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A New Bayesian Inference Methodology for Modeling Geochemical Elements in Soil with Covariates. Characterization of Lithium in South Iberian Range (Spain) Pablo Juan¹ ¹ University Jaume I of CastellAn Received: 15 December 2015 Accepted: 2 January 2016 Published: 15 January 2016

8 Abstract

⁹ When the scientific need to model geochemical elements in soil, is using geostatistical

¹⁰ methodologies, for instance krigings, but we can use a new possibility with Bayesian Inference.

¹¹ The models for the analysis were specified by the authors and estimated using Bayesian

¹² inference for Gaussian Markov Random Field (GMRF) through the Integrated Nested Laplace

¹³ Approximation (INLA) algorithm. The results allow us to quantify and assess possible spatial

¹⁴ relationships between the distribution of lithium and other possible explanatory elements. Are

these other elements significant to the study? We believe the methods outlined here may help to find elements such as lithium, as well as contributing to the prediction and management of

¹⁷ new extractions or prospection in a region in order to find each chemical element. The

¹⁸ application for the modeling is to study the spatial variation in the distribution of lithium and

¹⁹ its relationship to other geochemical elements is analyzed in terms of the different possibilities

²⁰ offered by geographical and environmental factors. All in all, Lithium presents many

²¹ important and meaningful uses and applications such as: ceramics and glass, electrical and

²² electronics standing out lithium ion batteries, as well as a lubricator for greases, in metallurgy,

²³ pyrotechnics, air purification, optics, organic and polymer chemistry, and medicine. This

study aims to examine the distribution of lithium in sediments from the area of Beceite, in the
Iberian Range and the Catalan Coastal Range (Catalànids), within the geological context of

²⁵ Iberian Range and the Catalan Coastal Range (Catalànids), within the geological context of
 ²⁶ the Iberian Plate. The Atlas Geoquímico de España (IGME, 2012) was used as the main

²⁷ geochemical data bank in order to carry out a statistical analysis study.

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29 Index terms— bayesian inference, iberian range, lithium, soil.

30 1 I. Introduction

ithium is a relatively rare element. Although lithium is found in many rocks and some brines, it is always present
 in very low concentrations. Lithium presents many important and meaningful uses and applications and the most
 important is electrical and electronics standing out lithium ion batteries, but not only this application, it is used

³⁴ as a lubricator for greases, in metallu rgy, organic and polymer chemistry or medicine. The average amount in

the earth's upper crust was estimated by Vine (1980) to be about 20 ppm, although others have reported values as low as 7 ppm (igneous rocks 6 ppm, sedimentary rocks 11.5 ppm) and as high as 60 ppm, this element ranking

as low as 7 ppm (igneous rocks 6 ppm, sedimentary rocks 11.5 pp
27th on the list of abundance (Garrett et al., 2004).

Because of its small ionic radius, lithium (+1, 0.68 Å) tends to replace magnesium (+2, 0.66 Å) or iron (+2, 0.74 Å)

³⁹ Å) in silicates rather than the much larger alkali ions, sodium (+1, 0.97 Å), potassium (+1, 1.33 Å), rubidium

40 (+1, 1.47 Å) or caesium (+1, 1.67 Å), to which it is chemically more closely related (Ju-chin- Chen, 1998).

Three main types of lithium ore can be differentiated: brines and related evaporites, pegmatites and sedimentary 41 rocks. Brines containing lithium make up 66% of the world's lithium resources, pegmatites make up 26%, and 42 sedimentary rocks account for the remaining 8% ??Grubber et al., 2011). The deposits formed are related 43 to lithium's greater capability for solubility than most other cations. Accordingly, it can sometimes be found 44 concentrated in flowing and cooling magma and/or its associated aqueous solutions, besides also being present 45 in evaporating brines. Lithium is associated with sedimentary rocks, including clay and lacustrine evaporites. 46 In clay deposits, lithium is a constituent of clay minerals, such as smectite. A remarkable smectite in this 47 environment From the geochemical point of view, one of the main goals of this paper is to study the relationships 48 of lithium with several other elements in an attempt to identify and establish trends in geochemical behaviour, 49 within the geological context of a linking area under the influence of two tectonic domains, namely: the East 50 Iberian range and the Catalan Coastal range. This area was remarkably tectonized by the Alpine orogeny in 51 the Late Mesozoic and Cenozoic. Going a step further, another important aim is the study of the distribution 52 of lithium, in relation with the potential atomic exchanges that occur in several crystalline structures -especially 53 with the existence of Fe, Mg, K and Na. Furthermore, this study is a part of a more ambitious project that 54 embraces a larger area. 55

