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1	Modeling singular mineralization processes due to fluid pressure fluctuations
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11	
12	Abstract
13	Mineralization in the Earth's crust can be regarded as a singular process resulting in large amounts of
14	mass accumulation and element enrichment over short time or space scales. The elemental concentrations
15	modeled by fractals and multifractals show self-similarity and scale-invariant properties. We take the view
16	that fluid-pressure variations in response to earthquakes or fault rupture are primarily responsible for
17	
	changes in solubility and trigger transient physical and chemical variations in ore-forming fluids that
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18 19 20 21	changes in solubility and trigger transient physical and chemical variations in ore-forming fluids that enhance the mineralization process. Based on this general concept, we investigated mineral precipitation processes driven by rapid fluid pressure reductions by coupling mineralization to a cellular automaton model to reveal the nonlinear mechanism of the orogenic gold mineralization process using simulation. In the model, fluid pressure can increase to the rock failure condition, which was set as lithostatic pressure at

23 pressure resulting from fault rupture or local hydrofracture may induce repeated gold precipitation. The

24 geochemical patterns generated by the model evolve from depletion to enrichment patterns, and from

25 spatially random to spatially clustered structures quantified by multifractal models and geostatistics. 26 Results show how metal elements self-organize to form high metal concentration patterns displaying self-27 similarity and scale-invariance. These transitions are attributed to the growth and coalescence of sub-28 networks with different fluid pressures up to the percolation threshold, resulting in a wide range of fluid 29 pressure reductions and mineral precipitation in the form of clusters. The results suggest that cyclic 30 evolution of fluid pressure and its effects on gold precipitation systems can effectively mimic the repeated 31 mineralization superposition process, and generate complex geochemical patterns characterized by a 32 multifractal model. The nonlinear behavior exhibits scale-invariance and self-organized critical threshold,s 33 where mineral phase separations result from fluid pressure reductions associated with fault failure.

34 Keywords: Singular mineralization process; Fluid pressure fluctuation; Cellular automaton; Self-organized
 35 criticality

36

37 1. Introduction

38 The interactions of fluid flow, seismicity, and mineral precipitation can control the mechanical 39 strength and permeability of faults in the earth's crust, and are indispensable components in the 40 development of hydrothermal systems and the formation of ore deposits (Sibson, 1987; Sibson et al., 1988; 41 Cox, 1995; Weatherley and Henley, 2013). Mechanisms responsible for generation and maintenance of high 42 fluid pressure are closely related to the spatial (Rice, 1992) and temporal (Walder and Nur, 1984) 43 variations of local permeability. In seismic zones, fluid pressure increases within impermeable zones and 44 occurs from creep compaction (Sprunt and Nur, 1977), pressure solution (Sleep and Blanpied, 1992), 45 fracture healing and sealing (Walder and Nur, 1984; Sibson et al., 1988; Blanpied et al., 1992). In addition, 46 direct fluid sources involving fluids at depth or from devolatilization reactions have also been considered 47 as mechanisms for elevated fluid pressure (Rice, 1992; Ko et al., 1995; Miller et al., 2003; Bodnar et al., 2007). Once the fluid pressure increases to a level sufficient to permit frictional slip at low fault shear stress, permeability instantaneously increases by several orders of magnitude to locally extremely high values (Miller and Nur, 2000). The rapid fluid pressure reductions due to fault ruptures can induce boiling, or phase separation of ore fluids contributing to mineral deposition (Sibson, 1987; Sibson et al., 1988; Wilkinson and Johnston, 1996; Weatherley and Henley, 2013; Peterson and Mavrogenes, 2014). Rapid deposition during a fluid pressure decrease seals fractures, returning permeability again to a very low value, and the cycle repeats (Sibson et al., 1988; Sibson, 1992).

