

Dear Editor:

Thanks very much for your kind decision. We have carefully read through the comments from referee #2 and made proper revisions. In addition, the writing was checked by an English speaking colleague again after edited by “American Journal Experts” and “ELSEVIER Language Editing Services” successively.

Our responses to the referee’s comments and some additional revisions are listed below. We greatly appreciate your time and efforts to improve our manuscript for publication.

Yours sincerely

Bin Zeng

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A point-by-point response to the referee #2

The comments from referee #2 below did not appear in the paper review system, but from the email by Editorial Support Anna Wenzel.

The paper on Mechanism of groundwater inrush hazard ... is interesting and worth to be printed but not in the present form.

The science in the paper is good and needs no further modification but the language and the style of the manuscript must be changed:

1- English is poor and somewhere it is difficult to understand the meaning of some sentences

Response: We invited an English speaking colleague to check the language of the manuscript and make appropriate revisions. (Before this minor revision, the manuscript was edited by “American Journal Experts” and “ELSEVIER Language Editing Services” successively.)

2- Often along the paper figures 3-4 etc. are wrongly cited (it seems that fig. 2 was added and this presence was not considered in citation along

the manuscript)

Response: We checked the whole manuscript to ensure that no figure was wrongly cited anymore.

3- In the reference list the alphabetic order is sometime wrongly followed

Response: We checked the reference list again, and made proper revisions to ensure that all the references listed in right alphabetical order.

The author also made following additional revisions:

- The Fig. 3 was redrawn so as to improve the clarity.
- All the data in the tables were revised to have the same number after the decimal point.
- We also checked the references in the main text to ensure that every reference cited in the text is also present in the reference list (and vice versa).
- We checked our manuscript carefully for typos, co-authors and their affiliations, terminology, updates of data in tables, and updates of variables in equations.

A marked-up manuscript version

Mechanism of groundwater inrush hazard caused by solution mining in a multilayered rock salt mining area: A case study from in Tongbai-County, China

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1 ABSTRACT

2 The solution mining of salt mineral resources may contaminate groundwater and lead to water
3 inrush out of the ground due to brine leakage. Taking a serious groundwater inrush hazard in a large salt
4 mining area in Tongbai County, China, as an example, this study mainly aims to analyze the source and
5 channel of the inrushing water. The mining area has three different types of ore beds ~~as follows:~~
6 ~~a)including~~ trona (trisodium hydrogencarbonate dihydrate, also sodium sesquicarbonate dihydrate,
7 with the formula $\text{Na}_2\text{CO}_3 \cdot \text{NaHCO}_3 \cdot 2\text{H}_2\text{O}$, is a non-marine evaporite mineral); ~~b)~~ glauber (sodium
8 sulphate, is the inorganic compound with the formula Na_2SO_4 as well as several related hydrates) and ~~c)~~
9 gypsum (a soft sulphate mineral composed of calcium sulphate dihydrate, with the chemical
10 formula $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). Based on the ~~understanding~~ characterizing of geological and hydrogeological
11 conditions, ~~the study first obtained~~ hydrochemical data of the groundwater at different points and depths,
12 ~~and then analysed~~ were used to analyze the pollution source and pollutant component from single or
13 mixed brines by using ~~both~~ physical-chemical reaction principle analysis and ~~a~~ hydrogeochemical
14 simulation method. Finally, possible leakage brine conducting channel to the ground was discussed
15 from both geological and artificial aspects. The results reveal that the brine from the trona mine is the
16 major pollution source; there is a fissure zone in the NW-SE direction controlled by the geological
17 structure that provides the main channels for the leakage brine to flow into the aquifer around the water
18 inrush regions, and a large number of waste gypsum exploration boreholes are the channels that supply
19 the polluted groundwater inrush out of the ground. This research can offer a valuable reference for
20 avoiding and assessing groundwater inrush hazards in similar rock salt mining areas, which is
21 advantageous for both groundwater quality protection and public health.

22 1. Introduction

23 Solution mining is commonly used in salt mine exploitation, as salts are soluble in water. In this
24 method, high-pressure and -temperature water with low salinity is injected into a mineral deposit
25 through production wells to dissolve the mineral salts. After being drawn from the wells, the soluble salt
26 is purified and further processed. However, the high-pressure and -temperature water used in this
27 process not only dissolves minerals but ~~can~~ also cause fractures in the strata, which usually results in
28 hazards such as brine leakage or groundwater inrush. In this situation, ~~underground~~ drinking
29 groundwater for the public is normally polluted following groundwater inrush, creating a hazard and
30 threatening the health of local residents.

31 Many scholars (Clark and Fritz, 1997; Liu et al., 2015; Wu et al., 2016) have studied groundwater
32 inrush hazards in both coal and metal mines, and some adopted methods are as follows: the use of water
33 level-/-temperature criterion (Yuan and Gui, 2005; Ma and Qian, 2014), stochastic simulation
34 (Fernandez-Galvez et al., 2007), numerical simulation (Liu et al., 2009; Kang et al., 2012; Shao et al.,
35 2013; Houben, et al., 2017), water chemical analysis (isotope analysis, water quality type correlation
36 analysis) (Robins, 2002; Fernandez et al., 2005; Hu et al., 2010; Cobbina et al., 2015; Lee et al., 2016;
37 LeDoux et al., 2016), multivariate statistics (discriminant analysis, clustering analysis) (Chen and Li,
38 2009; Lu, 2012) , fractional advection dispersion equations (Ramadas et al., 2015) and nonlinear
39 analysis (fuzzy mathematics, grey correlation analysis, etc.) (Hao et al., 2010; Gao, 2012). However,
40 due to the particularity of the solution mining method and the complex chemical-physical reactions
41 during the high-pressure and -temperature mining process, researches regarding solution mining ~~was~~
42 ~~more~~were mainly focused on mining techniques (Jiang and Jiang, 2004; Kotwica, 2008; Namin et al.,
43 2009), mining cavity stability analysis and sinkhole problems (Staudtmeister and Rokahr, 1997; Bonetto
44 et al., 2008; Ezersky et al., 2009; Goldscheider and Bechtel, 2009; Closson and Abou Karaki, 2009;

45 Vigna et al., 2010; Frumkin et al., 2011; Ezersky and Frumkin, 2013; Qiu, 2011; Blachowski et al.,
46 2014), and geohazards particularly in karst areas due to human-induced underground caving (Waltham
47 and Fookes 2003; Parise and Gunn 2007; Zhou and Beck 2011; Parise and Lollino 2011; Lollino et al.,
48 2013; Gutierrez et al., 2014; Parise et al., 2015), but rarely on source and channel analysis of inrush
49 water in a solution mining accident.

