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3	A new perspective in radon risk assessment: mapping the geological hazard as a first step to
4	define the collective radon risk exposure
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19

# 20 Graphical abstract (mandatory)



21 22

# 23 Abstract (300 words)

24 Radon, a radioactive gas, is the largest source of ionizing radiation exposure for humans. As a

25 result, its accumulation in confined environment can pose serious threats to health. In order to

address the issue of radon exposure in dwellings, the European BSS directive 2013/59/EURATOM

established national reference level and guidelines to define Radon Priority Areas (RPAs). The 27 Geogenic Radon Potential (GRP) is considered an effective hazard indicator for assessing the 28 potential accumulation of this gas in buildings from geological sources. Several approaches, 29 including multivariate geospatial analysis and artificial intelligence algorithms, have been applied to 30 generate spatial continuous maps of the GRP based on soil gas point measurements and other 31 related geo-environmental proxies. 32 The goal of this study is to map GRP of the central sector of the Pusteria Valley by a supervised 33 machine learning algorithm (Random Forest), and use this map as a basis for identifying RPAs. The 34 Pusteria Valley (North-eastern Italy) has been chosen as a pilot site due to its well-known 35 geological, structural, and geochemical features. We then incorporate land use and population as 36 37 vulnerability and exposure factors, respectively, to provide a final risk map. Results indicate that high Rn risk areas are associated with GRP values higher than 50 kBqm<sup>-3</sup>. The use of the GRP map 38 39 as a hazard component of radon risk leads to a new geological definition of RPAs. GRP is an important tool for mapping Collective Risk Areas (CRAs) since the BSS directive only addresses 40 41 the identification of Individual Risk Areas (IRAs). 42 43 6 Highlights 44 Mapping the Geogenic Radon Potential (GRP) using a robust supervised machine learning 45 • 46 (i.e., random forest) as the most important spatial predictor for Indoor Radon Concentration (IRC). 47 • Apply the risk definition (i.e., product of hazard, vulnerability and exposure) in order to 48 define the CRAs by intersecting the GRP map (hazard) with the type of location 49 (vulnerability) and total population (exposure) 50 • Construct geological-based Collective Risk Areas (CRAs) map starting from GRP, coupled 51 with land use (location type) and population density of the census tracts to define those areas 52 53 subject to territorial planning by local authorities 54 **Keywords** 55 Geogenic Radon Potential, Machine learning, Pusteria Valley, Radon risk, Collective risk areas 56 57 Abbreviations: GRP = Geogenic Radon Potential; RPA = Radon Priority Area; IRC = Indoor 58 Radon Concentration; BRS = Background Radon Source; TER = Tectonically Enhanced Radon; 59 SRE = Surface Radon Exhalation; CRA = Collective Risk Area; IRA = Individual Risk Area; 60

- 61 SGRC = soil gas radon concentration; GRHI = Geogenic radon hazard index; BSS = Basic Safety
- 62 Standards, TGDR = Terrestrial gamma dose rate
- 63

# 64 Introduction

Radon (<sup>222</sup>Rn, hereafter Rn) is a radioactive gas considered the major source of ionizing radiation
exposure for the population. Its potentially harmful effects on human health have been extensively

- 67 documented (WHO, 2009). In particular, radon gas represents a serious hazard when it accumulates
- 68 in indoor environments (Indoor Radon) such as in residential houses and workplaces.
- 69 Exposure to indoor radon is a serious problem that has prompted Europe to introduce legislation
- 70 (Basic Safety Standards Directive 2013/59/EURATOM) which, on the one hand, establishes
- 71 maximal national reference level aimed to reduce the exposure to Indoor Radon Concentration
- 72 (IRC); on the other hand, promotes the public administrations to define Radon Priority Areas
- 73 (RPAs). For this reason, it is fundamental to identified areas characterised by the highest Rn hazard
- 74 for the population.
- 75 The concentration of radon gas in the environment can vary depending on the geological
- characteristics of an area. Radon produced within the Earth can migrate through permeable
- pathways (faults and fractures) in rocks and soil, or dissolved in groundwater, up to the shallow
- renvironment. Then, radon can diffuse into the atmosphere, or enter buildings. Geogenic Radon
- 79 Potential (GRP) can be considered an optimal Rn hazard indicator. It is conceptualised as "*what*
- 80 Earth delivers in terms of radon" from the geogenic sources (e.g., radionuclides content, faults and
- 81 fractures) towards the atmosphere (Bossew, 2015; Bossew et al., 2020).
- 82 In particular, GRP is characterised by the interaction of three natural processes:
- the Background Radon Source (BRS), the process that produces Rn isotopes (<sup>222</sup>Rn and <sup>220</sup>Rn)
- 84 through the natural decay of uranium (U) and thorium (Th), which are found in varying
- 85 concentrations in rocks and soil;
- the Tectonically Enhanced Radon (TER, Benà et al., 2022), the additional process allowing radon
- to migrate more easily to the surface through permeable pathways (e.g., faults and fractures in the
- crust) from deeper sources, caused by the stress increase and pressure conditions associated with
- 89 tectonic activity;
- the Surface Radon Exhalation (SRE), the process by which radon gas is released from the ground
- 91 into the surrounding environment. SRE includes the variables that affect radon movement in the
- shallow soil up to the soil/atmosphere interface (e.g., land morphology, soil permeability, humidity
- and temperature). This quantity of radon, which has not yet been extensively measured, represents
- 94 the amount of radon that could potentially enter buildings.

95 However, BRS and TER represent the dominant geological radon sources.

96 Over the years, several approaches have been applied to estimate the GRP over an area (e.g., Neznal

97 et al., 2004; Bossew et al., 2015; Pasztor et al., 2016; Ciotoli et al., 2017; Giustini et al., 2019;

98 Petermann et al., 2021; Coletti et al., 2021).

99 Neznal et al. (2004) proposed an early method to define the GRP that has been widely used due to

its simplicity up until this day. The method was based on the measure of two quantities: the Rn

101 concentration in the soil and the soil permeability. Equation 1 reports the Neznal formula to

102 calculate the GRP (dimensionless):

103

104

 $GRP_{Neznal} = \frac{SGRC}{-\log_{10} k - 10}$ 

(1)

105

where SGRC is the soil gas radon concentration (kBqm<sup>-3</sup>) measured at a depth of about 0.8 m, and k is the soil gas permeability in  $m^2$ .