In this paper is to present the prediction and the basic features of the Integrated Nested Laplace Approximation (INLA; Rue et al., 2009) approach as applied to geochemical spatial data. In previous studies of geochemical elements, it has been in Geostatistical Analysis context (Jordan et al., 2004; Juan et al., 2011), but now the INLA approach can be employed to improve the results. The INLA is designed for latent Gaussian models ranging from linear mixed to spatial and spatio-temporal models. For this reason, the INLA can be successfully used in a great variety of applications.

The paper is structured as follows. First, in Section 2 we review the main characteristics of the geochemical analysis of the data. In Section 3 the sampling data are presented, and this is followed by Section 4, where we detail the analysis of the data. We provide an overview of the theory underlying the INLA and we review the SPDE approach to dealing with spatial data in Section 5. The practical application to area-level data is presented

in Section 6. After that, in Section 7, issues are discussed and some conclusions are set out.

⁶⁷ 2 II. Geological Background

In order to understand better the geochemical content and its arrangement throughout the range is revised ina short way the geological context. In this way, the study area lies within the geological context of the Iberian

⁷⁰ Plate, close to the Iberian Range and the Catalan Coastal Range (Catalànids), and forms a single tectonic unit ⁷¹ (Vera, 2004). The east-central margin of the Iberian Peninsula a relief which often exceeds 1000 m. in height

⁷² as the result of two alpine structures that were partially destroyed there (Fig. 1). Both structures, the Iberian

73 Range and the Catalan Coastal Range, converge at their eastern and southern ends to form the Maestrat, where

⁷⁴ the area known in Spanish as "zona de enlace" is located (Fig. 2).

75 3 Figure 2 Geological map of the Iberian Range and

⁷⁶ Catalan Coastal Range (Vera, 2004).

It is made up of a fold-and-thrust belt running NW-SE (Iberian Range) and NE-SW (Catalànids) formed as a result of the Alpine reversion of the Mesozoic Iberian Basin. Its structure includes the fractured Hercynian basement, which has Alpine structures superimposed upon it that later underwent Neogene distension, especially in the south-east of the Iberian Range.

Most of the structural features of the Iberian Range are the result of the interaction of two factors: first, the reactivation of old compression fractures in the late Hercynian, NW-SE and NE-SW, and, secondly, the relationship between the cover and the underlying basement (Guimerà and Alvaro, 1990).

The tectonic style develops three structural levels characterized by the involvement of stratigraphic succession and deformation behaviours. The lower level consists of the Hercynian basement and overlying Permian and Triassic, while the upper level includes the Mesozoic (Jurassic and Cretaceous) and Cenozoic cover (bed) (preand syntectonic PaleogeneThe two are separated by a detachment level located in marks materials and evaporatic materials from the middle and upper Triassic, and hence they develop more folds and thrusts, and their main characteristic is their plastic behaviour and incompetence.

Between the Oligocene and lower Miocene a new extensional process was produced that is framed within the context of the Western European rift system. The distension mainly affects the eastern margin of the Iberian Plate and causes a complex system of horsts and grabens, parallel to the Mediterranean coast, related to the formation of the Valencia trench (Fontboté et al., 1990; ??ock and Guimerà, 1992)

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4 III. Sampling Information

The soil samples belong to the Geochemical Atlas of Spain, produced by the IGME in 2012, which studies the geochemical components of soils and other superficial materials in the Spanish territory. A multielement geochemical characterization of soils and superficial materials was performed. These georeferenced data were