50 The ~~studied~~study case of rock salt mining area is located in Tongbai County, Henan Province,
51 China. This mining area has the second largest trona reserves in the world, while its glauber salt
52 reserves reach 45 million tons. Since trona and glauber salt were put into production in 1990 with
53 single- and double-well convection mining as the main producing method, five inrush points appeared
54 in the town of Anpeng, Tongbai County, from June 2011 to May 2013. Among these five inrush points,
55 four (Y1~Y4) were long-term (longer than 2 years) inrush points with stable discharge, while one (Y-5)
56 was a sudden inrush point (as shown in Fig. 1 and Fig. 2).~~On 1 February 2013, almost~~ Almost 200 m³
57 of mud and sediment erupted out of the ground at the Y-5 point on 1 February 2013. The area of the
58 inrush point was ~~almost~~ ~4 m²; the average water inflow was ~~from~~ 20-30 m³/d while the greatest inflow
59 reached 200 m³/d. The water inrush lasted for approximately three months. During the Y-5 inrush
60 accident, according to the field investigation, a trona production well named “S02,” which is 200 m far
61 from the inrush point, broke at a depth of 234 m and remained broken for a long period of time. It was
62 repaired on 15 March 2013. During the entire water inrush process, the groundwater inrush~~of~~
63 ~~groundwater~~ led to a phenomenon of salinization at the house base ~~of the houses~~ of many villagers, and
64 made water in many residents’ wells no longer drinkable.

65 Since the groundwater inrush hazard involved a wide geographic area and the inrush source was
66 quite hard to distinguish due to the multi-layer distribution of the different ore bodies and the
67 complexity of the inrush water component. Therefore, in order to put forward a targeted treatment

68 program to stop the water inrush as soon as ~~is~~-possible, and mitigate the groundwater pollution in
69 research region, the source and channel of the inrush water were taken as the research emphasis in this
70 study. Furthermore, this research can provide a valuable reference for avoiding and assessing
71 groundwater inrush hazards in similar rock salt mining areas, which is advantageous for both
72 groundwater quality protection and public health.

73 **2. Geological and hydrogeological setting**

74 ***2.1. Geological conditions***

75 The mining area is located in northwestern Tongbai County. The landscape is
76 ~~characterised~~characterized by hollows and ridges, ~~and has~~with an elevation ranging from 140 to 200 m
77 above sea level. The strata, lithology, aquifer, and the position of different ore beds in the research area
78 (Shi et al., 2013) are shown in Fig. 3.

79 According to geologic references and field investigation, in the northeastern mining area, a hidden
80 east-west oriented fault is developed at the bottom of the first segment of the Hetaoyuan Formation, and
81 another four, hidden, south-north oriented faults are developed at the bottom of the second segment of
82 the Hetaoyuan Formation. These five faults are outside the scope of trona mine, so they have ~~had~~-little
83 effects on the ore bed. A few small-scale hidden faults are developed at the bottom of the third segment
84 of the Hetaoyuan Formation, although within the scope of the glauber salt mine, they have ~~had~~ little
85 effects on the glauber salt ore bed which is distributed at the top of the first segment of Hetaoyuan
86 Formation~~5~~5. A hidden east-west oriented fault is developed at the bottom of the Liaozhuang Formation
87 in the range of the glauber salt mine, but it has ~~had~~ little effects on the glauber salt mine because of its
88 small scale.

89 ***2.2. Hydrogeological conditions***

90 The groundwater in the mining area can be divided into pore water in the loose rock mass and

91 bedrock fissure water according to the lithology and hydrogeological features. In the upper part of the
92 Liaozi Formation, a mudstone interbedded with gypsum is considered a relative weak permeable
93 stratum especially under the condition of high-pressure and -temperature water injection during the
94 mining period. The shallow aquifer is unconsolidated pore water above this weak permeable stratum,
95 while the deep aquifer is a bedrock fissure beneath this weak permeable stratum.

96 The flow direction of the shallow groundwater is controlled by the ~~overall~~-regional terrain. Taking
97 the underground watershed as the boundary, the groundwater on the south side of the watershed is
98 mainly flowing from northeast to southwest with the Yanhong River as the drainage base ~~of the drainage~~,
99 while the groundwater on the north side of the watershed is mainly ~~slowing~~-flowing from south to north
100 with the Xia River as the drainage base ~~of the drainage~~. The deep groundwater ~~has-is in~~ relatively closed
101 burial conditions, with slow velocity, and nearly the same flowing direction as the shallow groundwater.
102 The water inflow of a single well with poor water content is approximately 100 m³/d, while it can reach
103 from 1000-2000 m³/d if it has rich water content. The annual amplitude ~~variation~~ of the groundwater
104 level is from 2- to 4 m, while the depth is stable at 2.3-4 m. Residents in Anpeng use groundwater as ~~the~~
105 ~~source of~~ their drinking water, which comes from wells and is from the porous aquifer.

106 ~~As shown in Fig. 3, g~~Gypsum mainly occurs on the top of the Liaozi Formation, glauber salt
107 occurs in the third member of the Hetaoyuan Formation, and the trona occurs at the bottom of the
108 second member of the Hetaoyuan Formation as well as on top of the first member of the Hetaoyuan
109 Formation (Fig. 3). The surrounding rocks of every mineral layer, ~~include~~-including mudstone, shale,
110 sandy conglomerate, psammitic rock and dolomite, ~~which~~-have sufficient thickness and good
111 water-resistance. Therefore, the effect of groundwater on the mineral deposit is minimal in the mining
112 area.

113 **2.3 Distribution and characteristics of the ore body**

114 The three ore bodies overlap in plane distribution, as shown in Fig. 4. The vertical distribution of
115 the ore bodies from deep-to-shallow is trona (buried depth: 1560.92-2929.53 m), glauber salt (buried
116 depth: 1003.66-1397.58 m) and gypsum (buried depth: 134-338 m). The trona and glauber salt bodies
117 are at least 250 m apart from each other vertically.