108 More recently, Pasztor et al. (2016) and Ciotoli et al. (2017) applied multivariate geospatial analysis 109 (regression kriging and geographical weighted regression, respectively) for GRP modelling by

(regression kriging and geographical weighted regression, respectively) for GRT moderning

using SGRC and selected environmental proxies for the first time.

111 In the past three years, researchers have developed more advanced multivariate techniques, such as

regression kriging (Coletti et al., 2021) and machine learning (ML) algorithms (e.g., random forest,

etc.), which include several predictors associated with the geogenic Rn component (Petermann et

al., 2021). However, it is important to emphasise that all these regression techniques require a

115 response variable (i.e., SGRC or IRC).

116 However, many European countries lack sufficient SGRC measurements and permeability data to

support the mapping of GRP. As a consequence, the calculation of a Geogenic Radon Hazard Index

118 was proposed by Bossew et al. (2020). The concept of the GRHI arose from the need to determine a

specific indicator using regionally accessible geological variables. However, it is difficult to

120 maintain consistency between GRHI scores in neighbouring regions when using multiple predictors.

In other words, GRHI values for areas with comparable geological factors but different data sourcesshould be nearly equivalent.

123 In order to produce consistent maps, Cinelli et al. (2015) proposed a first method to assign weights

to continuous or categorical input variables (covariates) based on their contribution to the index.

125 The weighted "mean class" is then used to calculate the Geogenic Radon Hazard Index (GRHI), a

dimensionless quantity. The weights allocated to each covariate are determined by the observed

127 correlations with the GRP in regions where GRP data are effectively accessible. Another way to

avoid the issue is the application of other more subjective techniques which does not require a 128 129 response variable (i.e., Spatial Multi Criteria Decision Analysis, SMCDA, Ciotoli et al., 2020). Since the GRP represents the amount of radon that could potentially enter buildings, it is considered 130 as the most significant spatial predictor of the IRC; therefore, it is crucial to map the GRP as 131 accurately as possible using a robust methodology. In this regard, the BSS European Directive 132 59/2013, transposed into Italian law by Legislative Decree n.101/2020, emphasizes further the 133 identification of RPAs, originally defined as those areas where the annual average IRC in a 134 significant number of dwellings is expected to exceed the reference level of 300 Bqm<sup>-3</sup>. However, 135 the concept and interpretation of "significant number of buildings" in the European Directive 136 remained unclear. 137

In a recent study, Petermann et al. (2022) emphasized that rather than the collective concept of geogenic risk, the interpretation of "significant number" of buildings is factually based on the concept of geogenic hazard, in that it is assumed to mean something like a relevant percentage of buildings within an area notwithstanding the number of houses or of people affected.

The latter accounts for the number of people affected, based on the notion that the possible detriment caused by Rn exposure in an area - the number of lung cancer fatalities - depends on hazard and presence of people who can be harmed. After all, according to the BSS, it is the detriment that should be reduced by Rn policy.

On the other hand, staying with the hazard- or individual risk-type notion of RPA, there is no 146 147 uniform decision at the regional scale regarding the selection of the reference level (RL) and the 148 threshold of probability percentage  $(p_0)$  of buildings exceeding the RL. In general, the majority of European nations (including Finland, Germany, Greece, Montenegro, and Spain) adhere to the 149 European Directive adopting the recommended reference level of 300 Bqm<sup>-3</sup> and a common 150 probability threshold of 10% (Bossew, 2018). Particularly, Germany only considers IRC 151 152 measurements in rooms on the ground floor of buildings with basements, whereas Spain only considers measurements in rooms on the ground or first level. Ireland has an RL of 200 Bqm<sup>-3</sup> and a 153 p<sub>0</sub> of 10%. Other countries, including Austria and Switzerland, define distinct priority levels based 154

- on RL and measurer IRC (Bossew, 2018). Italy has an RL of 300 Bqm<sup>-3</sup> and a  $p_0$  of 15% (D. Lgs. n.
- 156 101/2020). A map of the confusing diversity of RPA definitions across Europe has been shown in
- 157 Bossew and Suhr (2023, see Fig. 2 in the cited paper).

158 As reported in Bossew et al., 2021, the goals of radiation protection from Rn indoor are twofold:

to protect people from high Rn exposure in order to reduce individual risk (even if few people are involved);

- to avoid high exposure to the community, because the harm to society is proportional to the
  collective risk.
- 163 But how the European Community protects people from Rn risk?
- 164 In order to limit radon exposure and thereby lower the population's likelihood of contracting lung
- 165 cancer, the regulatory program aims to identify RPAs and implement ad-hoc mitigation plans.
- 166 European legislation aims to reduce the detriment from Rn exposure (i.e., the number of lung cancer
- deaths) and as a consequence, reduce the collective exposure. Collective exposure can be assessed
- by introducing the concept of collective risk, a complement of the individual risk concept (as
- 169 interpreted by the BSS directive, "classical" RPA). Collective risk can be figured as consisting of
- 170 many little individual risk zones. Accordingly, the demand to identify RPAs focused only on
- building remediation rather than land use planning is a controversial subject affecting both
- 172 European and regional legislation.
- 173 In the light of these considerations, and in the absence of any specific approach for defining RPAs
- at the European level, we propose to map collective risk areas (or detriment, CRAs) as the
- 175 combination of the GRP, vulnerability and exposed factors, as a complement to mapping individual
- 176 risk areas (IRAs) associated with IRC (i.e., "traditional" RPA). Rn abatement policy must take care
- 177 of these areas in order to minimize the damage, while also preserving areas of high individual risk.
- 178 The goal of this study is to show the effectiveness of a CRAs map to define Rn risk areas in a test
- site, the Pusteria Valley (Bolzano, northern Italy), based on the GRP map (i.e., hazard factor)
- 180 generated by using multivariate machine learning (MML) technique (e.g., Random Forest, RF).
- 181 The Pusteria Valley was chosen because it is well-known from a geological, structural, and
- 182 geochemical point of view with a lot of available data (Benà et al., 2022), and also because it was
- already the target of IRC measurements (Minach et al., 1999). The obtained GRP map (hazard) was
- 184 merged with the land use type (vulnerability) and population (exposure) of census tracts available
- 185 from the ISTAT (i.e., *Istituto Nazionale di Statistica*) website in order to identify risk areas that may
- be subject to territorial planning by local authorities. The CRAs map can be merged with the RPAs
  map (*sensu stricto*).
- The construction of GRP maps is a fundamental tool for both Rn hazard and risk analysis and,
  according to a new more geological view, as a base map for the identification of Radon Priority
  Areas (RPAs). This is important for collective risk assessment (land-use planning and prevention),
- and for individual risk assessment (i.e., more strategic planning of indoor surveys, and specific
- 192 remediation actions).
- 193
- 194 **2. Methods**

