

Calculating flux to predict future cave radon concentrations

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Original citation & hyperlink:

Rowberry, MD, Marti, X, Frontera, C, Van De Wiel, MJ & Briestenský, M 2016, 'Calculating flux to predict future cave radon concentrations' Journal of Environmental Radioactivity, vol 157, pp. 16-26

http://dx.doi.org/10.1016/j.jenvrad.2016.02.023

DOI 10.1016/j.jenvrad.2016.02.023 ISSN 0265-931X ESSN 1879-1700

Publisher: Elsevier

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Elsevier Editorial System(tm) for Journal of

Environmental Radioactivity

Manuscript Draft

Manuscript Number: JENVRAD-D-15-00654R1

Title: Calculating flux to predict future cave radon concentrations

Article Type: Research Paper

Keywords: cave radon concentration; radon flux; cave ventilation; radioactive decay; fault slip; numerical modelling.

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Abstract: Cave radon concentration measurements reflect the outcome of a perpetual competition which pitches flux against ventilation and radioactive decay. The mass balance equations used to model changes in radon concentration through time routinely treat flux as a constant. This mathematical simplification is acceptable as a first order approximation despite the fact that it sidesteps an intrinsic geological problem: the majority of radon entering a cavity is exhaled as a result of advection along crustal discontinuities whose motions are inhomogeneous in both time and space. In this paper the dynamic nature of flux is investigated and the results are used to predict cave radon concentration for successive iterations. The first part of our numerical modelling procedure focuses on calculating cave air flow velocity while the second part isolates flux in a mass balance equation to simulate real time dependence among the variables. It is then possible to use this information to deliver an expression for computing cave radon concentration for successive iterations. The dynamic variables in the numerical model are represented by the outer temperature, the inner temperature, and the radon concentration while the static variables are represented by the radioactive decay constant and a range of parameters related to geometry of the cavity. Input data were recorded at Driny Cave in the Little Carpathians Mountains of western Slovakia. Here the cave passages have developed along splays of the NE-SW striking Smolenice Fault and a series of transverse faults striking NW-SE. Independent experimental observations of fault slip are provided by three permanently installed mechanical extensometers. Our numerical modelling has revealed four important flux anomalies between January 2010 and August 2011. Each of these flux anomalies was preceded by conspicuous fault slip anomalies. The mathematical procedure outlined in this paper will help to improve our understanding of radon migration along crustal discontinuities and its subsequent exhalation into the atmosphere. Furthermore, as it is possible to supply the model with continuous data, future research will focus on establishing a series of underground monitoring sites with the aim of generating the first real time global radon flux maps.

Calculating flux to predict future cave radon concentrations

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Keywords cave radon concentration; cave radon flux; cave ventilation; radioactive decay; fault slip; numerical modelling.

Highlights

- Cave radon concentrations pitch flux against ventilation and radioactive decay
- Exhalation is dominated by radon advection along discontinuities such as faults
- Our numerical model isolates flux and then predicts future radon concentrations
- Independent observations confirm the close relationship between flux and fault slip

1 1. INTRODUCTION

Radon (²²²Rn) is a radioactive noble gas that results from the decay of solid radium 2 (²²⁶Ra). The release of radon is controlled by the alpha particle recoil mechanisms that 3 expel radon from radium. Whether a newly formed radon atom remains in the mineral 4 grain or whether it enters the intergranular pore space is determined by the position of 5 the radium atoms and the direction of radon atom recoil (Appleton 2013). The vast 6 majority of radon atoms remain within the mineral grain only to decay once again into a 7 solid product while the tiny minority that enter the intergranular pore space then begin 8 9 the process of migration towards the surface. Migration is controlled largely by the water retention and fluid transmission characteristics of the bedrock (Åkerblom & 10 Mellander 1997). The latter include its permeability, its porosity, and its pore size 11 12 distribution as well as the nature of any crustal discontinuities such as faults, fractures, and joints (Appleton 2013). It is far more common for radon to be emitted into a liquid 13 phase rather than into a gas phase. Radon migration in the liquid phase occurs with the 14 15 help of carrier fluids and the radon will remain in the liquid phase until a gas phase is introduced. Clearly radon migration in the liquid phase is going to be influenced by 16 factors such as groundwater circulation whereas migration in the gas phase is going to 17 be influenced by factors such as the diffusion characteristics of the gas. 18

19

Radon is generally abundant in confined underground spaces such as caves, tunnels, 20 and mines (Stannard 1988). The numerical model presented in this paper is based on 21 input data recorded in a cave. Measurements of cave radon concentration reflect the 22 outcome of a perpetual competition which pitches flux against ventilation and 23 radioactive decay (Wilkening & Watkins 1976). In the absence of ventilation it is 24 possible for the radon concentration in such settings to approach that characteristic of 25 soil gas (Wilkening 1990). Cave radon clearly accumulates as a result of exhalation from 26 27 the confining rock mass but it is important to have a basic understanding of the contributions made by diffusive transport and advective transport. The distances over 28 which radon atoms can be transported by diffusion are limited by the short half life of 29 radon ($t_{1/2} = 3.82$ d) while the distances over which they can be transported by 30 advection along structural discontinuities is significantly further, perhaps more than 31 one hundred metres (Appleton 2013). Exhalation by diffusion from solid limestone 32 containing 2.2 mg kg⁻¹U may be expected to result in cave radon concentration 33

measurements in the order of 100 Bq/m³ (Appleton 2013). The fact that radon
concentration measurements in such settings are generally greater by at least one order
of magnitude emphasises the importance of exhalation by advection along structural
discontinuities. This importance may be heightened in caves compared to other
underground settings as their passages often develop along precisely the same faults
and fractures as those used for radon migration.