118 The trona has 11 horizontal layers, with an average thickness of 2.11 m. The chemical composition
119 of trona is mainly NaHCO_3 (average of 77.06%) and Na_2CO_3 (average of 16.33%) (Wang, 1987). The
120 glauber salt has 4 layers, with an average thickness of 8.93 m. The dip angle of the ore bed layer is less
121 than 10° . The average mineral grade is 60.14%. The main composition of the glauber salt is Na_2SO_4
122 (>90%) with a small amount of NaCl.

123 **3. Methods**

124 Based on the field investigation results, the chemical characteristic analysis of the inrush water at
125 different sites and time, analysis of the physical and chemical reaction principles for the different brines,
126 and combined with the PHREEQC simulation method, the source of the inrush water was determined.

127 **3.1. Sampling and testing**

128 The five groundwater inrush points (Y1~Y5) and some shallow groundwater points (resident wells:
129 SY1~SY6) near the accident site were chosen as groundwater quality sampling points, as shown in Fig.
130 4. Water from each point was sampled on 9 March 2013.

131 Water samples were filtered using a $0.45\ \mu\text{m}$ millipore filtration membrane in the field, and then
132 filled with a polyethylene bottle which had been soaked in acid and washed with deionised water.
133 Filtered water samples were acidified until the $\text{pH}<2$ by addition of ultra-pure HNO_3^- for the
134 determination of cations; water samples for the determination of anions were not treated.

135 Elements tested in the laboratory included 26 cations (K^+ , Na^+ , Ca^{2+} , Mg^{2+} , Sr^{2+} , etc.) and 5 anions

136 (F⁻, Cl⁻, NO₃⁻, SO₄²⁻, NO₂⁻). The instrument used for the determination of cations was an inductively
137 coupled plasma atomic emission spectrometer (Agilent ICP-OES 5100), and the minimum detection
138 limit was 0.0001mg/L. The instrument used for the determination of anions was an ion chromatograph
139 (ICS-1100), and the minimum detection limit was 0.001 mg/L. CO₃²⁻ and HCO₃⁻ were tested according
140 to the “Groundwater quality test method: Determination of carbonate and bicarbonate by hydroxide
141 titration (DZ/T 0064.49-93),” and the minimum detection limit was 0.01 mg/L.

142 In addition, from March to April 2013, at the Y-5 and Y-3 points, three water quality automatic
143 recorders (Levellogger gold, Canada) were arranged for inrush water monitoring. Monitoring indicators
144 were temperature, water level and electrical conductivity. The purpose of the monitoring was to fully
145 understand the inrush water quality during the whole accident, especially in the process of well
146 reparation.

147 ***3.2. Analysis of the physical and chemical reaction principles in different brine mixing conditions***

148 During the accident, the leakage brine of the trona (2000 m belowground) or glauber salt (1000 m
149 belowground) might flow through the gypsum deposit (200-400 m belowground), which is comprised
150 primarily of CaSO₄, and cause physical and chemical reactions while it inrushes out of the ground. Thus,
151 the formation of the ~~inrush water~~ chemistry component [in inrush water](#) might be from glauber brine, or
152 trona brine, or a mixture of the two brines, flowing through the gypsum layer ~~with~~ accompanying
153 physical and chemical reactions. To provide the basis for further analysis of the inrush water source, the
154 physical solubility of the gypsum and the reaction when the glauber salt brine, trona brine, or a mixture
155 of trona and glauber salt brine flowing through the gypsum deposits were ~~analysed~~[analyzed](#).

156 ***3.2.1. The physical solubility of gypsum (CaSO₄)***

157 Gypsum is slightly soluble; when in water, its acidity is apparent. Eq. (1) provides the dissolution
158 rate equation of gypsum in water:

159
$$R_{Gypsum} = k_1 \times \frac{A_g}{V} \left(1 - \left(\frac{IAP}{K}\right)_{Gypsum}\right)$$
 (1)

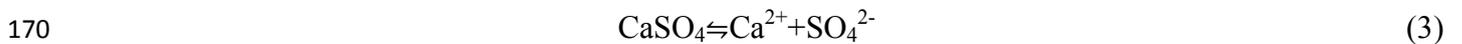
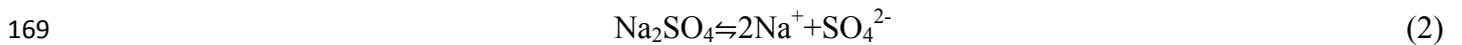
160 R_{Gypsum} : the dissolution rate of gypsum; k_1 : rate constant; A_g : the surface area of gypsum; V : the
 161 liquid volume in contact with the gypsum surface; IAP : the product of ion activity; and K : ion solubility
 162 product.

163 $\left(\frac{IAP}{K}\right)_{Gypsum}$ is affected by the temperature; thus, it is the same as R_{Gypsum} .

164 The solubility of gypsum in water reaches a maximum of 0.2097 g/100 g at 40°C. The solubility
 165 decreases when the temperature is below or above 40°C. The content of SO_4^{2-} and Ca^{2+} obtained by
 166 physical dissolution is very low.

167 *3.2.2. Gypsum ($CaSO_4$) dissolved by glauber salt brine (Na_2SO_4)*

168 Equations (2) and (3) show the reactions of Na_2SO_4 and $CaSO_4$ with water.

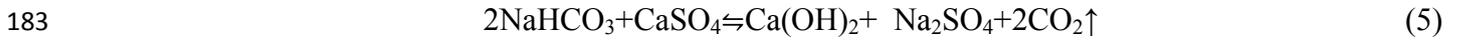


171 Because of the common-ion effect, the solubility of the electrolyte will decrease when a strong
 172 electrolyte with the same ion is placed into an electrolyte-saturated solution. Thus, the solubility of
 173 gypsum will be reduced when glauber salt brine flows through and dissolves the gypsum deposits; the
 174 gypsum will be even harder to dissolve in this situation. Thus, if the glauber salt brine flows through the
 175 gypsum deposits, the brine characteristic would not apparently change.