- A dataset including different variables (e.g., response and predictors) was used to elaborate the GRP
  map of the study area by applying a ML technique (i.e., Random Forest, Breiman, 2001) and predict
- 197 the radon values at the center points of a 50x50 m fishnet. The resulting GRP map will be then used
- as the hazard factor in the risk equation (Eq. 2, see section 2.4) and multiplied by census tract
- indicator of land use and population density representing the vulnerability and the exposure factors,
- 200 respectively.
- 201 Figure 1 shows the flowchart of the applied procedures. Data were processed using ArcGIS Pro
- 202 3.1.2 (copyright 2023@ESRI Inc.) and Scikit learn library in Python PyCharm 2023.1.2 (Copyright
- 203 © 2010–2023 JetBrains s.r.o.).
- 204



205

Figure 1. Flowchart of the mapping process and procedures. SGRC = soil gas radon concentration; 206 perm = soil permeability; TGDR = terrestrial gamma dose radiation;  $^{220}$ Rn = thoron; CO<sub>2</sub> = carbon 207 dioxide concentration in soil gas;  $H_2O$  = concentration of radon dissolved in water; FD = fault 208 209 density; dtm = digital terrain model; slope = slope; aspect = aspect ratio; solar = solar radiation; loc type = location type: P dens = population density; GRP map = geogenic radon potential map; RPAs 210 = radon priority areas. SGRC, permeability, TGDR, thoron, carbon dioxide, radon dissolved in 211 water, faults, DTM were pre-processed in order to apply the forest-based classification and 212 regression (first step) to construct the GRP map (hazard factor). The GRP map was then multiplied 213 by the location type (vulnerability factor) and population density (exposure factor) to construct the 214 collective risk map. 215

- 216
- 217 **2.1 Dataset**

- The dataset used consists of one response variable (SGRC) and ten independent variables that were
  either measured on-site or derived from primary base-maps, available online from the Bolzano
  province's Geo-catalogue (http://geokatalog.buergernetz.bz.it/geokatalog/#!). These ten variables
- 221 were selected as potential predictors for machine learning regression models.
- 222 Soil gas surveys (<sup>222</sup>Rn, <sup>220</sup>Rn, CO<sub>2</sub>) (Benà et al., 2022), TGDR, and permeability measurements
- were collected on-site during two separate field campaigns in summer 2021 and 2022 under similar
- and stable climatic conditions. The Digital Terrain Model (DTM 2.5 m resolution) and fault
- distribution were obtained directly from the base maps of the Bolzano Province Geo-catalogue.
- The examined predictors were pre-processed using geospatial analysis to generate 50x50m raster
- 227 maps (see Fig. S1 in supplementary materials). The "Extract multi-value to point" tool of ArcGIS
- Pro was used to assign the values of the predictor grids to each observation of the collected soil gas
- samples. The obtained dataset, containing the predictors and the response variable (SGRC), was
- used to train the Random Forest (RF) model. Once the best model was found, it was applied to a
- regular point 50x50m fishnet corresponding to the centroids of the predictors' raster grid pixels. The
- final dataset consists of 27,758 points that includes complete information for all predictors. The
- following sections provide a detailed description of the response variable and the predictors.
- 234

### 235 **2.1.1 Response variable**

- SGRC (kBqm<sup>-3</sup>) was used as a response (dependent) variable in the Forest Regression algorithm to
  describe the GRP map. The Rn dataset consists of 278 measurements obtained in the field using the
  methodology and sampling pattern described by Benà et al. (2022).
- 239

# 240 2.1.2 On-site predictor variables

- 241 Five predictors were measured in the field: thoron (<sup>220</sup>Rn), carbon dioxide (CO<sub>2</sub>), TGDR,
- permeability and  $^{222}$ Rn dissolved in water. The same sampling technique and pattern adopted for the measurement of Rn concentrations in soil gas was also adopted for the measuring of thoron and carbon dioxide (CO<sub>2</sub>) (Benà et al., 2022).
- 245
- 246 TGDR measurements
- 247 TGDR measurement have been performed at 76 sampling points using the NaI γ-ray portable
- scintillometer (Scintrex GRS-500) pre-set on the total count rate window corresponding to an
- energy interval range between 80-3000 keV. The device was held 1 m above the ground for a
- 250 measuring time equal to the time needed to reach a 3% accuracy. The sensitivity factor of the

251 Scintrex GRS-500 is 3.40 cps/nGyh<sup>-1</sup> and allows the counting rates to be converted into the IS unit 252 of the gamma dose rate ( $\mu$ Sv/h, Giustini et al., 2019, 2022).

Geostatistical analysis (experimental variogram calculation, modelling, kriging) was used to obtain a prediction map of the TGDR (see Fig. S2 a and b in supplementary materials). This variable is used as a proxy of the BRS contribution (i.e. radionuclides content in rocks) of the geogenic radon component.

257

#### 258 *Permeability*

259 Soil gas permeability directly affects radon gas migration from the deep source (mainly by

advection along faults), and in the shallow soil (by diffusion prevalent mechanism) (Nuhu et al.,

261 2021; Neznal et al., 2005). High permeability allows the upward migration of radon, enabling its

exhalation to the atmosphere, while the presence of a shallow soil layer with low permeability could

- increase the accumulation of radon in the soil with a consequent decrease of exhalation rate at the
- soil-atmosphere boundary (Castelluccio et al., 2015; Johner et al., 2001). The radon concentration in

soil gas is directly dependent on the geological characteristics of the area (i.e., radionuclide content,

266 presence of fractures and faults) and can be strongly influenced by soil permeability in terms of soil

pore dimensions and soil water content (i.e., soil moisture) (Benavente et al., 2019; Lara et al.,

268 2015). Additionally, some other physical characteristics of soils, such as soil texture and grain size,

have a significant impact on the mechanisms of radon emanation and exhalation in the soil

environment (Huynh Nguyen et al., 2018; Yang et al., 2019).