40

Faults and fractures permit the efficient transmission of radon to the surface due to the 41 fact that fluids readily migrate along such crustal discontinuities. Consequently many 42 studies have used high radon concentration measurements to infer the presence of 43 discontinuities under soil or glacial drift. High radon concentration is more likely to be 44 45 encountered if the discontinuities are active (Swakoń et al. 2005; Ielsch et al. 2010; Neri et al. 2011). Discontinuities in the near surface environment may be thought of as active 46 if they are subjected to thermal expansion as this leads to dilation and constriction 47 whereas those at greater depths tend to be more susceptible to slip caused by either 48 gravitational or tectonic processes. The EU-TecNet fault displacement monitoring 49 network has been making direct experimental observations of fault slip at more than 50 51 one hundred sites across central Europe. More than a decade of data demonstrate that fault motion in this intracratonic region is commonly characterised by steady 52 progressive creep trends: these may be horizontal (strike-slip), vertical (dip-slip), or a 53 combination of the two (oblique-slip). However, the steady progressive creep trends 54 are sometimes interrupted by short periods of anomalous activity, interpreted to reflect 55 a short term perturbation in the regional stress field (Stemberk et al. 2010; Košťák et al. 56 2011; Briestenský et al. 2015). During these periods the progressive creep trends may 57 be subjected to, for example, a conspicuous reversal; a sudden enduring displacement; 58 or a series of oscillatory displacements. It follows that significant displacements should 59 also be evidenced by radon anomalies especially given that numerous studies have 60 related radon concentration anomalies with other geodynamic phenomena such as 61 62 earthquakes (Igarashi et al. 1995; Briestenský et al. 2014; Hwa Oh & Kim 2015). 63

Once radon has been exhaled into a confined underground space it is then subject to the
processes responsible for liberating it into the atmosphere. The most comprehensive
recent account of underground meteorology is that of Badino (2010). Air exchange is

strongly influenced by convective circulation caused as a result of internal-external 67 buoyancy pressure differences and barometric circulation caused as a result of internal-68 external pressures differences. The former is particularly important for caves with more 69 than one entrance at different heights whereas the latter is more important for caves 70 with only one entrance or for caves with only extremely small entrances. Diurnal and 71 seasonal circulation changes often result in diurnal and seasonal fluctuations in natural 72 gas and aerosol concentrations (Bezek et al. 2012). The effects of such changes on radon 73 concentration are particularly well known because radon is commonly used as a tracer 74 for cave ventilation modelling (Cunningham & Larock 1991; Hakl et al. 1997; Tanahara 75 et al. 1997; Przylibski 1999; Perrier et al. 2004; Kowalczk & Froelich 2010; Gregorič et 76 al. 2014). However, although the processes governing air exchange are well understood, 77 78 the mass balance equations used to model changes in radon concentration through time 79 routinely treat flux as a constant. This mathematical simplification sidesteps an intrinsic geological problem: the majority of radon exhalation occurs as a result of advection 80 from crustal discontinuities whose motions are clearly inhomogeneous in both space 81 and time. In this paper the dynamic nature of flux is investigated and the results are 82 used to predict cave radon concentration for successive iterations. 83

84

85 **2. STUDY AREA**

The input data used for our numerical model were recorded at Driny Cave in the 86 geodynamically active Little Carpathian Mountains of western Slovakia (Figure 1). This 87 mountain range trends SW-NE along the southeastern margin of the Bohemian Massif 88 89 and forms part of the Alpine-Carpathian Orogenic Belt (Lenhardt et al. 2007). It comprises a mesh of clearly defined morphostructural units (Marko et al. 1991) 90 bordered to the northwest by the Vienna Basin and to the southeast by the Pannonian 91 Basin (Plašienka et al. 1997). The range is characterised by moderate seismicity: the 92 strongest earthquake, with a magnitude of $M_s = 5.7$, occurred in the epicentral area of 93 Dobrá Voda on 9 January 1906 and was followed by a large aftershock, with a 94 magnitude of $M_s = 5.3$, on 16 January 1906 (Zsíros 2005). Other events with 95 magnitudes of greater than $M_L = 4.0$ occurred in 1904, 1930, and 1967 (Fojtíková et al. 96 97 2010). Notable recent earthquakes were recorded close to the town of Vrbové, with a magnitude of M_L = 3.3, on 13 March 2006, and close to the town of Studienka, with a 98 magnitude of M_L = 3.4, on 5 March 2012. The stress field appears to be dominated by 99

NE-SW horizontal extension (Kováč et al. 2002) although NE-SW horizontal
compression has been identified around the epicentral area of Dobrá Voda (Fojtíková et
al. 2010).