99 introduced into a high-resolution geo-database. 1. It should be based on samples of different media, which

include soils (two depths), stream sediments, and floodplain sediments (2 depths). These later, in alluvial 100 systems of a certain magnitude, will make it possible to determine the original uncontaminated values, prior to the 101 industrialization period, and the variations that have taken place over time by comparison with present and future 102 values. 2. It must be multi-element, including a wide range of chemical elements (63 in this case), comprising all 103 those that are potentially dangerous for the ecosystem and those of any interest to geology and mineral resource 104 research. They will be analysed by reliable techniques and with very low detection limits. Determination of the 105 total or almost total contents and of the partial contents that will allow the easiest extractable or bio-available 106 part to be estimated. This latter point is of particular interest for the environmental analysis and would allow 107 an assessment of pollution processes to be carried out. 3. It should be available in digital form, to facilitate the 108 integration of the geochemical information with other datasets (geology, metallogeny, land uses, animal or human 109 morbidity and mortality). 4. It must be standardized across national boundaries, as many of the problems and 110 concerns that arise regarding the surface make it necessary to go beyond the frontiers and perform analyses on 111 a more global scale. The follow-up of the IGCP project 259 recommendations and the protocols derived from 112 the FOREGS project entitled "Global Geochemical Baselines for Europe" will act as a guarantee for compliance 113 with this requirement. 114

115 5

:

Co, Ti, Zr, Nb, Ta, Ni, Cu, Pb, Zn, Sn, Rb, Cd, and Sb. Our goal is to analyse the concentration of lithium and its relationship with the concentration of the other elements considered in the study, considering those elements as covariates. The main objective is to assess how the covariates are related to the concentration of lithium in the study area and to asses which of such elements show a statistically significant relatonship with lithium.

Figure 4 shows the histogram of lithium concentrations as well as a map showing the sampling locations and the observed lithium concentrations at each site. The data show a non-symmetric distribution with a shape that not resembles the normal distribution. It shows a Gamma distribution. Bottom: Distribution of lithium in the

123 region.

¹²⁴ 6 V. Methodology Statistical Inference

The basic idea behind the Bayesian approach is that in any statistical model of a random phenomenom, only one source of uncertainty exists, which can be described by suitable probability distributions of model parameters (Box and Tiao, 1973). Thus, there is no fundamental distinction between observable data or unobservable parameters, which are also considered random-vector quantity

The uncertainty about the value of the parameter vector before observing any data is described by a prior distribution

The inferential process combines the prior and the likelihood of observed data (x) given the model, to derive the posterior distribution

x) , which is typically, but not necessarily, the objective of the inference (Bernardo and Smith, 2000; Lindley,
 2006).

There are several advantages to the Bayesian approach: for instance the specification of prior distributions allows the formal inclusion of information that can be obtained through previous studies or from expert opinion; the (later) probability that a parameter does/does not exceed a certain threshold is easily obtained from the posterior distribution, thus providing a quantity that is more intuitive and interpretable than a frequentist pvalue. In addition, within the Bayesian approach, it is easy to specify a hierarchical structure on the data and/or parameters, which presents the added benefit of making predictions for new observations and missingdata imputation relatively straightforward. $+ = GF \ge x \ge b$ a Gamma $\ge F \ge i \ge 2$, (1)

where Fi is a vector of covariates made by the concentration of the other elements considered in this study, ? 142 a vector of coefficients and x a latent Gaussian random field, with a covariance matrix function with parameters 143 ?, ? and ?x2. With the Gamma likelihood, In this case, for all the models, we obtained the parameters of the 144 posterior distribution of the intercept (?), the dispersion parameter of the Gamma likelihood, the latent field 145 parameters log(?) and log(?), the fixed coefficients (?) for the covariates (Al, K, Na, Ca, Mg, Fe, S, Mn, Ba, 146 Co, Ti, Zr, Nb, Ta, Ni, Cu, Pb, Zn, Sn, Rb, Cd, Sb) and the values of (1/?e2). The nominal variance and the 147 nominal range (distance with correlation equal to 0.1) are also presented. For the spatial covariance structure 148 we used the Matérn family of covariance functions and a nugget term, over j.) '() 1(),,, '(), cov(2222' 149 j j I r r j j M j j = ? + ? = ? ? ? ? ? ? (2)150

¹⁵¹ 7 c) Stochastic Partial Differential Equation Approximation

The SPDE approach allows a GF with the Matérn covariance function to be presented as a discretely indexed spatial random process (GMRF), which has significant computational advantages. Gaussian Fields are defined directly by their first-and second-order moments and their implementation is highly time-consuming and leads to the so-called "big n problem" (Lindgren et al., 2011). The idea is to construct a finite representation of a Matérn field by using a linear combination of basis functions defined in a triangulation of a given domain D. This representation leads to the SPDE approach given by (2), which is a link between the GF and the GMRF, and allows the spatio-temporal covariance function and the dense covariance matrix of a GF to be replaced with a neighbourhood structure and a sparse precision matrix, respectively, both of which are typical elements that
 define a GMRF. This, in turn, produces substantial computational advantages (Lindgren et al., 2011).