271 In the study area, the soil permeability was measured at 76 sampling points with a permeameter

developed by the University of Roma Tre and directly connected to the soil gas sampling probe

273 (Castelluccio et al., 2015). The soil is assumed to be homogeneous and isotropic, and standard state

is considered; the air is assumed to be incompressible. The calculation of the final soil permeability

(k) is based on Darcy's equation and expressed in  $m^2$ . Geostatistical analysis (i.e., experimental

variogram calculation, modelling, kriging) was used to obtain a prediction map of the soil

277 permeability (see Fig. S3 a and b in supplementary materials).

278

279 Radon dissolved in groundwater

280 Dissolved <sup>222</sup>Rn was measured at 22 captured water springs in the study area. Water samples from

selected springs were already studied for their chemical-physical conditions by the *Agenzia* 

282 provinciale per l'ambiente e la tutela del clima - Laboratorio analisi acque e cromatografia

283 (Bolzano province) in 2022.

The water was sampled directly from the captured springs using glass bottles. Rn concentrations 284 were measured using RAD7 in the sniff mode connected to Big Bottle RAD H<sub>2</sub>O and drystick 285 (drierite desiccant) accessories. Prior to the measurements, the system was purged to guarantee that 286 287 the moisture (water content) inside the system was reduced to less than 10% humidity. The sampled bottle was then connected in a closed air-loop mode to the RAD7 (Durridge Company Inc.). During 288 system operation, continuous circulation gradually enriches the air contained in the closed loop with 289 the Rn dissolved in the water sample. Each measurement was performed with a 5-minute 290 integration period and was repeated until the difference between the last two readings is less than 5-291 292 10%. The final result was calculated by averaging the previous two integrations. Thiessen polygons was constructed to create a map of areas of influence around the water springs. Water springs 293 294 represents the centroid of the Thiessen polygons in which the measured dissolved radon value (i.e., the centroid) is assumed to be representative of the area underlying the entire polygon. The resultant 295 296 map was transformed in a 50x50m raster grid and used as predictor in the RF model.

297

#### 298 2.1.3 Derived predictor variables

### 299 Fault density

Faults and fractures represent the main pathway for radon, and other gases (CO<sub>2</sub> and CH<sub>4</sub>) migration in the subsoil from deep sources (see Ciotoli et al., 2007, 2014, 2017, 2020; Giustini et al., 2019). Therefore, the network of the fractured zone characterising the study area has been used as a proxy of the secondary permeability. The distribution of the main faults in the study area (Keim et al., 2013) was converted into a fault density (FD) map using the quadratic kernel density function (Silverman, 1986), as described in Benà et al. (2022).

306

#### 307 Digital terrain model

The Digital Terrain Model (DTM) of the study area (i.e., elevation) was used as a proxy of the 308 309 meteorological conditions which may strongly affect radon migration and exhalation mechanisms. The mobility of radon can be impacted by the presence of slopes, hills, and depressions, which can 310 311 alter air flow and soil pressure (Gundersen et al., 1992). Radon may not build up as much in areas with rough terrain because air circulation and groundwater drainage may be improved. On the other 312 313 hand, low-lying areas and depressions may act as radon traps, resulting in higher levels of the gas (Sukanya et al., 2021). Furthermore, Griffiths et al. (2014) highlighted how crucial it is to take 314 topographic interactions into account when estimating radon concentrations across different 315 geographical areas. The DTM (2.5 m/pixel) of the Bolzano province is available on the Geo-316

- 317 catalogue of the Bolzano province (*Rete Civica dell'Alto Adige*,
- 318 *https://geoportale.retecivica.bz.it/default.asp*).
- The "Surface Parameters" tool of Spatial Analyst" in ArcGIS Pro was applied to the DTM to create 319 maps of further potential proxies: slope, solar radiation (e.g., Areal Solar Radiation) and aspect 320 ratio. The slope can be used as a proxy of soil moisture and shallow soil meteorological conditions; 321 the solar radiation is used as a proxy of the microclimate/temperature. Aspect (i.e., slope exposure) 322 refers to the compass direction of the downhill slope faces in relation to the sun. Into details, slope 323 conditions such as the angle, aspect, and elevation of a land surface can strongly influence local 324 weather patterns and microclimates acting as a proxy of meteorological conditions in different ways 325 (e.g., sun exposure, rainfall distribution, wind patterns, temperature gradients), all of which may 326 327 impact radon generation and movement (Zalloni et al., 2018).
- 328

# 329 2.2 Predictor selection

- Predictor selection was conducted using Least Absolute Shrinkage and Selection Operator (Lasso) 330 331 regression. Least Absolute Shrinkage and Selection Operator (Lasso) regression is an extension of ordinary least squares (OLS) regression used in statistical modelling and machine learning (ML) to 332 333 estimate the relationships between variables and make predictions (Tibshirani, 1996, 2011; Durrant et al., 2021). This technique aims to find an equilibrium between model simplicity and accuracy by 334 introducing a penalty term into the traditional linear regression model, which enables sparse 335 solutions in which some coefficients are forced to be exactly zero. LASSO is especially useful for 336 variable selection because it can automatically identify only the most significant and discard 337 irrelevant or redundant variables, especially if we assume that many of the features do not 338 contribute significantly to the target variable (Durrant et al., 2021; Handorf et al., 2020). It also 339 helps to prevent overfitting by removing variables with low predictive value, potentially making the 340 model more robust across datasets. Furthermore, because it can choose between correlated 341 explanatory variables, it can aid in the optimization of models with high multicollinearity. In simple 342 words, the Lasso regression adds a penalty term to the MSE used in linear regressions. This penalty 343 344 term is proportional to the sum of the absolute values of the variable coefficients. The Lasso regression seeks the coefficient values that minimize the sum of the MSE and the penalty. 345 346 The Lasso regression cost function is defined as follows (Eq. 3):
- 347
- 348

$$J(\beta) = \left(\frac{1}{n}\right) * \sum (y_i - \hat{y}_i)^2 + l * \sum |\beta_j|$$

(3)

- 349
- 350 where

- $J(\beta)$  is the cost function 351 ٠ 352 • n is the number of data or physical samples (statistically, the sample size) y<sub>i</sub> is the actual output for the i-th sample 353 •  $\hat{y}_i$  is the predicted output for the i-th sample 354 • βj represents the coefficients (weights) associated with each feature 355 • l is the regularization parameter that controls the amount of regularization applied to the 356 ٠ model. Higher values of  $\lambda$  led to more regularization, resulting in a more pronounced feature 357 shrinkage and potentially some coefficients becoming exactly zero. 358 359 In this work, Lasso regression was applied in Python code using the scikit-learn module
- 360 (sklearn.linear\_model.Lasso).
- 361

### 362 **2.3 Machine learning and GRP mapping**

Machine learning (ML) algorithms allow to solve very complex problems. First, generating a model
based on processing the dataset and then, predicting the values of a new input data point by
executing the created model (supervised machine learning) (Rebala et al., 2019).