103

Driny Cave is located at the contact between the Little Carpathian Mountains and the 104 adjacent Pannonian Basin. The area around the cave hosts a range of slope 105 deformations while the limestone cliffs which host the entrance to the cave are 106 characterised by numerous open fissures (Briestenský et al. 2011a). The cave itself has 107 developed in the lower Cretaceous locally schistose marly limestones of the Hlboč 108 Formation (Michalík et al. 1992) as a result of corrosion by meteoric waters seeping 109 along splays of the NE-SW striking Smolenice Fault and a series of transverse faults 110 111 striking NW-SE (Droppa 1951). Its main entrance is situated at 399 m asl, its smaller upper entrance is situated at 431 m asl, its chimney descends a total of 36 m, and its 112 passages attain a total length of 680 m (Bella 2006). Faults within the cave often exhibit 113 striations and slickencrysts. These features indicate that the splays of the NE-SW 114 striking Smolenice Fault are primarily affected by dip slip displacements while the 115 transverse faults striking NW-SE are primarily affected by strike slip displacements 116 117 (Briestenský et al. 2011a). Its walls are commonly covered by flowstones while the ceilings and floors host numerous types of dripstone. The presence of tiny, fresh, cracks 118 in dripstones developing close to fault outcrops suggests that these crustal 119 discontinuities are still active (Briestenský et al. 2011a,b). 120

121

122 Three mechanical extensometers have been installed to measure fault slip in the cave. The monitoring points Driny 1 and Driny 2 are installed across parallel splays of the 123 Smolenice Fault whereas Driny 3 is installed across one of the transverse faults 124 (Briestenský et al. 2011a,b). Results from Driny 1 and Driny 2 present evidence of 125 sinistral strike slip and uplift of the northwestern block whereas results from Driny 3 126 present evidence of dextral strike slip and uplift of the northeastern block (Table 1). In 127 128 detail, however, the data are more complicated. At Driny 1, large reversals in the sense of dip slip displacement were recorded towards the end of 2009 and at the beginning of 129 2010 while dextral strike slip dominated between 2010 and 2012. At Driny 2, large 130 reversals in the sense of dip slip displacement were again recorded towards the end of 131 2009 and at the beginning of 2010 while dextral strike slip dominated slightly later, 132

between 2012 and 2013. At Driny 3, subsidence of the northeastern block was recorded
between 2011 and 2013 while sinistral strike slip dominated between 2010 and 2012.
The anomalous fault slip trends recorded between 2010 and 2013 are interpreted to
reflect a short term perturbation in the regional stress field (see, for example, Stemberk
et al. 2010; Košťák et al. 2011; Briestenský et al. 2015).

138

139 **3. EXPERIMENTAL METHODS**

Measurements of equilibrium equivalent radon concentration, ambient temperature, 140 and barometric pressure were obtained every thirty minutes from January 2010 to 141 August 2011 while measurements of fault slip have been recorded at Driny Cave once 142 every two weeks since January 2006. The measurements of equilibrium equivalent 143 144 radon concentration were obtained by a radon progeny monitor TS96¹. This was installed in a distal part of the cave in which the host limestone is characterised by low 145 calculated mass activity (Štelcl et al. 2002). These instruments use a 200 mm² 146 semiconductor barrier detector to measure the alpha activity of ²¹⁸Po and ²¹⁴Po with an 147 accuracy of +/-10 %, a resolution of 2 Bq/m³, and a measuring range between 0 Bq/m³ 148 and 1999 Bq/m^3 . EEC_{Rn} measurements were converted to radon concentration using a 149 150 constant radioactive equilibrium coefficient of F = 0.5. It should be noted that the validity of applying a constant coefficient is discussed in Section 6.3. The measurements 151 of ambient temperature were obtained using two Comet System instruments R0110. 152 One was installed outside in a cleft above the lower entrance to the cave and one was 153 installed at the distal end of the cave, approximately five metres from the radon progeny 154 155 monitor. These instruments have an accuracy of ± 0.4 °C, a resolution of 0.1 °C, and a measuring range between -40° C and $+80^{\circ}$ C. The measurements of barometric pressure 156 were recorded by an Eijkelkamp BaroDiver. This was installed in a hydrogeological 157 borehole in the nearby epicentral area of Dobrá Voda. These instruments have a 158 resolution of \pm 0.1 cm H₂O, a typical accuracy of \pm 0.5 cm H₂O, and can withstand a 159 maximum pressure of 15 m H₂O. The readings are converted to give atmospheric 160 161 pressure in hPa. The measurements of fault slip are recorded using three mechanical extensometers TM-71 (Košťák 1991). Two are installed across splays of the Smolenice 162 Fault (dip→dip direction: $70^{\circ} \rightarrow 290^{\circ}$ and $70^{\circ} \rightarrow 110^{\circ}$) while the third is installed across 163

¹ The TS96 was developed at the Institute of Particle & Nuclear Physics at Charles University in Prague and approved for equilibrium equivalent radon concentration monitoring by the Czech Metrology Institute.