55 This well-known fault-valve process is widely linked to the formation of mesothermal gold mineralization 56 because the rapid fluid pressure reductions due to fault ruptures can result in anomalous enrichment of 57 elements in small ore bodies within a relatively short period of time (Sibson et al., 1988; Wilkinson and 58 Johnston, 1996; Weatherley and Henley, 2013; Peterson and Mavrogenes, 2014; Sanchez-Alfaro et al., 2016; 59 Moncada et al., 2019). This process is in accordance with the singular mineralization process, which can 60 result in anomalous amounts of mass accumulation and element enrichment within a narrow spatio-61 temporal interval (Cheng, 2007, 2008; Zuo et al., 2009). The end products of the singular mineralization 62 processes often show complex non-linear properties, and can be modeled by fractals and multifractals 63 (Cheng et al., 1994; Cheng and Agterberg, 1996; Agterberg, 1995; Cheng et al., 2000; Cheng, 2007, 2008; Zuo 64 et al., 2009; Zuo and Wang, 2016; Zuo, 2016, 2018).

The concept of fractals introduced by Mandelbrot (1983) primarily represents irregular geometry by its Hausdorff (or fractal) dimension, which is greater than its topological dimension. Multifractals are spatially intertwined fractals with a continuous spectrum of fractal dimensions, which can be used for describing complexity and self-similarity in nature. Examples include the spatial distribution of geological and geochemical quantities, such as mineralization-related element concentrations in rock or related surface media, such as water, soils, and stream sediments (Cheng et al., 1994; Cheng and Agterberg, 1996; Cheng, 1999; Cheng, 2007). Both deterministic and stochastic physical models, such as self-organized 72 criticality, multiplicative cascade processes, diffusion limited aggregation, turbulence and Brownian 73 motion (Bak et al., 1987; Schertzer and Lovejoy, 1987; Evertsz and Mandelbrot, 1992) illustrate the 74 generation of fractals or multifractals. For example, the theory and concept of multiplicative cascade 75 processes play an important role describing intermittent turbulence and nonlinear processes (Schertzer 76 and Lovejoy, 1987). The de Wijs model (De Wijs, 1951; Agterberg, 2001) is a simple multiplicative cascade 77 model widely applied for explaining the generation mechanism of multifractal patterns and their basic 78 singularity characteristics in regional exploration geochemistry (Agterberg, 2001, 2007; Cheng, 2005; Xie 79 and Bao, 2004). However, they cannot efficiently reflect the effects of the variations in extreme physical 80 processes (e.g. fluid pressure fluctuation) on the evolution of the hydrothermal systems. As a numerical 81 equivalent of the fault valve model, the coupled cellular automaton with shear stress and fluid pressure 82 proposed by Miller et al. (1996, 1999) is expected to simulate the singular mineralization process from fluid 83 pressure fluctuation. In this model, fluid flow within a fault zone is modeled as a simple cellular 84 automaton model with a 'toggle switch' permeability assumption (Miller and Nur, 2000). That is, 85 permeability is set to two extreme states, either zero when the fluid pressure fails to reach the failure 86 conditions along the fault plane, or infinite to the nearest neighbors when the fluid pressure reaches the 87 condition, and a dilatant slip event occurs (Miller and Nur, 2000). The dynamical system between shear 88 stress and the state of the fluid pressure exhibits an evolution to a complex stress state that results in scale-89 invariant and self-organizing behavior (Miller et al., 1996, 1999; Miller and Nur, 2000; Fitzenz and Miller, 90 2001; Miller, 2002; Miller et al., 2003).

91 However, these models focused on the dynamic interaction between earthquakes and dehydration 92 reactions, and did not address mineral precipitation processes associated with rapid fluid pressure 93 reductions. Gold solubility in hydrothermal solutions is dominantly controlled by temperature, pressure, 94 pH, and redox (e.g., Seward, 1973). A drop in pressure alone initiates gold precipitation (Loucks and 95 Mavrogenes, 1999), yet decompression also triggers phase separation where the exsolution of volatiles 96 drastically alters fluid chemistry to induce precipitation. Recent studies have suggested that precious 97 metal solubilities are strongly dependent on water vapor phase as the density of the fluid changes, which 98 is an indirect measure of changing fluid pressure (Migdisov and Williams-Jones, 2013). The abrupt 99 reductions in fluid pressure may have a dramatic effect on the aqueous solubility of quartz (Walther and 100 Helgeson, 1977) and are likely to play a major role in co-precipitation of gold with silica during each fault 101 rupture (Helgeson and Lichtner, 1987; Migdisov and Williams-Jones, 2013; Weatherley and Henley, 2013).