More particularly the SPDE approach consists in defining the continuously indexed Matérn GF X(s) as a

discrete indexed GMRF by means of a basis function representation defined on a triangulation of the domain D:? = n l l l s s X 1) () (?? (3)

where n is the total number of vertices in the triangulation; $\{?|(s)\}$ is the set of basis functions and $\{?i\}$ are the zero-mean Gaussian distributed weights. The basis functions are not random, but rather are chosen to be piecewise linear on each triangle:? ? = elsewhere vertix at s l 0 1) (? (4)

¹⁶⁷ The key is to calculate {?i}, which reports the value of the spatial field at each vertex of the triangle.

The values inside the triangle will be determined by linear interpolation (Simpson et al., 2011). Year 2016 ($G \otimes 2016$ Global Journals Inc. (US)

170 In our case, we obtain this representation of the mesh with the response variable, see Fig. 5. We can see

171 the relation between the data and the vertex for the implementation. This is the usual way of representing the

172 regular region data when using INLA//to be able to use INLA.

173 8 Results

At this point, we know the data and the statistical framework and we have presented the statistical process with 174 the applied study. First of all we can observe the advantage of this process because, in general, the processing 175 time is long. In our case, the time needed for pre-processing: 8.2529 seconds, for running each INLA model: 176 3.4478 seconds, and for post-processing: 0.4212 seconds. We have produced many previous models but the 177 one presented here is the final version used, with this processing time. In this study, different models have been 178 tested. The first one (M1) includes only the spatial effect, the second one (M2) includes the spatial effect and the 179 covariates (chemical elements) and, finally, the third one (M3) has only the covariates without the spatial effect. 180 M was the Matérn function (R-INLA, 2011); ?2 denoted the sill (the total variance of the innovation process); r 181 2? 2 was the variance of the spatially correlated portion of the process; (1-r 2)? 2 corresponded to the nugget 182 (the variability that is unique to a given station); ? was the range of the process (the size of the region where the 183 process was significantly correlated); and ? was the smoothness of the process (specifically, we tried ?=1 for the 184 model, R-INLA project). All the analyses were carried out with the freeware statistical package R (version 3.0.2) 185 (Lindgren et al., 2011) and the R-INLA package. Now, the results of the model, the fixed effects, the random 186

187 effects and the comparison between the real and the predicted data are presented.

¹⁸⁸ 9 a) Fixed effects

189 In our models, the fixed effects are presented by the covariates.

Table ?? shows the mean, the standard deviation and the 2.5, 50 and 97.5 percentiles for the Intercept of the models (elements other than lithium). In Tables ??Ia and IIb, we can see positive values for the elements that increase the value of lithium and the negative values for the elements that decrease the amount of lithium. But the most important thing is to check which one has a significant statistical effect.mean

For Model M2, we can see that only Nb offers a significant result and Zr, Ta, Cu, Pb, Zn and Rb are elements close to being representative in the study of lithium.

For Model M3, we can see that K and Nb are positively significant in the study of lithium and Ta is significant in the opposite way.

¹⁹⁸ 10 b) Random effects

In this case, the hyperparameters correspond to the Gamma distribution and, since we do not know the values (Bayesian inference), they could be compared with the posterior. We have only the structure. In this case, the parameters are 1/?e2 or marginal variance nominal, marginal kappa and marginal range nominal. ??ODEL If we want to know the form of the structure of the parameters, they can be seen in Fig. 6. For M3, the parameter associated to the Gamma likelihood has a value of 10.7411 with a standard deviation of 2.8977.