176 *3.2.3. The reaction of trona brine or a mixture of trona and glauber salt brine with gypsum*

177 The HCO_3^- and CO_3^{2-} contents in trona brine or in mixed brine are very high as is the solution
 178 alkalinity and pH. If the reaction kinetics is not taken into account, the pH has little influence on the
 179 dissolution of gypsum (Yang, 2003; Xu and Li, 2011). The reaction occurs when the brine with high
 180 concentrations of HCO_3^- and CO_3^{2-} flows through the gypsum deposits. The main chemical reactions are

181 as follows:



184 In Eq. (4), CaSO_4 is slightly soluble, while CaCO_3 is insoluble. The reaction easily occurs when an
185 insoluble substance is produced by a slight soluble substance, and the ionic equation is as follows:



187 The Gibbs Free Energy (ΔG) is -22.7 kJ/mol under the standard state. When ΔG is negative, the
188 reaction, which is endothermic, occurs freely. The reaction is faster at higher temperatures. Eq. (5)
189 shows that ΔG is 2102 kJ/mol under the standard state. When ΔG is positive, the reaction will not freely
190 occur.

191 Thus, the reaction shown in Eq. (5) will not occur, but the chemical reaction will still proceed as
192 shown in Eq. (4), when trona brine or mixed brine flow through the gypsum deposits.

193 3.2.4. The carbonate equilibrium effect during the reaction of different brines

194 The carbonate equilibrium that exists in the trona brine or mixed brine is affected by pH. The
195 carbonate in groundwater exists in three forms: free carbonic acid, bicarbonate and carbonic acid.

196 In the trona brine (pH>10), the concentration of HCO_3^- is 5-20 times that of the ~~concentration of~~
197 CO_3^{2-} concentration, and CO_3^{2-} in the brine is dominant in this case. When the trona brine flows through
198 the gypsum, CaSO_4 reacts with CO_3^{2-} and CaCO_3 precipitates. If the concentration of CO_3^{2-} in the brine
199 decreases, a reversible reaction will take place and drive the equilibrium to the right. Thus, the reverse
200 reaction will occur when the trona brine flows through the gypsum as follows:



203 The circular reactions as shown in Eqs. (7) and (8) will occur when mixed brine flows through the

204 gypsum because it has similar properties to the trona brine. Thus, taking the carbonate equilibrium
205 effect into account, the concentrations of HCO_3^- and CO_3^{2-} will decrease, while SO_4^{2-} increases after
206 CaCO_3 precipitates.

207 *3.3. Simulation of groundwater inrush source*

208 For further quantitative analysis of the inrush water source and component, the international
209 hydrological and geochemical simulation software PHREEQC was used to simulate the water-rock
210 interaction. The PHREEQC software was developed by the U.S. Geological Survey, and it is able to
211 calculate geochemical action within a temperature range of from 0~300 degrees (Wei, 2010).

212 Based on the deduction that the main water inrush source around Anpeng was trona leakage brine,
213 the simulation method PHREEQC was used and combined with the possible channel of inrush water to
214 establish a conceptual model and then hydrogeochemical simulation of the water-rock interaction was
215 conducted. Subsequently, the mixed ratio of inrush groundwater and shallow groundwater around
216 Anpeng were quantified, which can better verify the source of the inrush water.

217 *3.3.1. Conceptual model*

218 Around Anpeng, the trona leakage brine flowed through the specified mineral assemblages and
219 mixed with shallow groundwater in different proportions.

220 *3.3.2. Initial data input*

221 The parameters of the trona brine were taken from the enterprise's production testing data; the
222 parameters of the shallow groundwater were taken from the same aquifer but outside the study area, and
223 can basically represent groundwater background values. The specific parameters are shown in Table 1.

224 *3.3.3. Setting of stratum and mineral*

225 The formations from the bottom to the top during the process of the leakage brine flowing into the
226 shallow groundwater and then flowing out of the ground were as follows: the third member of the

227 Hetaoyuan Formation of Paleogene, ~~and~~ the Liaozhuang Formation and the Fenghuang Formation of
228 Neogene and Quaternary, ~~respectively~~. To simplify the mining area, according to the thickness of the
229 rock stratum and the proportion of mineral composition, it can be assumed that the layer through which
230 the trona brine flowed contains Ca-montmorillonite, kaolinite, gypsum, potash feldspar and potash
231 mica.

232 The main ingredients are as follows: Kaolinite: $Al_4[Si_4O_{10}](OH)_8$; Gypsum: $CaSO_4 \cdot 2H_2O$;
233 Ca-montmorillonite: $(Na,Ca)_{0.33}(Al,Mg)_2[Si_4O_{10}](OH)_2 \cdot nH_2O$; Dolomite: $CaMg(CO_3)_2$; Potash feldspar:
234 $K [AlSi_3O_8]$; Potash mica: aluminium silicate as K, Al, Mg, Fe and Li.

235 **4. Results and Discussion**

236 On 9 March 2013, in Anpeng, water samples from five groundwater inrush points and six
237 surrounding water quality monitoring points (resident well) were tested. The results of water chemical
238 composition are shown in Table 2, and the distribution of the sampling points is shown in Fig. 4.

239 According to the water quality analysis, the inrush brine had a relatively high salinity, with some
240 inrush water samples containing SO_4-Na and some containing HCO_3-Na . The crystals mainly consisted
241 of $NaSO_4$, Na_2CO_3 , and $NaHCO_3$. The composition of the inrush water and the crystals was the same as
242 that of the high-concentrated ions in the trona brine (Na_2CO_3 , $NaHCO_3$, etc.) and glauber salt brine
243 (Na_2SO_4).

244 **4.1. The source of the inrush water**

245 An automatic water quality recorder was set up at the Y5 inrush point on 4 March 2013. The
246 monitoring lasted from 5 March to 20 March 2013. Thus, the relationship between the inrush points and
247 the S02 well can be assessed according to the correlation of the changes between temperature/electrical
248 conductivity and the concentration of brine during the S02 production well reparation period (5 March
249 to 14 March 2013).

250 The production of glauber ceased during the investigation (2 March to 15 March 2013), so it could
251 be determined how the glauber mining affects the water inrush hazard based on a dynamic water quality
252 situation.