In this study, we integrated the mineral precipitation process into the fluid flow cellular automaton to investigate the basic nonlinear behaviors of the orogenic gold mineralization process during rapid fluid reductions due to fault failure at a depth of 10km (270MPa). We coupled gold precipitation processes to the model of Miller and Nur (2000) to investigate how metal elements self-organize to form ore deposits with high metal concentrations showing self-similarity. The complexity and self-similarity of generated metal concentrations was further quantified by a multifractal model and geostatistics (Matheron, 1962; Goovaerts, 1999).

109

110 2. Models

111 2.1 Numerical model

112 Cellular automata can generate very complex forms according to a simple set of local rules 113 governing interactions among nearest neighbors, and thus are attractive for the study of critical 114 phenomena and phase transitions (Wolfram, 1984; Bak et al., 1987; Bak and Tang, 1989; Miller et al., 1996). 115 We assume a grid of cells at depth representing a cross section through an active fault zone. The state of each cell within a fault plane is determined by fluid pressure P_{f_r} which is in hydraulic isolation from its 116 117 neighbors until a failure condition is reached. When the fluid pressure is sufficient to induce hydro-118 fracture or other failure mechanism such as frictional sliding, the permeability is assumed infinite to the 119 nearest neighboring cells and fluid pressure equilibrates with neighboring cells by conserving fluid mass.

120 The fluid pressure in each cell within the impermeable fault zone is increased at a uniform driving rate at121 each time-step (t):

122
$$P_f \rightarrow P_f + \frac{\partial P_f}{\partial t}|_{noflow}, \ \frac{\partial P_f}{\partial t}|_{noflow} = \frac{(\dot{\Gamma} - \dot{\phi})_i}{\phi_i(\beta_{\phi} + \beta_f)_i}$$
 (Eq. 1)

where $\dot{\Gamma} - \dot{\phi}$ represents the fluid pressure source coupled with a time dependent porosity reduction $(-\dot{\phi})$ 123 and a direct fluid source ($\dot{\Gamma}$), ϕ_i represents the initial porosity in cell *i*; and β_{ϕ} and β_f represent the pore 124 and fluid compressibility, respectively, often lumped into a single parameter $\beta = \beta_{\phi} + \beta_{f}$ (Segall and Rice, 125 126 1995; Wong et al., 1997). Porosity reduction mechanisms (e.g., fault compaction and pressure solution) and 127 a direct fluid source (e.g., dehydration/decarburization reactions) contribute to the increases of fluid 128 pressure acting on discrete cells of a zero permeability fault plane. Once fluid pressure exceeds the 129 lithostatic load, failure occurs, and the failed cells and their immediate neighboring cells are labeled. The 130 fluid pressure instantaneously equilibrates with these hydraulically connected cells by conserving fluid 131 mass, ignoring any gravity effect. The equilibrium fluid pressure within the affected cells updates to:

132
$$\overline{P} = \frac{\sum_{i=1}^{N} (\phi \beta)_i P_i}{\sum_{i=1}^{N} (\phi \beta)_i}$$
(Eq. 2)

where P_i and \overline{P} represent the pre-failure and post-failure pore pressure among the affected cells, respectively, and *N* is the number of affected cells. The fluid pressure redistribution might cause the neighboring cells to reach the failure condition, leading to further pressure equilibrium and cascading failure until the stress value in all the cells recovers to below the failure condition. It is important to note that the numerous mechanisms (e.g., crack porosity production due to hydro-fracture, variation of mechanical strength, and time-dependent healing), that are responsible for the evolution of fluid pressure, are simplified in this model (Miller and Nur, 2000). 140 Once the fault rupture occurs, the abrupt drop in fluid pressure toward hydrostatic values triggers 141 mineral precipitation in the fracture network, which can seal fractures to rebuild the fluid pressure and 142 ensure that the cycle repeats. The relationship between the solubility of elements and fluid pressure is 143 different for different temperature ranges. Some studies suggest that the solubility of metal ion species (e.g. 144 Ag, Au, Cu and Sn) decreases log-linearly with decreasing water vapor pressure (Migdisov et al., 1999; 145 Archibald et al., 2001, 2002; Migdisov and Williams-Jones, 2005). Other studies show an exponential 146 relationship between metal ions and water vapor pressure with the change of temperature (Bischoff et al., 147 1986, 1988; Rempel et al., 2006; Migdisov and Williams-Jones, 2013; Migdisov et al., 2014). These studies 148 indirectly reflect that the fluid pressure can efficiently affect the solubility of precious metal solubilities.