We can see the structure of this parameter in Fig. 7 below. Fig. 8 shows the relation between the observed and the predicted values for the three models. The results are clearly optimum in the second one, M2. In Table **??II** we can also see the results for the correlation coefficients and for the root-mean-square error (RMSE). In the same way, the results are satisfactory and the best model is the second one (M2). In all cases, the highest

²⁰⁸ predicted values of lithium under these conditions are found for the North of the Beceite area.

²⁰⁹ 11 Discussion and Conclusion

This work presents the predicted values of lithium in the study region. They were obtained with a new methodology in Bayesian probabilities called INLA. The results showed that the lithium values could be studied by knowing the different covariates.

Henceforth, from the results of the model with the fixed effects, the random effects, and the comparison between real and predicted data it is possible to appreciate that lithium does not increase or decrease its concentration noticeably with any other element. On the one hand, if model 2 (M2), which fits the data better, is taken as a reference, at most it is possible to appreciate a slight increase in niobium and a minor decrease in thallium.

On the other hand, model 3 (M3) showed an increase in potassium and niobium, and lithium decreased in the 217 presence of thallium. Lithium might have been expected to connect with some specific geological materials of 218 the area, such as clay minerals, with an important Al and K content and to a lesser extent Fe. It formed part of 219 220 clay minerals, frequently appearing mixed with sandstones. In relation with the age of the materials, lithium was related mostly to Lower Cretaceous and Tertiary deposits, which cover an important surface area of the region, 221 second to colluvial and alluvial Quaternary materials, and finally, to a much lesser extent, to Triassic formations. 222 In this study, we have focused in the content of Lithium at soils and sediments in relation to several elements, 223 especially heavy metals, in order to determine its geogenic presence through the INLA modelization, taking into 224 account that Lithium shows different geochemistry in different origin deposits, especially marine, as the main 225 studied area. 226

Finally, regardless the results, the use of the model could provide new insights in order to face other geochemist studies in soils and sediments for environment, agrarian, or mining prospections.

As we known, Lithium presents many important uses and applications such as: ceramics and glass, electrical and electronics standing out lithium ion batteries, as well as a lubricator for greases, in metallurgy, pyrotechnics, air purification, optics, organic and polymer chemistry, and medicine. For all of these applications, the results we got for predicting the quantity and the location of Lithium it should be taken into account.

233 On the other hand, the phyllosilicates would contain potassium and would not appear with sodium clays. 234 Furthermore, lithium would not be related with brines, for instance the marine Keuper deposits represented in the area, which contain sulphate and chlorine minerals (Fig. 12). ??985). All in all, the geological context 235 was defined during the Late Jurassic to Early Cretaceous rift stage, where several sub-basins were developed (in 236 the Spanish provinces of Castelló, Teruel and Tarragona), which were filled by fluvial to transitional sandstones 237 and lacustrine carbonates (Weald facies), later evolving to marine limestones (Urgon facies). These sub-basins 238 trapped clastic sediments from the Tertiary, where potential source areas are metamorphic and pre-rift Triassic 239 and Jurassic sedimentary cover rocks. 240

In summary, the most important idea is that the presence of a geological element in a region could be predicted

²⁴² when we know the others using the INLA procedure for the statistical inference, which is faster than the use of MCMC techniques.



Figure 1: Figure 1

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Figure 2:



Figure 3:



Figure 4: Figure 3



Figure 5: Figure 4



Figure 6:



Figure 9:



Figure 10: Figure 6



Figure 11: Figure 7







Figure 13: Figure 9

	sd	0.025quant	
M1	3.1050.165	2.774 $3.1043.449$	
M2	1.0651.312	- 1.0843.606	
		1.590	

Year M3 For the covariates, the fixed effects are presented in models M2 and M3 (in bold, the significative 0 2016 44 Fixed effects of the model M2. Mean, sd and 2.5, 50 a

[Note: $G \ b:A$]

Figure 14: Table 1 :

 $\mathbf{2}$

Figure 15: Table 2

3

Model	Correlation coefficient	RMSE
M1	0.7833763	16.17654
M2	0.9909199	2.59907
M3	0.9667658	5.061028

[Note: c) Prediction MapFinally, we can see the prediction maps of the latent field and the response variable for M1, with the corresponding standard deviation and the response variable for M2 (Fig.[9][10][11]. As an example of the application of this methodology the reader is referred to Jordan et al.(2004).A]

Figure 16: Table 3 :

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