- one of the transverse faults ($75^{\circ} \rightarrow 040^{\circ}$). These instruments use the moiré phenomenon of optical interference to record displacements in a three dimensional Cartesian coordinate system with a resolution of ± 0.007 mm (Martí et al. 2013).
- 167

168 4. NUMERICAL METHODS

169 **4.1** Ventilation mechanisms, temperature gradients, and air flow velocities

The presented numerical model assumes a system whose geometry is based on that of 170 Driny Cave (Figure 2A). The first step in the first stage of the modelling procedure 171 specifies the ventilation mechanism. In our model, as at Driny Cave, the air flow 172 direction reflects the presence of two entrances at discordant heights. The specific 173 mechanism is governed by Bernoulli's Principle. When the inner temperature is greater 174 175 than the outer temperature, the comparatively warm, less dense, air inside the cave is forced out of the upper entrance as a result of the chimney effect. When the outer 176 temperature is greater than the inner temperature, the comparatively cold, denser, air 177 inside the cave is forced out of the lower entrance as a result of Torricelli's Law. 178 179

180 The second step defines an intermediate parameter, T^* , to account for the difference 181 between the temperature inside the cave, T_i , and the temperature outside the cave, T_o , 182 as illustrated in Eq. 1:

183

$$T^* = \sqrt{\frac{|\Delta T|}{T_j}} \tag{1}$$

184

185 Where: ΔT is the observed difference between the outer temperature and the inner 186 temperature ($\Delta T = T_o - T_i$) and T_j is the temperature corresponding to each ventilation 187 mechanism ($T_j = T_i$ for chimney effect and $T_j = T_o$ for Torricelli's Law). This parameter 188 is needed at various points in the numerical modelling procedure and it allows us to 189 present more concise mathematical formulas in subsequent steps. 190 191 The third step calculates air flow velocity through the system on the basis of Bernoulli's

192 Principle, irrespective of the specific ventilation mechanism, as illustrated in Eq. 2:

$$v = T^* \sqrt{2gH} \tag{2}$$

194

195 Where: *g* is gravity acceleration and *H* is the height difference between the uppermost 196 and lowermost points of the cave. The modelled flow velocities for air passing through 197 the upper entrance, if $T_i > T_o$, or the lower entrance, if $T_o > T_i$, are presented in Figure 198 2B while an extracted air flow velocity profile is presented in Figure 2C. These 199 demonstrate, first, that air flow ceases when there is no temperature gradient and, 200 second, that the air flow velocity is not a linear function of the temperature gradient. 201

202

4.2 Modelling radon flux and predicting radon concentration

The first step in the second stage considers the seminal mass balance differential
equation of Wilkening & Watkins (1976), routinely used to model cave radon
concentration changes, as illustrated in Eq. 3:

$$\frac{\mathrm{d}c}{\mathrm{d}t} = \phi - \lambda \cdot c - \frac{Q}{V}\Delta c - \frac{D}{L}\Delta c \tag{3}$$

207

Where: ϕ is radon flux; *c* is radon concentration; λ is the radioactive decay constant; *Q* is 208 the displaced volume of air; *V* is the total volume of air in the cave; *D* is the radon 209 diffusion coefficient between two points separated a length L in the absence of 210 ventilation; Δc is the difference between radon concentration measured inside and 211 outside the cave. This term can be simplified by assuming that the radon concentration 212 outside the cave is zero (i.e. $\Delta c = c$) and by neglecting the diffusion term because, in the 213 absence of ventilation, radon is only transported for short distances compared to the 214 dimensions of a typical cave. 215

216

The second step consolidates the variables related to cave geometry into one symbol,
allowing us to present more concise mathematical formulas in subsequent steps, as
illustrated in Eq. 4:

220

$$\gamma \equiv \frac{k_j}{l_j} \sqrt{2gH}.$$
(4)

Where: *k_j* is a coefficient from 0 to 1 that accounts for the proportion of the ceiling or entrance which acts as an aperture for air flow and *l_j* is a length scale equal to the height of the cave or the magnitude of the length of the cave. The proportion of the ceiling and the height of the cave are used when ventilation is dominated by the chimney effect whereas the proportion of the entrance and the magnitude of the length of the cave are used when ventilation is dominated by Torricelli's Law. The influence that this variable exerts on the outputs of the numerical model is discussed in Section 6.1.

The third step isolates flux in the mass balance equation in order to simulate real timedependence among the variables, as illustrated in Equation 5:

232

$$\phi(t) = \lambda \cdot c + \gamma T^* \cdot c + \frac{\mathrm{d}c}{\mathrm{d}t}$$
(5)

233

This equation connects the perpetual competition pitching flux, the ultimate source of the radon inside the cave, against ventilation. It demonstrates that the retrospective analysis of flux requires only routinely available data: radon concentration, inner temperature, and outer temperature.

238

The fourth step delivers an expression to compute radon concentration for successive
time instants. It is possible to predict radon concentration, *c*, for successive time
instants, *t*, using the preceding radon concentration, *c*₀, and the ambient temperatures,
as illustrated in Equation 6:

243

$$c \approx c_0 + t \cdot \left(\phi - c_0(\lambda + \gamma T^*)\right) \tag{6}$$

244

As in the mass balance differential equation of Wilkening & Watkins (1976), flux is
thought of as favourable, the term preceded by a plus sign, while ventilation and
radioactive decay are thought of as unfavourable, the terms preceded by a minus sign.
The relevance of this equation is illustrated by a simulation presented in the following
section while a flow diagram summarising the outlined numerical modelling procedure
is presented in Figure 3.