149 We recognize that fluid pressure is not the only factor that controls metal solubility in hydrothermal 150 systems, and other physical and chemical factors, such as temperature, pH and redox, may be as 151 important or more important in some cases (Seward, 1973). Here, we ignore these other factors for the sake 152 of simplicity and focus only on the role of pressure decrease as the mechanism of metal 153 deposition. However, if the hydrothermal fluid is still undersaturated after the solubility decreases due to 154 an abrupt pressure drop, the metal will not precipitate. Thus, to simplify the model, we suppose that each 155 solubility decrease can lead to metal precipitation because the subsequent recovery stage flow of fluid 156 from the surroundings into the sealing factures can progressively build high concentration hydrothermal 157 fluid cycle by cycle (Weatherley and Henley, 2013).

In this study, we used three different relationships between metal ions and fluid pressure. Specifically, we investigate linear, exponential and power-law functions (Eq. 3) to estimate the volume of mineral precipitation.

161
$$\begin{cases} Linear : \log S_{metal} = A + B \cdot \log P_{f} \\ Power : \log S_{metal} = A + B \cdot (\log P_{f})^{C} \\ Exponential : \log S_{metal} = A + B \cdot C^{\log P_{f}} \end{cases}$$
(Eq. 3)

162 Taking the linear relationship as an example, the volume of mineral precipitation due to rapid fluid163 pressure reductions can be estimated by:

164
$$C_{metal} = \left(S_{metal} - \overline{S}_{metal}\right) \cdot \phi \beta = 10^A \cdot \left(P_f^B - \overline{P}_f^B\right) \cdot \phi \beta$$
 (Eq. 4)

Here C_{metal} represents the mineral element concentration in the cells. S_{metal} and \overline{S}_{metal} represent the solubility of metals corresponding to pre-failure fluid pressure P_f and post-failure fluid pressure \overline{P}_f , respectively, and *A*, *B* and *C* are constants. The cell storage capacity ($\phi\beta$) is considered as the fluid mass of each cell.

169

170 2.2 Multifractal model and singularity

We assume that the total concentration of deposited metal elements (e.g., Au or Ag) in the *i*-th cell with a linear measuring scale ε satisfies $\mu_i(\varepsilon) \propto \varepsilon^{\alpha_i}$ from a multifractal perspective, where α_i represents the singularity index. Different cells possess different singularity indices, hence the total number of cells covering the entire subset bearing the singularity α , $N_{\alpha}(\varepsilon)$, is proportional to $\varepsilon^{-f(\alpha)}(N_{\alpha}(\varepsilon) \propto \varepsilon^{-f(\alpha)})$. The fractal dimension function $f(\alpha)$ is known as a multifractal spectrum, which is usually estimated via

176 the moment method (Halsey et al., 1986). The partition function $\chi_q(\varepsilon)$ is defined as:

177
$$\chi_q(\varepsilon) = \sum_{N(\varepsilon)} \mu_i^q(\varepsilon)$$
 (Eq. 5)

178 The partition function $\chi_q(\varepsilon)$ shows a power-law relationship with cell size ε for any $q \in [-\infty, +\infty]$ if the 179 distribution of $\mu_i(\varepsilon)$ is multifractal,

180
$$\chi_a(\varepsilon) \propto \varepsilon^{\tau(q)}$$
 (Eq. 6)

181 Here $\tau(q)$ represents the mass exponent of order *q*. The index $M = \tau(2) - 2\tau(1) + \tau(0) < 0$ suggests that the

182 measure corresponds to a multifractal, whereas M=0 suggests a fractal or non-fractal.

183 The singularity exponent $\alpha(q)$ and the multifractal spectrum value can be calculated through the mass 184 exponent by differentiation and the Legendre transformation, respectively (Evertsz and Mandelbrot, 1992)

185
$$\alpha(q) = \frac{d\tau(q)}{q}$$
 (Eq. 7)

186 $f[\alpha(q)] = \alpha(q)q - \tau(q)$ (Eq. 8)

187 An asymmetry index $R = (\alpha(0) - \alpha_{\min})/(\alpha_{\max} - \alpha(0))$ is defined to quantify the shape of the entire 188 multifractal spectrum (Xie and Bao, 2004; Cheng, 2014). R > 1 or R < 1 represent a left- or right-skewed 189 shape of the multifractal spectrum indicating that a local enrichment or depletion pattern dominates 190 among the whole set of cells, respectively.