366 In the literature, recent works have applied ML techniques for spatial prediction in a number of

studies that deal with environmental science (e.g., landslide applications, Micheletti et al., 2014,

Tehrani et al., 2022; soil mapping, Hengl et al., 2017, GRP mapping, Petermann et al., 2021; time

369 series analysis, Janik et al., 2018). ML can handle complex multi-dimensional non-linear

relationships and mostly makes no or weak assumptions of the underlying distribution of the data

371 (Fouedijo and Klump, 2019). Furthermore, ML based approaches have been proven to outperform

classical geostatistical models for several prediction tasks dealing with highly complex systems

373 (e.g. Nussbaum et al., 2018; Hengl and MacMillan., 2019; Li et al., 2019). ML models display a

high performance due to their ability to reflect the influence and interplay of a multitude of factors.

Random Forest (RF) is an ensemble classifier algorithm developed by Breiman (2001) typically

used in classification and regression problems providing an output based on a Decision Trees

377 structures. Decision Tree is a regression model built using a series of decisions based on variable

values. Splitting values are determined to best separate subsets of data to take one path or the other.

Random Forest is a method of averaging many Decision Trees created from a bootstrap sample of

the full training set using a subset of predictors (=mtry) at each split in order to reduce overfitting

by a single Decision Tree. It uses bagging (i.e., bootstrap aggregation) to create numerous Decision

382 Trees by sampling a subset of training data with replacement and constructing the model based on

the sampled training set (Rebala et al., 2019).

- In this study, we have used Scikit learn code in python to apply a supervised machine learning
- method (i.e., Random Forest) to model the relationships between the SGRC (response variable) and
- the nine predictors described in the section 2.1.2 (<sup>220</sup>Rn, CO<sub>2</sub>, TGDR, permeability, fault density,
- 387 digital terrain model DTM, slope, aspect ratio and solar radiation).
- 388 389
- 390 2.4 Radon risk mapping

#### 391 **2.4.1 Risk concept**

- 392 The development of GRP maps is a valuable tool for hazard analysis; this map, coupled with
- 393 vulnerability and exposure factors, it is critical to assess the collective risk, i.e., the risk to which the 394 general public is exposed by geological causes.
- Furthermore, the map of the collective risk can be combined with the indoor measurements (thus
- including the knowledge of the geological base processes) to better delineate Radon Priority Areas,
- and manage the individual risk in terms of remediation activities.
- As above mentioned, we can define the risk as the product of hazard, vulnerability and exposure(Eq. 2).
- 400
- 401

### *Risk* = *Hazard* \* *Vulnerability* \* *Exposure*

(2)

- The application of the risk definition in order to mapping the CRA represents a first and easymethod to assess the collective Rn exposure in the study area.
- 404

## 405 2.4.2 Construction of CRA map

According to the risk equation, in order to construct the CRA map we identified the GRP as the hazard term, the location types and the total population of the census tracts of the study area

408 (available on the ISTAT web site, www.istat.it/it/archivio/104317#accordions) as vulnerability and
409 exposure factors, respectively.

410 The location type in the ISTAT dataset is marked by a number identifying the specific type of

building areas from 1 (residential areas) to 4 (sparse houses). These numbers were reclassified in

- 412 order to assigned the highest weight (4) to the area with the highest expected mean population
- 413 density, as follow: (i) location type 4 = residential areas; (ii) location type 3 = housing unit; (iii)
- location type 2 = industrial areas; (iv) location type 1 = sparse houses.
- Then, the total population and the location type have been used to calculate the population density
- 416 as the ratio between the total population living in a specific location type and the total area (in  $km^2$ )
- 417 of the census tract. The maps of the location type and the population density were converted in 50m

- 418 x 50m raster grid and normalised to the maximum value before constructing the final Rn risk map;
- the GRP map was also normalized to the maximum value.
- 420 Furthermore, these three factors (GRP, Location Type and Population Density) were multiplied
- 421 using the Raster Calculator tool in ArcGIS Pro according to Eq. 2. The resulting risk map has been

422 further standardized and the Zonal Statistic tool of Spatial Analyst in ArcGIS Pro was applied to

- 423 assign a risk value to each polygon of the census tract. We considered the maximum risk value
- 424 assigned to the polygon in order to visualize the risk map and to create the risk classes. The final
- risk map is divided into three risk classes expressed in percentage of risk (i.e., low, medium andhigh).
- 427

#### 428 **3. Results**

## 429 **3.1 Selected predictors, RF modelling and predictors importance**

Results of LASSO regression identified 7 predictors out of the 10 candidates: TGDR, CO<sub>2</sub>, FD,
<sup>220</sup>Rn, slope, aspect and soil permeability (see table S1 in supplementary materials). DTM, solar
radiation, and Rn in groundwater all show coefficients of 0, so they are excluded from the model
because they are considered as being misrepresentative. Furthermore, though slope and aspect show
not significant coefficients, they were however included in the RF model. The Variance Inflation

- Factor (VIF) was also calculated for the 7 selected predictors to evaluate multicollinearity, in order
- to be sure that among the 7 predictors there is no redundancy. All the selected predictors show VIF
- 437 < 7 (see table S2 in supplementary materials). The selected predictors include: one geophysical
- 438 parameter (TGDR), geochemical parameters (<sup>220</sup>Rn and CO<sub>2</sub>), geological parameters (Fault and
- Permeability), and geomorphological parameters (slope and aspect). All of these parameters are
  representative of the overall process at the core of Rn production (source), migration, and behaviour
- in shallow soil, as well as at the soil-atmosphere interface.
- Before to execute the RF model, the number of trees was set at 1000. The analysis of the model performance shows  $R^2$  of 0.93 and 0.47 for training and test data, respectively, and a RMSE of 0.30 and 0.83 for training and test data, respectively (see Fig. S4 in supplementary materials displaying the predicted vs observed values for training and test data).
- 446 The importance of the individual predictors in the RF model is considered as the relative influence
- of an individual predictor on the model performance (Fig. 2). The variable percentage importance
- shows that TGDR, CO<sub>2</sub>, fault density, <sup>220</sup>Rn, slope, aspect and permeability have the main influence
- in the model performance, respectively. In particular, TGDR, a proxy of the Rn source in rocks and
- soil, and the CO<sub>2</sub> (the main carrier gas in the study area, Benà et al., 2022) represent the most
- 451 influencing predictors with an importance higher than 30%. The fault density (FD) (i.e., proxy of

452 secondary permeability) highligths an important decrease in the percentage range of 10-15%. <sup>220</sup>Rn
453 and slope show an importance lower than 10% followed by the aspect ratio and soil permeability
454 lower than 5%, respectively.