252 **5. MODEL IMPLEMENTATION**

An illustrative numerical simulation is presented in Figure 4 to demonstrate the 253 relevance of Equation 6. Figure 4A outlines a hypothetical ventilation scenario, Figure 254 4B outlines a hypothetical flux scenario, and Figure 4C presents iterative estimations of 255 future radon concentration. The first half of the numerical simulation is characterised 256 by an increase and decrease in the air flow velocity while the second half is 257 characterised by a complete absence of ventilation. Flux remains constant throughout 258 this period except at those times when it is interrupted by a spontaneous anomaly. 259 During the first half of the simulation the radon concentration decreases rapidly as 260 ventilation successfully counteracts flux. Furthermore, the first pair of flux anomalies 261 clearly influence the radon concentration. During the second half of the simulation the 262 263 radon concentration increases steadily as ventilation is no longer able to counteract flux. However, the second pair of flux anomalies do not clearly influence the radon 264 concentration. This simulation suggests that radon concentration is not necessarily a 265 reliable indicator of radon flux. 266

267

Figure 5A-C presents time series of inner temperature, T_{i} outer temperature, T_{o} , and 268 radon concentration, c, recorded at Driny Cave from January 2010 to August 2011. It is, 269 unfortunately, not possible for us to extend this term as it represents the only period in 270 which the input data were recorded simultaneously. Figure 5D maps the relationship 271 between measured radon concentration, *c*, and the outer temperature, *T*_o, by plotting 272 the number of events that represent a single pair of the variables. The vertical line to the 273 274 left denotes the freezing point of water while the vertical line to the right denotes no temperature gradient. This panel reveals a complex, nonlinear, seasonal dependency as 275 higher radon concentration pairs with both lower outer temperatures during the 276 winter, when cave ventilation is dominated by the chimney effect, and higher outer 277 temperatures during the summer, when ventilation follows Torricelli's Law. The 278 assumption that ventilation ceases in the absence of a temperature gradient and, 279 consequently, increases radon concentration is not observed in our model. Far from 280 being a maximum, at $T_o \approx T_i$, the number of events that represent a single pair of the 281 282 variables appears to be close to a minimum.

Given that cave radon concentration measurements reflect the outcome of a perpetual 284 competition which pitches flux against ventilation and radioactive decay then the 285 aforementioned contradiction adds further weight to the idea that flux is not a constant. 286 Figure 5E maps the relationship between measured radon concentration, *c*, and the 287 intermediate parameter, T*, by plotting the number of events that represent a single 288 pair of the variables. The intermediate parameter, *T**, captures the majority of the 289 290 experimental data, depicted by warmer colours, in a manner that cannot be attained by linear combinations. This panel, first, demonstrates the close relationship between 291 radon concentration and the temperature gradient and, second, suggests that greater 292 radon concentration measurements pair with faster ventilation rates. The second point 293 is worthy of elaboration. Irrespective of whether radon migrates by diffusion or 294 295 advection, its ultimate source has to be the confining rock mass, so the suggestion that greater radon concentration measurements pair with faster ventilation rates implies 296 that ventilation is able to act as a pump which draws radon into the cave. It confirms the 297 nonlinear dependence between flux and the temperature gradient. 298

299

Figure 6 interrogates flux modelled as a function of the input data obtained at Driny 300 301 Cave. Figure 6A maps the relationship between the measurements of radon concentration, *c*, and the modelled flux, φ , by plotting the number of events that 302 represent a single pair of the variables. This panel demonstrates, first, the close 303 relationship which exists between the measurements of radon concentration and the 304 modelled flux and, second, that the modelled flux is characterised by a generally narrow 305 306 set of values. The second point is once again worthy of elaboration. There appears to be a reasonable correspondence between the flux values we have obtained though our 307 modelling and those used as constants during cave ventilation modelling (see, for 308 example, Gregorič et al. 2011). It emphasises that this mathematical simplification is 309 acceptable as a first approximation. Figure 6B plots modelled flux as a function of time. 310 This panel shows a series of flux anomalies rising above a seemingly oscillatory 311 312 background. The four most significant anomalies occurred in January 2010, October 2010, November 2010, and March 2011. In the following section the timing of these 313 anomalies is discussed in relation to independent experimental observations of fault 314 slip. 315

Figure 7 compares measurements of radon concentration and radon concentration 317 predictions for three different time horizons made using Equation 6. The input data are 318 presented in Figure 5 and the flux is obtained using Equation 5. Figure 7A-C presents 319 data pertaining to a thirty minute time horizon, Figure 7D-F presents data pertaining to 320 an eight hour time horizon, and Figure 7G-I presents data pertaining to a twenty four 321 time horizon. For each time horizon the measurements of radon concentration and 322 323 radon concentration predictions are compared as a function of time; the relationship between the measurements of radon concentration and radon concentration 324 predictions are mapped; and the difference between the measurements of radon 325 concentration and radon concentration predictions are plotted - positive residuum 326 indicates overestimation of the radon concentration while negative residuum indicates 327 328 its underestimation. It is clear that there are close relationships between the measurements of radon concentration and radon concentration predictions. With a 329 thirty minute time horizon our radon concentration predictions never deviate from the 330 measured values by more than 100 Bq/m^3 while with a twenty four hour time horizon 331 our radon concentration predictions rarely deviate from the measured values by more 332 than 1000 Bq/m³. It is also clear that overestimating the predicted radon concentration 333 334 is more common than underestimating it.