191

192 2.3 Semivariogram

Semivariograms are a key component in geostatistics, and are typically used to quantify the degreeof spatial variability. The semivariogram function can be expressed as:

195
$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_i + h)]^2$$
 (Eq. 9)

196 where $\gamma(h)$ represents the semivariance that quantifies the average dissimilarity between the measured variable at different spatial locations. $Z(x_i)$ and $Z(x_i + h)$ represent the value of a variable at locations *i* 197 198 and i + h, respectively, and N(h) is the number of data pairs separated by a given lag vector h. The basic 199 concepts involved in the semivariogram include a measure of the total variance (the sill, $C_0 + C$), the 200 average length of the spatial dependence (range), and the local variation due to sampling or measurement 201 error (the nugget, C_0). The degree of spatial dependence is determined by the ratio of the nugget to sill: $C_0/$ 202 $C_0+C < 0.25$ (strong spatial dependence), $0.25 < C_0 / C_0+C < 0.75$ (moderate spatial dependence) and C_0 / C_0 203 $C_0+C > 0.75$ (weak spatial dependence) (Cambardella et al., 1994).

204

207 We started the simulation of fluid pressure increases and mineral precipitation process within a test 208 grid of $L \times L$ (L = 200) cells. Different distributions of fluid sources and material properties (simplified to 209 initial porosity and the compressibility of pore space and fluid) may determine the distribution of the fluid 210 pressure rate of increase (Miller and Nur, 2000). In our study, we considered a heterogeneous fault zone 211 where the initial value of compressibility varies, and the source term remain constant. The compressibility 212 was set between 1×10⁻³ MPa⁻¹ and 1×10⁻²MPa⁻¹ based on experimental results from David et al. (1994). The 213 storage capacity was varied from 5×10⁻⁵ MPa⁻¹ to 5×10⁻⁴ MPa⁻¹ by setting the initial porosity to 0.05. The 214 source term was set to a constant value of 1×10⁻⁶ yr⁻¹. The initial fluid pressure in each cell was distributed 215 between the hydrostatic and lithostatic pressures, in which lithostatic pressure was regarded as the failure 216 condition, and set to 270MPa corresponding to a depth of 10km. These parameters result in fluid pressure 217 increasing towards the failure condition at rates ranging from 2-20 kPa/yr. At early simulation times, cells 218 reaching failure are independent in space because the neighbors of the failed cells are far from failure, thus 219 preventing the propagation of the failure event and limiting the event size. We define event size as the 220 total number of cells reaching the failure condition during one discrete time-step. As the simulation 221 evolves, many cells approach the failure condition, and the high pressure in the failed cells can propagate 222 quickly to generate large events. Once failure occurs during the simulation, the fluid pressure in an 223 individual cell or cluster of cells experiences abrupt fluctuations from hydraulic connectivity to low 224 pressure cells. The abrupt fluid pressure drops reduce the solubility of precious metals (e.g. Ag and Au), 225 which is likely to induce co-precipitation of gold with silica during each fault rupture, and further 226 contributes to geochemical variations.

227 Similar to the evolution of high fluid pressure, the spatial distribution of metal elements evolves
228 from a spatially random structure to spatially clustered structures. Nine temporal sequence snapshots (Fig.
229 1) show the evolution and variations of geochemical patterns caused by rapid fluid pressure reductions
230 corresponding to the later time evolution. At early time steps, cells experiencing mineral precipitation are

231 randomly observed in the system due to the random failure of one or only a few cells (Figs. 1a-1d). As the 232 system evolves, more clustered spatial distributions of geochemical patterns are produced due to the 233 occurrence of mineral precipitation among a wider range of more clustered cells (Figs. 1e-1i). The ratio of 234 nugget to sill increases before the evolutionary time of 13.6 kyr and decreases after 13.6 kyr (Fig. 2), 235 indicating that the degree of spatial dependence of geochemical patterns decreases at first and then 236 increases with the further evolution of the system. This transition might be attributed to the establishment 237 of the initial structure of incipient failures and both marks the onset of a correlation length and identifies 238 the percolation threshold of the system. Around this transition point, the number of failure events, 239 cumulative event sizes and correlation length show significant fluctuations (Miller and Nur, 2000). The 240 mineral depositional process occurs in isolated failure cells that increase the spatial randomness of the 241 geochemical patterns before the transition point. When the structure of the incipient failure is established, 242 the sub-networks at different fluid pressures merge and equilibrate according to Eq. 2; thus, a wider range 243 of fluid pressure reductions results in mineral precipitation in the form of clusters, which may enhance the 244 degree of spatial dependence of geochemical patterns.