456

Figure 2. Feature importance based on SHAP value percentage in the RF model. The predictors are ordered by decreasing importance.; X-axis: SHAP percentage; Y-axis = selected predictors. TGDR = terrestrial gamma dose rate;  $CO_2$  = carbon dioxide; FD = fault density; <sup>220</sup>Rn = Thoron; perm = soil permeability. 461

Furthermore, we constructed the SHAP diagram by using the "shap" library in Python to highlight 462 the impact of each selected predictors on the model prediction (Fig. 3). The Y-axis of the SHAP 463 diagram reports the 7 selected predictors in descending order of importance in the RF model from 464 TGDR (the most influent) to the soil permeability (the less influent). The X-axis of the SHAP 465 diagram represents the SHAP values quantifying the impact of a single feature on the model's 466 output: positive SHAP values indicate that the feature positively contributes to the output, while 467 negative values suggest a negative contribution. Red and blue dots represent the contribution of 468 individual features to the prediction compared to a reference value. Red dots represent positive 469 contributions and indicate that the feature is increasing the predicted output. Blue dots represent 470 negative contributions and indicate that the feature is decreasing the predicted output. In particular, 471 the SHAP diagram pointed out that positive values of TGDR, CO<sub>2</sub>, FD, <sup>220</sup>Rn, slope and 472 permeability exert the main influence in the model output; while, aspect is the only variable that has 473 influence in the model output for negative values. 474





Figure 3. SHAP diagram. Y-axis: reports the 7 selected predictors in descending order of 478 importance in the RF model; X-axis: the SHAP values quantifying the impact of a single feature on 479 the model's output: positive SHAP values indicate that the feature positively contributes to the 480 output, negative values suggest a negative contribution. Red and blue dots represent the contribution 481 of individual features to the prediction compared to a reference value. Red dots represent positive 482 contributions indicating that the feature is increasing the predicted output. Blue dots represent 483 negative contributions indicating that the feature is decreasing the predicted output. TGDR = 484 terrestrial gamma dose rate;  $CO_2$  = carbon dioxide; FD = fault density; <sup>220</sup>Rn = Thoron; perm = soil 485 permeability. 486 487

The next step in model interpretation is understanding the effect of an individual predictor on the model output. Partial dependent plots (PDPs) were constructed by using "pdpbox" library in Python to analyse the relationship between a target feature and the model's predicted outcome while considering all other features as fixed (see Fig. 5S a-g and the related explanation in supplementary materials). It helps to visualize the relationship between a target feature and the model's predicted outcome. The PDP of each predictor is calculated by accounting for the average effect of the other predictors in the model (Petermann et al., 2021).

495

## 496 **3.2 GRP map**

497 Random forest algorithm has been applied to construct the GRP map of the study area by using

498 SGRC as response variable and the 7 selected predictors (i.e., TGDR, CO<sub>2</sub>, fault density, <sup>220</sup>Rn,

slope, aspect, permeability). The final predicted GRP map ranges between a minimum value of 7.21

 $kBq \cdot m^{-3}$  and a maximum value of  $182 kBq \cdot m^{-3}$  (Fig. 4). According to the results reported in Benà et

al. (2022), we consider high GRP values those exceeding 50 kBqm<sup>-3</sup>, i.e., the local background.

502 Higher GRP values extend along the E-W direction from Falzes to Chienes (central part), to Terento

- 503 municipalities, accordingly to the direction of the wide fracture zone belonging to the Pusteria fault
- system. High GRP values are linked to the Tectonically Enhanced Radon (TER) quantity (Benà et
- 505 al., 2022).
- 506



- 508 Figure 4. Geogenic Radon Potential (kBqm<sup>-3</sup>) map of the study area.
- 509

507

# 510 **3.3 The CRA map**

Figure 5 shows the CRA map of the study area, representing the density of collective risk and 511 obtained by multiplying the GRP map, the location type (vulnerability) and the population density 512 (exposure factor). The map was divided into three risk classes using the natural breaks method as 513 follow: i) risk < 5%, low risk (in white); ii) 5%<risk<50%, medium risk (in orange); iii) risk >50% 514 high risk (in red). The CRA map is linked to table 1 which summarizes some parameters 515 characterizing the three defined risk classes: (i) the average GRP value in kBqm<sup>-3</sup>; (ii) the average 516 population density expressed in number of people per  $\text{km}^2$ ; (iii) the location type (i.e., 4, 3, 2, 1); 517 (iv) the total area covered by the considered risk class. 518

519



Figure 5. Map of the Collective Risk Areas. 521

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Collective risk class	Risk level (%)	GRP mean (kBqm <sup>-3</sup> )	Population density (people km <sup>-2</sup> )	Population (people)	Location type	Area (km <sup>2</sup> )
Low	< 5	63.50	546	5927	4, 3, 2, 1	68.51
Medium	5 - 50	65.11	6116	3072	4, 3, 2, 1	0.75
High	> 50	75.88	17549	622	4	0.05

<sup>523</sup> 

Table 1. The table reports the risk class and the correspondent percentage of risk, the mean GRP value, the population density, the location type and the extension of the area covered by the 524 considered risk class. 525 526

Most of the study area (68.51 km<sup>2</sup>) falls within low risk areas; this agrees with the mountainous 527 morphology of the territory where most of the population is concentrated in the residential areas of 528 the main municipalities (Terento, Chienes and Falzes). In general, the mean GRP values (hazard) 529 exceed the local background value of 50 kBqm<sup>-3</sup> in all the three risk classes and slightly increases 530 from low risk (63.50 kBqm<sup>-3</sup>) to high risk (75.88 kBqm<sup>-3</sup>). The progressively increase of the mean 531 population density (e.g., exposure) from low to high risk areas are strictly related to the location 532 type (e.g., vulnerability): (i) in the low risk areas most of the census tracts (33) are described as 533 residential areas (location type = 4) and sparse houses (location type = 1, 43 census tracts); (ii) in 534 the medium risk areas most of the census tracts are considered as residential areas (location type 4, 535 22 census tracts); (iii) all census tracts falling in the high risk areas are described as residential areas 536

(location type = 4) with the highest population density. In fact, the population density increases
accordingly from low to high risk areas.