335

336 **6. DISCUSSION**

337 6.1 The variable γ

The variable γ was introduced in the second stage of the numerical model to consolidate 338 339 all the parameters related to cave geometry. It accounts for the proportion of the ceiling or entrance which acts as an aperture for air flow and incorporates a length scale equal 340 to the height of the cave or the magnitude of the length of the cave. Three plots of flux 341 modelled as a function of the input data from Driny Cave are presented in order to 342 interrogate the extent to which our model is sensitive to the parametric inputs used to 343 define the variable γ (Figure 8). In all three cases the height is fixed to forty metres, as 344 345 at Driny Cave, and the length is fixed to one hundred metres. These parameters have been fixed because they are generally easier to estimate than the proportion of the 346 ceiling or entrance which acts as an aperture for air flow. The first case estimates that 347 the proportion of the ceiling or entrance which acts an aperture for air flow equates to 348 0.1 % (Figure 8A). The second case estimates that the proportion of the ceiling or 349

entrance which acts an aperture for air flow equates to 1 % (Figure 8B). The third case
estimates that the proportion of the ceiling or entrance which acts an aperture for air
flow equates to 10 % (Figure 8C). It should be noted that the values used for our
numerical model are very similar to those presented in the second case (cf. Figure 6B &
Figure 8B).

355

Figure 8 demonstrates, first, that the flux anomalies occur at precisely the same times 356 irrespective of the proportion of the ceiling or entrance assigned to act as an aperture 357 for air flow and, second, that the magnitude of the flux anomalies remains comparable 358 irrespective of the proportion of the ceiling or entrance assigned to acts as an aperture 359 for air flow. The most conspicuous changes relate to the magnitude of the flux in the 360 361 oscillatory background but such changes are not relevant in the context of the present study. Inspection of the parameters consolidated by γ reveals that its range must fall 362 between $\gamma = 0$, if air flow is completely impeded, and $\gamma \approx 1$, if the entirety of the 363 entrance or ceiling is completely open (this scenario is clearly a physical impossibility). 364 Typical cave geometries will result in γ being ascribed a value ranging from 365 approximately 0.01 to approximately 0.0001. If the ascribed value is smaller than the 366 367 actual value then this would underestimate the role played by ventilation whereas if the ascribed value is greater than the actual value then this would underestimate the roles 368 played by flux and radioactive decay. The comparison presented here demonstrates that 369 the use of approximations will not fundamentally change the results of the numerical 370 modelling, as long as completely unrealistic values are not ascribed. The variable γ , 371 372 which itself has no physical meaning, simply acts as a scaling coefficient.

373

6.2 Flux anomalies and fault slip

The validation of our numerical model is hindered because flux cannot be measured 375 directly and measurements of radon concentration are needed as inputs in the model. 376 To overcome this problem we compare our flux anomalies to independent experimental 377 378 observations of fault slip recorded by three mechanical extensioneters in Driny Cave. This site is one of more than one hundred which comprise the European fault 379 displacement monitoring network EU-TecNet. The extensometers measure 380 displacement in a three dimensional Cartesian coordinate system: the *x*-coordinate 381 represents dilation across the fault; the *v*-coordinate represents strike slip 382

displacements; and the z-coordinate represents dip slip displacement. Typically the 383 strike slip and dip slip displacements at any given monitoring point are characterised by 384 progressive creep trends. These are sometimes interrupted by shorter periods of 385 anomalous activity during which one or both of the slip components may be affected by, 386 for example, conspicuous reversals in the progressive creep trend; a sudden enduring 387 displacement; or a series of oscillatory displacements (see, for example, Briestenský et 388 al. 2015). It is important to note that parallel faults tend to move simultaneously but 389 their sense of movement may differ while perpendicular faults may or may not interact 390 with one another. 391

392

The four most significant flux anomalies occurred in January 2010, October 2010, 393 394 November 2010, and March 2011. Figure 9A presents the strike slip and dip slip displacements from Driny 1; Figure 9B presents the strike slip and dip slip 395 displacements from Driny 2; Figure 9C presents the strike slip and dip slip 396 397 displacements from Driny 3. The anomaly in January 2010 is reflected by a significant dip slip displacement at Driny 1 and a significant strike slip displacement at Driny 2. 398 The anomalies in October 2010 and November 2010 are reflected by a series of dip slip 399 400 displacements, incorporating reversals in the sense of movement, at Driny 3. The anomaly in March 2011 is reflected by a significant dip slip displacements at Driny 3. In 401 all instances the anomalous fault displacements preceded the modelled flux anomaly. It 402 has been proposed that fault slip at Driny Cave reflects both slope deformation and 403 tectonic deformation (Briestenský et al. 2011a). Irrespective of the specific geodynamic 404 405 process, it is clear that fault slip plays an important role in facilitating radon migration along discontinuities in the crust. The novelty of our experimental design should not be 406 overlooked. It is only possible for us to compare our flux anomalies to experimental 407 observations of fault slip because, first, we monitor faults with different orientations 408 and, second, we are able to record displacements in a three dimensional Cartesian 409 coordinate system. 410

411

412 **6.3 Using the numerical model**

To summarise, the presented numerical model is straightforward to use, as illustrated
by Figure 3. The dynamic variables are represented by the outer temperature, the inner
temperature, and the radon concentration while the static variables are represented by

the radioactive decay constant and a range of parameters related to geometry of the 416 cavity. In Section 3 it was stated that the EEC_{Rn} measurements recorded by the radon 417 progeny monitor had been converted into radon concentration by applying a constant 418 radioactive equilibrium coefficient of F = 0.5. It is accepted that the use of a constant 419 equilibrium coefficient does introduce uncertainties into the reported radon 420 concentration. This approach, however, is justified on the following grounds. The focal 421 point of the presented study is the mass balance equation and, in particular, the 422 dynamic nature of the flux term. In this context, the precise calculation of radon 423 concentration is less important than being able to identify the general patterns. If we 424 were to apply a dynamic radioactive equilibrium coefficient then each of the 425 equilibrium equivalent concentration measurements is effectively subjected to a 426 427 different scaling coefficient. This may serve to modulate the amplitude of a flux anomaly but it cannot change the timing of an anomaly nor can it create or eradicate flux 428 anomalies. In this regard it is important to recall that the four most significant flux 429 anomalies rise above baseline values by a factor of approximately 4. Furthermore, the 430 close relationship between our modelled flux anomalies and independent observations 431 of fault slip also indicates that our use of a constant radioactive equilibrium coefficient 432 433 is appropriate.