245 A transitional phenomenon also occurs with multifractal geochemical patterns. Figure 3 shows the 246 relation between the multifractal spectrum value $f(\alpha)$ and the singularity index α for different time 247 periods. The multifractal spectrum curves vary from right deviation to left deviation at approximately 13.5 248 kyr. The increasing asymmetry index (R) with the evolution of the system demonstrates that the 249 geochemical pattern evolves from a dominant local depletion pattern to local enrichment within the entire 250 matrix near the percolation threshold. Fluid pressure reductions varying from small-scale to large-scale 251 determine the scale of superimposition of metal material at different evolutionary stages. The large-scale 252 superimposition makes the components of higher value in the whole system become more pervasively 253 distributed. Thus, the enrichment of element concentrations (asymmetry indexes R in Fig. 3) rapidly increases within a short period of 1.5 kyr. The index *M* decreases with the increase of evolutionary time,indicating increasingly higher degrees of multifractality (Fig. 4).

256 Simulations end when the average fluid pressure of the system reaches the failure condition. Figure 5 257 shows the corresponding final geochemical pattern, with a high degree of spatial dependence and local 258 enrichment. The highly enriched area was found to be distributed at or near the cells with high storage 259 capacity because high storage capacity equalizes the fluid pressure to a greater extent. Therefore, more 260 dramatic fluid pressure variations occur when the cells near the high storage capacity cells fail, resulting in 261 a higher magnitude of mineral depositions. This phenomenon coincides with field observations that 262 mineral deposits or veins occur at or near the faults that determine the random and clustered features of 263 ore deposits in their spatial distributions. For example, Wang et al. (2015) revealed a clustered distribution 264 of Fe deposits in space along NNE-NE trend in Fujian Province, China. The distribution of singularity 265 index α , estimated from $\log[\mu(\varepsilon_1)/\mu(\varepsilon_2)]/\log(\varepsilon_1/\varepsilon_2)$, can quantify the properties of enrichment (α <2) and 266 depletion (α >2) of geochemical elements caused by mineral depositions (Fig. 5b).

267 Producing maps of singularities can provide new information, complementing results based on the 268 original concentration distribution (Fig. 5a) and help to recognize metal concentration anomalies from 269 complex geological regions. The multifractal spectrum of the geochemical patterns is calculated via the 270 method of moments (q), and varying from -10 to 10 in steps of 1. The corresponding parameters, partition 271 function $\chi_{q}(\varepsilon)$, mass exponent $\tau(q)$ and singularity exponent $\alpha(q)$, are shown in Figs. 6a-6c. The 272 multifractal spectrum obtained through a Legendre transformation (Fig. 6d) shows an asymmetric left-273 skewed shape. This asymmetry may reflect the fact that the spatial distribution of concentrations shows a 274 continuous multifractal characteristic, which can be attributed to the periodic local mineral deposition due 275 to fluid pressure fluctuation. The results shown above are based on linear relationships between solubility 276 and fluid pressure, while Figure 7 shows the simulation results with the other two relationships (Eqs. 3b 277 and 3c). These three geochemical patterns show a similar spatial structure (Figs. 7b and 7c), and both of these spatial structures are highly dependent on the distribution of the cells' storage capacity. However,
due to different solubility relationships, the accumulation of mineral precipitation varies, resulting in
different degrees of local enrichment patterns at the end of the simulation as shown by the multifractal
spectrum and the asymmetry index (Fig. 7d).