253 4.1.1. The source of inrush water at the Y-5 point

254 After successful reparation of the S02 well, the conductivity and temperature of the inrush water
255 decreased significantly. The CO_3^{2-} concentration remained at 0, the ~~concentration of~~ HCO_3^-
256 concentration decreased to 500 meq/L, while the ~~concentration of~~ SO_4^{2-} concentration increased to 600
257 meq/L. Subsequently, the concentrations of these three ions were in a state of dynamic balance. The
258 analysis shows that the source of the inrush water at the Y-5 point is closely related to the S02 trona
259 well.

260 In order to ensure whether the glauber brine exists at this point as part of an inrush source, further
261 analysis was performed. The depth of the well rupture was 234 m; the gypsum deposit was developed to
262 this depth. While the leakage of the trona brine flowed through the gypsum deposit, reactions would
263 occur as shown in Eqs. (7) and (8).

264 According to the ion milliequivalent concentrations (Ca^{2+} : 0.61 meq/L; CO_3^{2-} : 905.3 meq/L; HCO_3^- :
265 1332.94 meq/L; Cl^- : 107.43 meq/L; and SO_4^{2-} : 267.89 meq/L) at the Y-5 point, the concentration of Ca^{2+}
266 was negligible compared to the other main ions. Only the reaction between CO_3^{2-} and CaSO_4 had to be
267 taken into account because of the large number of CO_3^{2-} , with fast velocity, the short contact time with
268 gypsum, and the high temperature. The reaction of CO_3^{2-} and CaSO_4 would take place at a ratio of 1:1
269 according to Eq. (7), and three types of inrush water sources could be assumed under this precondition
270 as follows:

271 (1) The inrush water source was only from the trona brine.

272 The CO_3^{2-} and CaSO_4 in the brine reacted at a ratio of 1:1, and the ~~concentration of~~ SO_4^{2-}

273 concentration was equal to the reacted γCO_3^{2-} content. Thus, the $\gamma\text{CO}_3^{2-}/\gamma\text{HCO}_3^-$ ratio in the trona brine
274 was equal to the $\gamma(\text{CO}_3^{2-}+\text{SO}_4^{2-})/\gamma\text{HCO}_3^-$ ratio in the inrush water. From this calculation, it could be seen
275 that $\gamma(\text{CO}_3^{2-}+\text{SO}_4^{2-})/\gamma\text{HCO}_3^-$ was equal to 0.88, while $\gamma\text{CO}_3^{2-}/\gamma\text{HCO}_3^-$ ranged between 0.86 and 1.26. The
276 content of $\gamma(\text{CO}_3^{2-}+\text{SO}_4^{2-})/\gamma\text{HCO}_3^-$ was similar to $\gamma\text{CO}_3^{2-}/\gamma\text{HCO}_3^-$; therefore, the source of the inrush
277 water was exclusively trona brine.

278 (2) The inrush water source was only from the glauber brine.

279 The $\gamma\text{SO}_4^{2-}/\gamma\text{HCO}_3^-$ ratio in the glauber brine was equal to 1237.8, compared to 0.19 in the inrush
280 water. Therefore, this assumption was incorrect because of the widely varying ratios.

281 (3) The inrush water source was from a mixed brine of glauber and trona.

282 Assuming that the contribution ratio of the glauber brine was X and that of the trona brine was Y
283 ~~was true~~, then $1237.8 \times X + (0.86 \sim 1.26) \times Y = 0.88$. This equation showed that when the contribution
284 ratio of the trona brine was equal to 1, the contribution ratio of the glauber brine was equal to 1.6×10^{-5}
285 ~~and which is was~~ too small ~~to~~ that can be ignored.

286 Thus, it could be confirmed that the water inrush source at Y-5 was exclusively the leakage of trona
287 brine from the broken S02 well.

288 4.1.2. The sources of inrush water at the Y-4, Y-3, Y-2, and Y-1 points

289 The inrush water quantity and the dynamic variation of the concentration of SO_4^{2-} and HCO_3^- at
290 points Y1-Y4 were not obvious when the S02 well was under repair and all the glauber wells were shut
291 down (~~2-15~~ from 2 to 15 March). This result shows that the sources of these water inrush points were
292 not due to the underground mining activities of the glauber brine or the rupture of the S02 well, but
293 because of the brine leakage from other trona wells.

294 4.1.3. Components and mixed proportions of the inrush water

295 The PHREEQC simulation conditions were assumed to be as follows: (1) the trona brine did not

296 mix with shallow groundwater after flowing through the mineral layer; or (2) the trona brine mixed with
297 shallow groundwater in a ratio of 1:2, 1:10, 1:100, 1:200, 1:500, 1:1000 and 1:5000 after flowing
298 through the mineral layer. The simulation results are shown in Table 3.

299 Table 3 shows that when the trona brine flowed through the stratum and shallow groundwater, the
300 concentrations of Na^+ , Cl^- and SO_4^{2-} decreased while the ~~concentration of~~ HCO_3^- concentration
301 increased with increasing proportion of ~~the~~ shallow groundwater. The ~~concentration of~~ Ca^{2+}
302 concentration decreased at first and then increased.

303 The ion concentrations at Y-5, except for SO_4^{2-} , were similar to the ion concentrations in the trona
304 brine. However, at the same time, the ~~concentration of~~ HCO_3^- concentration was nearly 0 meq/L. When
305 the trona brine flowed through the layer, it would react rapidly and pour out of the ground directly
306 because of the fast velocity of the inrush water at Y-5. Meanwhile, the trona brine was not continuously
307 provided in the simulation. Thus, the concentration of HCO_3^- would be near to the concentration of
308 trona brine in reality. Therefore, the trona brine must have a rapid inrush, ~~nearly~~ almost not mixing with
309 shallow groundwater.

310 The PHREEQC simulation ~~analysis~~ results show that: 1) the water inrush source of Y-5 was ~~nearly~~
311 ~~all of~~ the trona brine almost all from the ruptured S02 well; 2) the water inrush source of Y-3 was a
312 mixture of trona brine and groundwater in a ratio of ~~from~~ 1:10~1:100; and 3) the water inrush sources of
313 Y-4, Y-2 and Y-1 were a mixture of trona brine and groundwater under the ratio of 1:200.

