrock is generally more
457 homogeneous at the landscape scale, with the only exception of certain geological complex
458 regions. These findings suggest that future improvements of our models will depend on the
459 quality of new lithological (or related soil chemistry) variables with direct effect on groundwater
460 pH and Ca. We also note that the lithological map we used here is simplified to large units, while
461 many regional and national geological maps are being produced at finer details. The combination
462 of such new predictors with accurate calibration data at the continental level and at fine scale
463 resolution seems the most likely opportunity to produce significantly better models, since the
464 influence of spatial-dependent (kriging) effects is rather limited by the distribution of sample
465 points.

466

467 **Conclusions**

468 Here, we provide the first European map of groundwater pH and Ca²⁺ content. We collected as
469 even as possible distributed field measurements of water pH and Ca²⁺ or EC from European
470 groundwater-dependent habitats, with relatively higher sampling effort in regions rich in
471 endangered groundwater-dependent ecosystems (springs, fens). Despite the general high
472 accuracy of our models, we note that prediction uncertainties may affect the reliability of models
473 computed with both Random Forests and kriging (Hengl et al. 2018; Szatmári & Pásztor 2019).
474 Another source of prediction uncertainty is related to the quality of the original chemical
475 measurements and the georeferentiation of their geographic position. Moreover, the predictor

476 variables rely on spatial models (soil pH, precipitation) or broad geographic maps (lithology)
477 which are based on their own uncertainties and assumptions. Future Improvements of
478 groundwater pH and Ca estimates should therefore consider a more accurate set of response
479 variables and fine-scale predictors, preferably including lithology and soil pH (i.e., variables
480 surrogating bedrock chemistry) and precipitation sum (i.e., residence time of groundwater).
481 Despite potential uncertainties and data gaps, this study uses an unprecedented combination of
482 data to provide freely accessible and realistic maps that can be used in any kind of spatial
483 modelling, showing better resolution and fewer gaps than previously published maps. The
484 character of our input data, which are also freely accessible, predetermines our map for being
485 used in ecological modelling to address the distribution and diversity of groundwater-dependent
486 ecosystems and associated species. We even believe that our maps could be also suitable for
487 ecological modelling of other than groundwater-dependent habitats. It may mirror the bedrock
488 chemistry better than the map of soil pH, because soil pH is a resultant of pedogenetic processes,
489 which are tightly associated with the character of the vegetation cover itself (Miles 1985,
490 Duchaufour 2012), We conclude that our European maps of near-surface groundwater pH and
491 EC provides the best solution currently available for modelling the biodiversity of groundwater-
492 dependent ecosystems, especially at the continental or supra-regional scale.

493

494 **Data availability**

495

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

499

500 **Code availability**

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

502

503 **Sample availability**

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

506

507 **Team list**

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518

519 **Author contribution**

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

524

525 **Competing interests**

526 The authors declare that they have no conflict of interest.

527

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542

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544

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