282

283 4. Discussion and Conclusions

284 We investigated the cycle of fluid pressure increase – hydrofracture – fluid pressure decrease – rapid 285 sealing from precipitation using a cellular automaton model to simulate the singular mineralization 286 process. With the continued increase of fluid pressure within the undrained system, the evolution of the connectivity structure shows the onset of a correlation length at the percolation threshold, after which the 287 288 correlation length increases until the system as a whole reaches a critical state. The relationship between 289 cluster size and the number of events shows a power-law with an exponential tail at the percolation 290 threshold that plays an important role for fracture connectivity and fluid flow in the formation of mineral 291 deposits (Roberts et al., 1998, 1999). At the critical state, the power law statistics of cluster size indicate 292 scale invariance of the fluid pressure evolution system (cf. Fig. 6 in Miller and Nur (2000)), namely the 293 constructed cell space can occur at the level of pore structure, or at the level of large scale fluid pressure 294 within a fault zone. This determines that the distribution of elemental concentration also exhibits the scale-295 invariance property and critical thresholds where mineral phase transitions are induced by fault failure 296 and the system seeks a new attractor (Bak et al, 1987). The scale-invariant property of geochemical patterns 297 suggests that the snapshots in Fig. 5a can be viewed as distributions of elemental concentration at a 298 microscopic scale (e.g. ore samples) or at a metallogenic zone scale. The non-uniform distribution of 299 elemental concentrations on different scales occurred in nature in mineralization systems. For example, the 300 Au concentration distribution in the Dayinggezhuang ore deposit, located in Jiaodong gold province, 301 eastern China, suggest different mineralization density at different scales (Deng et al., 2011). Self302 organized criticality reflects complex mineralization behavior, which is characterized by a bottom-up 303 nature where complex behavior emerges from independent but interdependent interactions of unlimited 304 cells. For example, fluid pressure in cells far from the failure condition increases independently, however, 305 fluid pressures in individual cells or many isolated networks will merge and equalize with each other after 306 the structure of incipient failure is established.

307 The interaction of physical, chemical and biological processes can contribute to mineral deposition 308 through phase transition or separation during the hydrothermal mineralization processes. We considered 309 fluid pressure fluctuations and cyclicity as a dominant process in mineralization, and this cyclicity was 310 responsible for the superposition of repeated mineralization events that ultimately produce complex 311 geochemical patterns that can be effectively modeled using a multifractal framework. Although this 312 idealized model is simple, it is not simplistic, and provides important insights into the singular 313 mineralization process. Future model developments will include other important processes not yet 314 considered, including tectonic stress increases from plate motion, to investigate how different ratios of 315 differential stress to fluid pressure can influence the fault failure patterns which may further determine the 316 ore deposit types (Stephens et al., 2004).

317

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490 Figure 1. Nine temporal sequence snapshots showing the evolution and variations of geochemical patterns491 caused by rapid fluid pressure reductions corresponding to time of evolution; At early times, from (a) to

- (d), cells with mineral deposition randomly occur in the system due to the random failure of one or only a
- 493 few cells. As the system evolves, from (e) to (i), structured spatial distributions of geochemical patterns are
- 494 produced due to the occurrence of mineral deposition among a wider range of clustered cells.





Figure 2. Time line of ratio of nugget to sill obtained from the semivariogram function.





Figure 3. Multifractal spectra of geochemical patterns at different evolution times.



502 Figure 4. Time line of multifractality measuring the irregularity of geochemical spatial dispersion patterns.



505 Figure 5. (a) Spatial distribution of element concentration based on linear relationships between solubility

506 and pressure; (b) Spatial distribution of singularity index α quantifying the properties of local enrichment 507 and depletion of geochemical patterns.



Figure 6. Results of multifractal analysis applied to geochemical pattern of Figure 5(a); (a) Log-log plot of 512 mass-partition function vs. edge size of cell, model parameter q varies from -10 to 10 with 1 interval; (b) 513 Estimates of mass exponent $\tau(q)$ involve slopes of the straight-lines in (a) vs. order *q*; (c) Singularity index 514

515 $\alpha(q)$ and order q; (d) Multifractal spectra value $f(\alpha)$ vs singularity index α .



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Figure 7. Row (a) Three different relationships (linear, exponential and power law) between solubility and
pressure in logarithmic coordinates; Row (b) Spatial distribution of element concentration based on three
different relationships based on Row (a); Row (c) and Row (d) are semivariograms and multifractal
spectrums corresponding to geochemical patterns in Row (b).