314 **4.2. The channel of the inrush water**

315 *4.2.1. Reasons for the brine leakage*

316 Trona is produced by either a single well or double/multiple well convection mining method, ~~a~~ that
317 is water-soluble mining method (Lin, 1987). The main mining unit consists of a salt cavity and
318 production well. Thus, the instability of the salt cavity and the rupture of the production well are the

319 main possible reasons for brine leakage.

320 (1) Analysis of salt cavity stability

321 The possibility of salt cavity collapse: Trona is distributed at the bottom of the second member of
322 the Hetaoyuan Formation and in the upper part of the first member of the Hetaoyuan Formation, with
323 dolomite strata developed in the roof and floor. The thick and hard surrounding rock structure
324 determined that the cavity is produced by hydrofracture but it is hard to fill with large-scale fractured
325 channels and can remain intact and stable.

326 The development of a roof fracture: When a mineral is under exploitation, the surrounding rock in
327 the cavity is under pressure from the inner brine. This pressure is equal to the sum of the water injection
328 pressure ~~plus and~~ the water column pressure in the production well. The water injection pressure of the
329 trona production well is approximately 10-20 MPa, while the 1560.92-2929.53 m (mineral buried depth)
330 water column pressure is approximately 15.3-28.71 MPa. Thus, the greatest water pressure on the
331 surrounding rock in the cavity is 48.71 MPa. The main lithology of the surrounding rock is dolomite
332 ~~which that~~ is 500 m in thickness and 142.66 MPa in compressive strength, which is nearly 3 times that
333 of the greatest possible water pressure. Therefore, large-scale fractures in the surrounding rock of the
334 trona mineral would be difficult to develop under the effect of sustained water pressure.

335 (2) Analysis of production well rupture

336 The phenomenon of brine leakage caused by the S02 well rupture in Anpeng indicates that
337 production well damage is an important cause of brine leakage. The depth of the S02 well rupture is 234
338 m underground, i.e. in the gypsum deposit, which is strongly hygroscopic. The pressure caused by the
339 water swelling is approximately 0.15 MPa (Li and Zhou, 1996), which may damage the production well
340 and induce brine leakage. The high concentration of SO_4^{2-} (>250 mg/L) generated by the reaction of
341 leakage brine and gypsum can also corrode the production well and lead to groundwater inrush.

342 4.2.2. Analysis of water-conducting channel

343 According to our analysis, the most probable reason for brine leakage in trona is production well
344 rupture. The leaking brine will flow along the water-conducting channel into the shallow aquifer and
345 even pour out of the ground. However, the geological structure in the mining area shows no
346 water-conducting fault development. Thus, the water-conducting channel, that the leakage brine flows
347 along, is probably a fissure or artificial channel.

348 ~~A~~sStructural fissure is the main type of fissure that occurs in groundwater inrush hazards when
349 using the solution mining method. The structural fissure is determined by the maximum horizontal
350 principal stress, which is controlled by the tectonic stress field in the mining area. The connection
351 direction of the S02 well and the other water inrush points is NW-SE, which is the same as that of the
352 structural fissure zone development direction. This [result](#) indicates that the main water-conducting
353 channel in Anpeng is controlled by the structural fissure zone.

354 The inrush points in Anpeng are all at the abandoned gypsum exploitation wells, which were not
355 closed properly. Thus, high-pressure cavity water or leakage brine can flow along the structural fissure
356 zone, finally connect with these wells, and then pour out of the ground through boreholes. Therefore,
357 the abandoned gypsum exploitation wells are the main channels through which the shallow polluted
358 groundwater flowed out of the ground, as shown in Fig. 5.

359 5. Conclusions

360 This study aimed to investigate the source and channel of the inrush water in a multilayer rock salt
361 mining area. To achieve the set objectives, ~~this study combined~~ an analysis of geological and
362 hydrogeological conditions, an analysis of physical and chemical reaction principles of different brines,
363 the PHREEQC simulation method, and an analysis of geological and artificial reasons for the
364 conducting channel where leakage brine flowed from the damage depth out to the ground [were](#)

365 [combined](#) as the study methodology.

366 Long-term solution mining with high-pressure and -temperature water not only dissolves minerals
367 but also may cause rupture of strata and damage of the production well, which usually results in brine
368 leakage or ~~the inrush of~~ groundwater [inrush](#). Geological and hydrogeological conditions are the basis
369 which determines the total risk of the groundwater inrush hazard. Physical and chemical reaction
370 principle analysis of different brines and hydrogeochemical simulation of water-rock interaction in
371 different assumed conditions using the PHREEQC simulation method can not only determine the exact
372 source of the leakage brine but also identify the mixed proportion of inrush water while the leakage
373 brine flows through the mineral layer in different way. Other than geological reasons, mining techniques
374 such as pressure control of injection water and groundwater quality monitoring of exploitation wells
375 may also determine the risk of a groundwater inrush hazard in a multilayer rock salt mining area.

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379 **Author Contributions**

380 Bin Zeng and Tingting Shi contributed to data analysis and manuscript writing; Zhihua Chen
381 proposed the main structure of this study; Liu Xiang and Muyi Yang designed and performed the
382 experiments; and Shaopeng Xiang performed the PHREEQC simulation. All the authors read and
383 approved the final manuscript.

384 **Conflicts of Interest**

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

386

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Figure captions

Fig. 1. One of the long-term (longer than 2 years) groundwater inrush points with stable discharge (Y-3).

Fig. 2. The sudden groundwater inrush point (Y-5). ~~As shown in this figure,~~ [The](#) high-temperature inrush groundwater was being pumped after the ground was broken.

Fig. 3. Information about strata, lithology, aquifers, and buried positions of each ore bed in the mining area.

Fig. 4. Sketch map of hydrogeological conditions and the distribution of groundwater inrush points in the mining area.

Fig. 5. Schematic diagram of [the](#) source and channel ~~analysis~~ of the groundwater inrush hazard in the multilayered rock salt mining area in Tongbai County.