434

The presented numerical modelling procedure incorporates a small number of 435 assumptions. First, that air flow velocity is driven solely by the temperature gradient, 436 second, that radon diffusion is negligible and, third, that flux is spatially uniform. It 437 would be possible to develop and tune more sophisticated versions of the numerical 438 model so that it is better able to account for site specific conditions including those 439 encountered in other underground spaces such as tunnels or mines. For example, in 440 certain situations, it may be important to incorporate a time lag between radon 441 concentration measurements and ambient temperature measurements; it may be 442 important to incorporate differences in hydrostatic pressure recorded inside and 443 444 outside the cave; and it may be important to reintroduce the diffusion term back into the mass balance equation at sites where the distance from any given entrance to the 445 radon monitoring point is small. There are many advantages to being able to produce 446 detailed numerical models that can precisely replicate a wide range of variables. The 447 ultimate aim of this research, however, is to establish a network of monitoring points in 448

order to deliver the first real time global radon flux maps. This turns out to be feasible
because we can isolate flux in the mass balance equation and compute radon
concentration for successive iterations using only routinely available data: radon
concentration, inner temperature, and outer temperature. The synchronised analysis of
data obtained from across extensive geographical areas would illuminate our
understanding of radon migration along crustal discontinuities and our understanding
of its subsequent exhalation into the atmosphere.

456

457 **7. CONCLUSIONS**

The dynamic nature of radon flux has been investigated in this paper. The first part of 458 the presented numerical modelling procedure focuses on calculating cave air flow 459 460 velocity while the second part isolates flux in a mass balance equation to simulate real time dependence among the variables. With this information it was then possible to 461 deliver an expression for computing cave radon concentration for successive iterations. 462 Input data for the model were recorded at Driny Cave in the geodynamically active 463 Little Carpathians Mountains of western Slovakia. Here the cave passages have 464 developed along faults striking broadly NE-SW and NW-SE. Independent experimental 465 466 observations of fault slip were provided by three permanently installed mechanical extensometers. Our numerical modelling has revealed four important flux anomalies 467 while extensometric measurements demonstrate that each of these was preceded by 468 conspicuous fault slip anomalies. The mathematical procedure outlined in this paper 469 will help to improve our understanding of radon migration along crustal discontinuities 470 471 and its subsequent exhalation into the atmosphere at the site specific scale. Furthermore, as it is possible to supply the model with continuous data, future research 472 will focus on establishing a series of underground monitoring sites with the aim of 473 generating the first real time global radon flux maps. 474

475

476 **ACKNOWLEDGEMENTS**

The authors would like to thank Peter Zvonár, Sara Argerich-Bergada, Amanda KeenZebert, Lenka Thinová, and Petr Otáhal as well as the reviewers whose constructive
comments have helped to improve the clarity of the manuscript. This study was
conducted with support from the long term conceptual development research
organisation RVO: 67985891. It is published in the framework of CzechGeo-EPOS

- 482 "Distributed system of permanent observatory measurements and temporary
- 483 monitoring of geophysical fields in the Czech Republic" (MŠMT Project: LM2010008).
- 484

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593 **FIGURE CAPTIONS**

594

The location of Driny Cave in the Little Carpathians Mountains of western Figure 1 595 Slovakia. This range trends SW-NE along the southeastern margin of the 596 Bohemian Massif and forms part of the Alpine-Carpathian Orogenic Belt. The 597 cave passages have developed along splays of the NE-SW striking Smolenice 598 Fault, which hereabouts marks the contact between the Little Carpathians 599 and the Pannonian Basin, and a series of transverse faults striking NW-SE. 600 Detailed information about the cave and its layout is presented in Michalík et 601 al. (1992) and Bella (2006). 602

603

604 Figure 2 (a) A schematic representation of a cave with an upper entrance and a lower entrance modelled with a height, *H*, a temperature, *T_i*, a radon concentration, 605 *c*, the latter provided by spontaneous flux, ϕ . The outer temperature, T_{o} , and 606 atmospheric pressure, *P*_{atm}, are also labelled while the arrows denote air 607 flow at velocities V. (b) Plot of the modelled flow velocities for air passing 608 through either the upper entrance, if $T_i > T_o$, or the lower entrance, if $T_o > T_i$. 609 (c) Profile extracted from the modelled air flow velocities which emphasises, 610 first, that air flow ceases when there is no temperature gradient and, second, 611 that the air flow velocity is not a linear function of the temperature gradient. 612

613

614Figure 3Flow diagram summarising the numerical modelling procedure needed to615isolate flux in the mass balance equation and to compute the radon616concentration for successive time instants. The dynamic variables in the617model are represented by the outer temperature, the inner temperature, and618the radon concentration while the static variables are represented by the619radioactive decay constant and a range of parameters related to geometry of620the cavity.