539

#### 540 **4. Discussion**

# 541 **4.1 Interpretation of predictors in the RF model**

The RF model demonstrates that all of the selected predictors influence Rn concentrations and 542 movement in the subsoil. This result is consistent with the dependence of Rn from the geochemical 543 and structural characteristics of the study area mainly linked to the generation and transport of Rn in 544 the geological environment (i.e., from deep source toward the subsoil) (Benà et al., 2022). In fact, it 545 is not surprising that the variable's importance shows clearly that GRP is primarily affected by TGDR 546 (35%, Fig. 2) which represents the BRS contribution (e.g., the radionuclide content <sup>238</sup>U and <sup>232</sup>Th) 547 of the main outcropping rocks (i.e., gneiss, granite, phyllite) (Tchorz-Trzeciakiewicz et al., 2021; 548 549 Giustini et al., 2019, 2022). Because the survey of ambient gamma dose rate was conducted at the ground level, the correlation of TGDR with soil gas radon concentrations is likely to be stronger than 550 551 with atmospheric concentrations. In the literature, Bossew et al., 2017; Cinelli et al., 2019; Melintescu et al., 2018; Sainz Fernández et al., 2017 reported a positive correlation between TGDR and GRP. 552 553 The BRS contribution to the Rn amount in soil gas generates a relatively high spatial variability of Rn concentration in the soil gas, reflecting the homogeneous characteristics of the soil/rock 554 environment at local scale (BRS). However, Rn spatial variability can increase (also at local scale) 555 near fault zones (TER), especially in seismic areas characterised by active faults. In these areas, Rn 556 migration from deeper sources can be increased by intense fracturing and the presence of carrier 557 gases (mainly CO<sub>2</sub>) that may play a dominant role for advective transport and redistribution of trace 558 gases at surface (Wilkening, 1980; Ciotoli et al., 2007, 2014; Prasetio et al., 2023, and reference 559 therein). This is observable in the study area along the Pusteria fault system, where radon 560 concentrations in soil gas have a positive correlation with CO<sub>2</sub> concentrations (importance of about 561 30%, Fig. 2), suggesting a possible advective up flow caused by pressure gradients. In this faulted 562 area, radon anomalies at surface could also be associated with elevated concentrations of 563 564 radionuclide concentrations (i.e., Ra and U) in small soil particles transported by CO<sub>2</sub> gas molecules (Etiope & Lombardi, 1995). Furthermore, the presence of dissolved CO<sub>2</sub> in groundwater may 565 566 promote radium dissolution and thus transport in solution (Giraults et al., 2014). The high importance (about 15%, Fig. 2) of the fault density (interpreted as fault secondary 567 permeability) confirms the effect of the Pusteria fault system on the Rn migration (as well as of 568 other gases); this predictor is strictly related to the TER component (Benà et al., 2022). Indeed, 569 570 damage zones related to high fracturing zones (fault areas) often exhibit a high permeability

- 571 compared to the surrounding rocks and may facilitate the fluids advective transport for SGRC, thus
- 572 potentially increasing radon release towards the surface and, as a consequence, Rn availability to
- 573 enter buildings (IRC) (Ciotoli et al., 2007, 2014, 2016; Seminsky et al., 2014; Chen et al., 2018;

574 Banrion et al., 2022; Zhou et al., 2023).

- 575 Similar importance of the other predictors (i.e., Tn, Slope, Aspect and Permeability) ranging from 4
- to 8% can be explained by shallower processes affecting Rn movement in the soil layer, and at the
- soil-atmosphere interface (SRE) (Fig. 2). In the shallow environment the influence of
- 578 meteorological conditions can be a complex issue, and the literature results are controversial. Air
- temperature and pressure on soil radon concentrations is small in comparing with total seasonal
- variability of this gas, and in any case the influence of these two variables is further lowered by
- conducting soil gas measurement campaigns during periods of stable and good weather conditions
- 582 (Ciotoli et al., 2014; Beaubien et al., 2013, 2008).
- 583 The principal drivers governing diurnal and seasonal changes of radon concentration in the soil are
- the water-saturation and moisture-retention in the soil pore (i.e., rainfall) (King and Minissale,
- 585 1994). These two parameters directly decrease soil permeability thus preventing radon gas diffusion
- 586 in the shallow soil layers (Nazaroff, 1992; Alonso et al., 2019; Beltran-Torres, 2023). High soil
- 587 permeability allows <sup>220</sup>Rn to be detected at surface despite its short decay time (56 seconds).
- 588 In addition, the slope can be used as a proxy of soil moisture and meteorological conditions in
- absence of any other meteorological variables. High slopes also constitute zones characterized by
- 590 increased soil permeability because they do not promote the retention of water and moisture in the
- soil pores. On the contrary, flat zones are characterized by low soil permeability because they
- favour the accumulation of water and moisture in the soil pore. At this regard, the SHAP diagram
- shows that high values of Tn, slope and permeability are positively correlated with high GRP (Fig.
- 5943). The soil permeability may be linked to the ability of radon to migrate and escape towards the
- Earth surface. In fact, where permeability is high radon escapes more easily. Permeability is alsolinked to the fault density representing the secondary permeability.
- All these predictors, except for the aspect, have an impact on the GRP values prediction for positive values and show an increasing trend up to the expected average radon value (see PDPs, in Fig. S5 in supplementary materials). On the contrary, low values of the GRP are correlated with high values of the aspect ratio (i.e., inverse correlation). The aspect identifies the compass direction that the downhill slope faces for each location; therefore, radon accumulation is easier in flat areas.
- 602 The model confirmed the correlations between geology and GRP and also provided insight into the
- 603 utility and significance of other predictors that reflect the physical, chemical, and hydraulic
- properties of soil, as well as climatic predictors. On the basis of these results, further work should

also consider meteorological parameters, such as soil temperature and humidity, rainfall, etc. This is
 especially fundamental to capture seasonal variability in models that uses IRC as response variable.