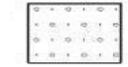
Fig.1



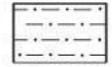
Fig.2

Stratigraphy				Thickness (m)	Lithologic profile	Petrographic description	Minerals	Aquifer
System	Series	Formation	Member					
Quaternary				0-290		Alternating layers of sandy conglomerate and sandy clay		Shallow aquifer
Neogene	Oligocene	Fenghuang zhen						
Paleogene	Eocene	Liaozhuang		500-634		Upper part: mudstones are interbedded with gypsum	Gypsum	Weak permeable stratum
		Hetaoyuan	Third segment	400-500		Mudstone with interlayers of sandy conglomerate, as well as thin layers of shale, muddy dolomite and glauber salt	Glauber	
			Second segment	700-800		Mudstone is interlayered with muddy dolomite and dolomite, as well as small amounts of trona		Trona
First segment	1100-1700		Mudstone, muddy dolomite, dolomite, shale and siltstone	Trona				

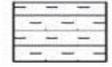
Legend



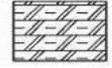
Sandy conglomerate



Sandy clay



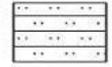
Mudstone



Muddy



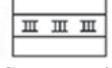
Shale



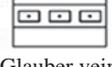
Siltstone



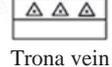
Dolomite



Gypsum vein

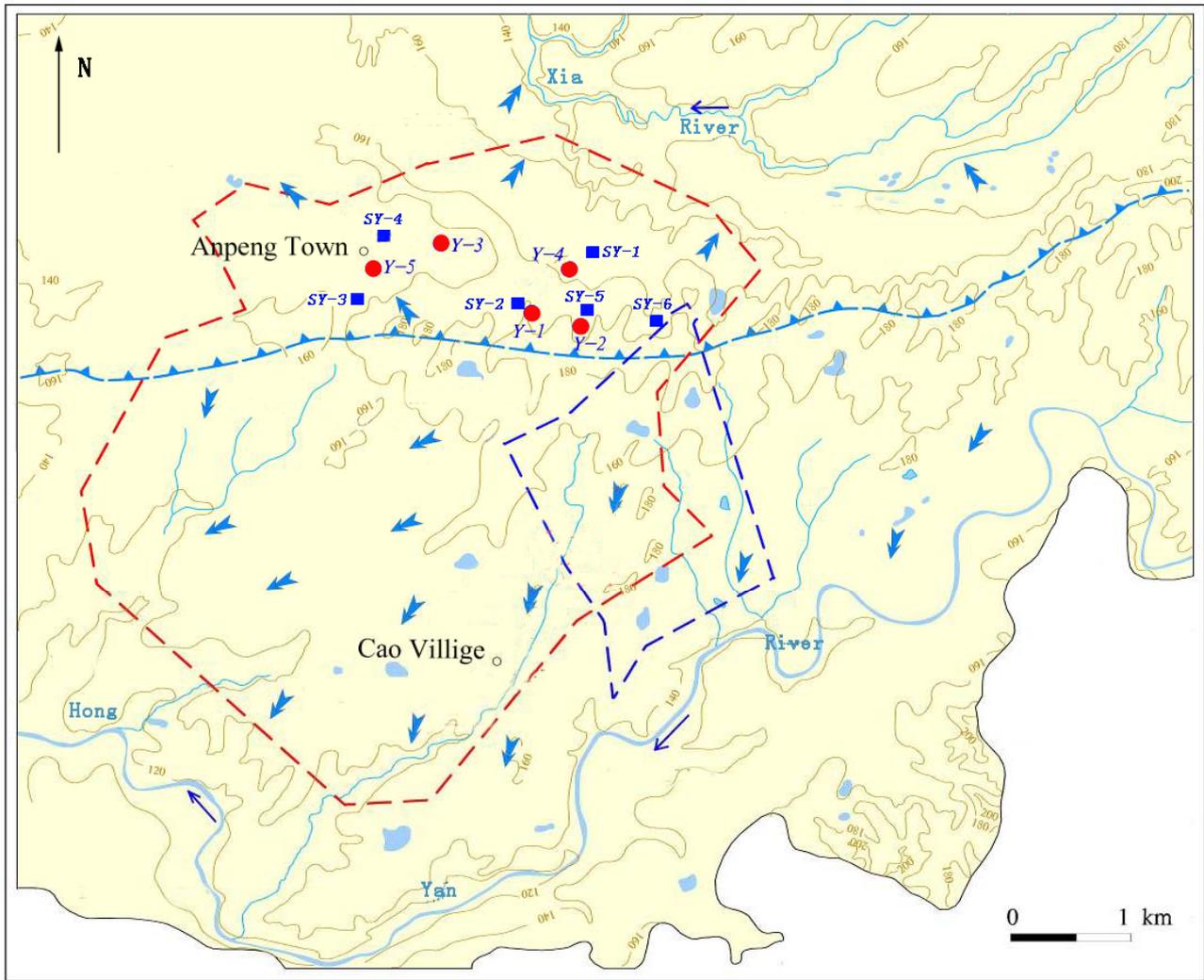


Glauber vein



Trona vein

Fig.3



- | | | |
|----------------------------|--------------------------------|-------------------------------|
| Quaternary pore water | The area of trona mine | The area of glauber salt mine |
| Contour and elevation | Drainage divide of groundwater | Rivers and lakes |
| Groundwater flow direction | Groundwater inrush points | Resident well points |