621

Figure 4 An illustrative numerical simulation demonstrating the relevance of
Equation 6. (a) A hypothetical ventilation scenario over an arbitrary period.
The first half of this period is characterised by increase and decrease in the
air flow velocity while the second half is characterised by a complete absence

626of ventilation. (b) A hypothetical flux scenario over the same arbitrary627period during which constant flux is interrupted by four spontaneous628anomalies. (c) The iterative estimations of radon concentration over the629same arbitrary period as calculated using Equation 6.

630

(a-c) Time series of inner temperature, T_{i} , outer temperature, T_{o} , and radon Figure 5 631 concentration, *c*, recorded at Driny Cave for the period January 2010 to 632 August 2011. (d) Plot mapping measurements of radon concentration, *c*, 633 against outer temperature, T_{o} , in which the vertical line on the left denotes 634 the freezing point of water and the vertical line on the right denotes no 635 temperature gradient. (e) Plot mapping measurements of radon 636 concentration, *c*, against the intermediate parameter, *T**, in which the red 637 dashed line emphasises the strong relationship between the measured radon 638 concentration and the temperature gradient. The colour scale represents the 639 number of times an event reoccurs on the corresponding ordinate and 640 abscissa pairing. 641

642

Figure 6 (a) Plot mapping radon concentration for successive time instants against
flux modelled as a function of the input data recorded at Driny Cave from
January 2010 to August 2011. (b) Flux through time modelled as a function
of the input data recorded at Driny Cave from January 2010 to August 2011.
This plot shows four significant anomalies in January 2010, October 2010,
November 2010, and March 2011.

649

Figure 7 (a) Measured radon concentration, indicated by red dots, compared to radon 650 concentration predictions made with a thirty minute time horizon using Eq. 651 6, indicated by a blue line. (b) Plot mapping measured radon concentration 652 against radon concentration predictions made with a thirty minute time 653 654 horizon. (c) The difference between the radon concentration predictions made with a thirty minute time horizon and the *a posteriori* measured radon 655 concentration. (d) Measured radon concentration, indicated by red dots, 656 compared to radon concentration predictions made with an eight hour time 657 horizon using Eq. 6, indicated by a blue line. (e) Plot mapping measured 658

659	radon concentration against radon concentration predictions made with an
660	eight hour time horizon. (f) The difference between the radon concentration
661	predictions made with an eight hour time horizon and the <i>a posteriori</i>
662	measured radon concentration. (g) Measured radon concentration, indicated
663	by red dots, compared to radon concentration predictions made with a
664	twenty four hour time horizon using Eq. 6, indicated by a blue line. (h) Plot
665	mapping measured radon concentration against radon concentration
666	predictions made with a twenty four time horizon. (i) The difference
667	between the radon concentration predictions made with a twenty four time
668	horizon and the <i>a posteriori</i> measured radon concentration.

669

670 Figure 8 Flux modelled through time as a function of the input data in order to interrogate the extent to which our model is sensitive to the parametric 671 inputs. (a) The proportion of the ceiling or entrance which acts an aperture 672 for air flow equates to 0.1 %. (b) The proportion of the ceiling or entrance 673 which acts an aperture for air flow equates to 1 %. (c) The proportion of the 674 ceiling or entrance which acts an aperture for air flow equates to 10 %. In all 675 three cases the cave height is fixed to 40 m and the cave length is fixed to 100 676 m. 677

678

Time series of direct experimental observations of fault slip recorded across Figure 9 679 three faults in Driny Cave. The red lines indicate strike slip displacement; the 680 blue lines indicate dip slip displacement; and the grey areas denote periods 681 of three weeks either side of the modelled flux anomalies. (a) The data 682 recorded at monitoring point Driny 1 show a significant dip slip 683 displacement before the flux anomaly in January 2010. (b) The data 684 recorded at monitoring point Driny 2 show a significant strike slip 685 displacement before the flux anomaly in January 2010. (c) The data recorded 686 687 at monitoring point Driny 3 show significant strike slip displacements before the flux anomalies in October 2010, November 2010, and March 2011. The 688 radon progeny monitor was located in the immediate vicinity of monitoring 689 points Driny 2 and Driny 3. More detailed information about this research is 690 presented in Briestenský et al. (2011a,b). 691

TABLES

 Table 1A summary of the fault slip trends recorded at the three monitoring points in
Driny Cave over a nine year period between January 2006 and January 2015.

Monitoring point	Monitored fault	Total strike slip (mm)	Annual slip rate (mm yr ⁻¹)	Sense of movement	Total dip slip (mm)	Annual slip rate (mm yr ⁻¹)	Sense of movement
Driny 1	Smolenice Fault (splay)	0.302	0.034	Sinistral	0.422	0.047	NW block uplift
Driny 2	Smolenice Fault (splay)	0.176	0.019	Sinistral	0.11	0.012	NW block uplift
Driny 3	Transverse Fault (unnamed)	0.223	0.025	Dextral	0.171	0.017	NE block uplift

Figure 1 Click here to download high resolution image



49°0'0"N

48°0'0"N

Figure 2 Click here to download high resolution image



Figure 3 Click here to download high resolution image









Figure 7 Click here to download high resolution image