#### 608 4.2 Map of the Collective Risk Areas (CRAs)

609 The GRP map obtained by RF regression represents radon hazard due to geological features of a

specific region. It is strictly related to Rn gas directly measured in the soil and to all geological

611 predictors (e.g., TGDR, CO<sub>2</sub>, fault density, etc.) that significantly influence its concentration in the

shallow environment, and potentially affect its movement towards homes. GRP maps, representing

- the most significant spatial predictor of IRC, are useful tools to evaluate the Rn risk (Bossew, 2015;Bossew et al., 2020).
- As already mentioned, the European regulations aims to identify RPAs and implement mitigation
- plans in order to limiting radon exposure and thus reducing the risk of lung cancer to population. In
- an unbuilt and inhabited area, the presence of high Rn values represents only a high hazard (i.e.,
- 618 GRP), but not a risk. This concept is highly known and applied in the case of other natural
- 619 phenomena such as in seismic microzonation studies. European legislation aims to reduce the
- 620 detriment from Rn exposure (i.e., the number of lung cancer deaths) and as a consequence, reduce
- 621 the collective exposure. In Figure 6, we show how GRP is a key factor in recognising of collective
- 622 risk areas (CRAs).
- 623 In this paper, for the first time, we introduce the concept and define the Collective Risk Areas
- 624 (CRAs) by applying the risk definition (section 2.4) consisting of three basic factors: i) the hazard,
- e.g., the Geogenic Radon Potential (GRP), ii) the vulnerability, e.g., the type of location, and iii) the
- 626 exposure, e.g., the population.
- 627



629 Figure 6. Summary sketch of the Collective Risk Areas concept.

628

- Mapping the GRP is clearly the first fundamental step in defining the Rn hazard, a characteristic
- which cannot be mitigated. For this reason, it is important to map it as accurately as possible (i.e.,
- by consider multiple geological variables and applying robust mapping techniques).
- As reported in Benà et al., 2022, Rn values exceeding the lithological background (50 kBq  $m^{-3}$ ) are
- 635 considered anomalous and linked to the wide fracturing zone of the Pusteria fault system that
- represents Rn enhanced by tectonics (TER). However, in Benà et al.,2022, this quantity is not
- 637 discussed in terms of GRP and thus it does not include the other important geological factors, such
- as gas permeability and deep circulation indicators (e.g., Rn in groundwater), as well as the shallow
  effects governed by the morphological parameters (e.g., DTM).
- 640 The identification of a threshold value of GRP is not significant to delineate CRAs, since the indoor
- radon risk exists even for "very low" concentrations of radon in the soil and, consequently, for very
- low GRP values. In fact, radon measured in the soil (GRP) is generally three order of magnitude
- 643 higher than indoor radon. It is clear that every area can be affected by a potential indoor risk and all
- the dwellings are considered vulnerable.
- However, GRP plays a key role in defining the CRAs that mainly occur along the Pusteria fault
- 646 system where Rn degassing is enhanced by the intense fracturing and the GRP values are high. This
- 647 is consistent with the fact that all the GRP values contribute to the risk. Therefore, the CRAs map
- 648 highlights those areas with low, medium and high collective risk and, as a consequence, here the
- 649 IRC values may be high for the residential areas.
- 650

## 651 5 Conclusions

- The mapping and analysis of GRP (e.g., Rn hazard), obtained by using ML approach, is a
- fundamental tool for the delineation of CRAs according to a new, more geological, interpretation of
- the RPAs with respect of that reported in the BSS directive (2013/59/EURATOM).
- We used the risk formula to combine the GRP map with the location type characteristic of the
- census tracts (e.g., the vulnerability factor) and the population density (e.g., the exposure factor).
- According to a geological-based interpretation of the RPAs, we can recognise hazard-based RPAs
- 658 (CRAs) and detriment-based RPAs (IRAs) as complementary concepts of territorial planning and
- remediation actions, respectively, and not in alternative.
- 660 In particular, the obtained results highlight the following conclusions:
- Machine learning model by using the random forest technique demonstrates as a robust and high-performance method to obtain a GRP map of the study area. In particular, the obtained GRP map uses seven predictors reflecting geology (BRS and TER), soil characteristics
   (groundwater circulation, permeability), and meteorological conditions (DTM derivatives).

- The variable importance highlights the dominant impact of Rn source but still significant 665 contributions of the other predictors. 666
- As GRP is considered the most important spatial predictor of IRC, it is clear that mapping 667 • this hazard factor well represents the total amount of radon that can potentially enter 668 buildings. 669
- Since GRP (e.g., soil gas concentration) values are three order of magnitude higher than the 670 • IRC, there is no reason to define GRP threshold, as the indoor radon risk can exists even for 671 "very low" concentrations of radon in the soil and, consequently, for very low GRP values. 672 GRP qualitative classes can serve only as delineation of zones (in the same way used in the 673 seismic micro zonation studies) in which different land use planning strategies and/or 674 construction types, and remediation actions should be adopted 675
- The absence of an unambiguous guidelines to define Radon Priority Areas (RPAs) led to the 676 • geological-based conceptualization of a complementary approach of mapping both the 677
- CRAs (in terms of prevention), as well as IRAs (in terms of building remediation actions). 678 This study may help policy makers to implement constructive preventive measures in those areas 679 where new buildings are planned, and to act in terms of remediation in the RPAs sensu stricto. 680 Future studies may aim to define the effective individual risk by constructing statistical models that 681 682 also consider IRC measurements and anthropogenic factors.
- 683

#### **Authors contribution** 684

Eleonora Benà: Conceptualisation, data curation, methodology, writing - original draft preparation, 685 reviewing and editing. Giancarlo Ciotoli: conceptualisation, methodology, writing - reviewing and 686 editing. Eric Petermann: methodology, software, reviewing and editing. Peter Bossew: 687 conceptualisation, reviewing and editing. Livio Ruggiero: reviewing and editing. Luca Verdi: 688 reviewing and editing. Paul Huber: water springs sampling. Federico Mori: software, Python 689 code. Claudio Mazzoli: conceptualisation, reviewing and editing. Raffaele Sassi:

- 690
- conceptualisation, project administration, supervision, funding acquisition, reviewing and editing. 691
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