Fig.4

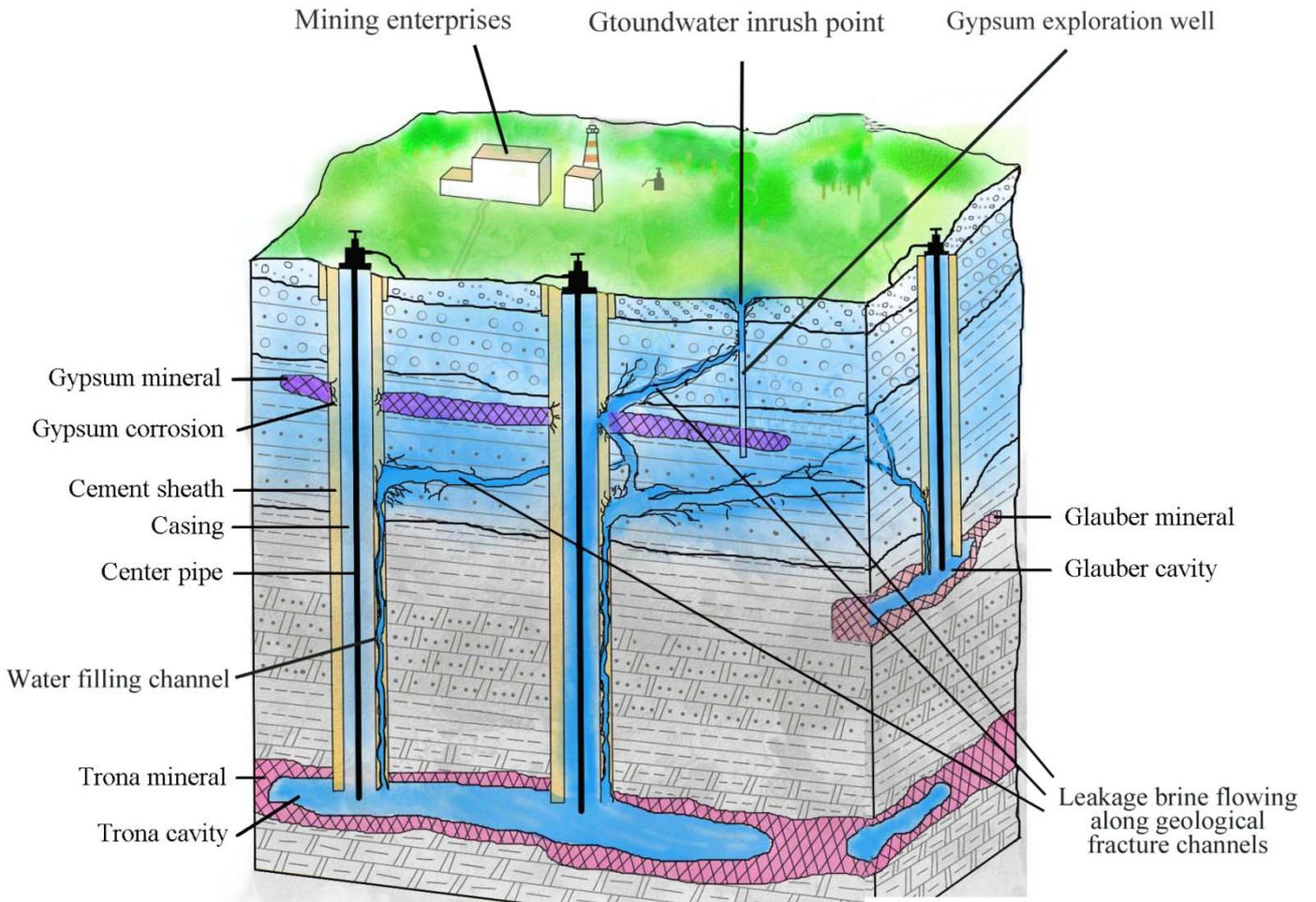


Fig.5

Table 1 Initial data of trona brine and background value of groundwater for the PHREEQC simulation

Type	Temperature (°C)	pH	Na ⁺	Ca ²⁺	Mg ²⁺	Cl ⁻	SO ₄ ²⁻	HCO ₃ ⁻	CO ₃ ²⁻
						(mg/L)			
Trona brine	70.00	10.80	85880.00	5.00	1.00	3819.00	206.00	104721.00	4565.00
Background value of groundwater	14.10	7.50	38.76	67.10	23.88	12.46	39.31	386.87	0.00

Table 2 Chemical composition of groundwater from the inrush hazard points and surrounding resident wells

Source	Point	Na ⁺	Ca ²⁺	Mg ²⁺	Cl ⁻	SO ₄ ²⁻	HCO ₃ ⁻	CO ₃ ²⁻	Salinity	Depth (m)
		(mg/L)								
Groundwater from inrush hazard points	Y-1	447.30	91.20	74.68	171.18	278.55	1488.89	0.00	1807.35	330.55 ~ 430.20
	Y-2	524.50	89.34	75.32	153.97	298.88	1525.00	0.00	1904.51	
	Y-3	1132.00	146.60	158.30	125.56	4296.44	1012.93	0.00	6365.37	
	Y-4	322.12	98.67	123.88	210.78	346.55	1122.77	0.00	1663.38	
	Y-5	50300.00	12.23	53.21	3813.80	12858.63	81309.15	27159.00	107692.40	
Groundwater from resident wells around the inrush points	SY-1	46.28	76.76	17.29	64.30	14.58	319.03	0.00	378.73	10.00
	SY-2	28.37	98.02	27.46	26.16	10.38	453.84	0.00	417.31	
	SY-3	43.14	46.20	14.42	31.02	117.12	319.03	0.00	316.26	
	SY-4	118.53	278.40	72.30	425.23	175.96	568.52	0.00	1354.68	
	SY-5	31.67	95.51	19.22	53.93	22.59	351.97	0.00	398.90	
	SY-6	36.77	68.82	19.60	18.51	21.55	340.38	0.00	335.43	

Table 3 Simulation results for a mixed proportion of inrush trona brine using the PHREEQC

Conditions	Mixed proportion with shallow groundwater	method (mg/L)				
		Na ⁺	Ca ²⁺	Cl ⁻ (mg/L)	SO ₄ ²⁻	HCO ₃ ⁻
Trona brine unmixed or mixed with different proportion of shallow groundwater after flowing through the mineral layer (simulation results)	Unmixing	87147.00	301.08	3880.15	68659.20	5.06
	1:1	48093.00	280.00	2145.62	37900.80	9.39
	1:2	33235.00	184.72	1485.68	26188.80	13.97
	1:10	9586.40	148.28	436.30	7561.92	57.95
	1:100	1098.25	90.40	141.63	873.89	306.34
	1:200	571.78	69.60	118.56	459.17	382.17
	1:500	252.77	68.32	104.60	207.84	453.66
Water quality test results in five water inrush hazard points	1:1000	144.81	67.52	99.94	105.12	481.60
	Y-1	447.30	91.20	171.18	276.55	1488.89
	Y-2	524.50	89.34	153.97	298.88	1525.00
	Y-3	1132.00	146.60	125.56	4296.44	1012.93
	Y-4	322.12	98.67	210.78	346.55	1122.77
	Y-5	50300.00	12.23	3813.80	12858.63	81309.15

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