GROUNDWATER CHEMICAL SIGNATURES – POSSIBLE CLUES FOR THE SEEMINGLY ERRATIC PRODUCTIVITY OF A GEOTHERMAL AQUIFER HOSTED BY A ~1 KM THICK CARBONATE SEQUENCE IN THE MOESIAN PLATFORM (ROMANIA)

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ABSTRACT

A several hundred meters thick carbonate reservoir located in the Moesian Platform (Romania) has been tapped by deep wells (2000-3000 m) drilled for geothermal exploration. Because of the carbonate reservoir progressive deepening – from its outcrop area, down to more than 3 km – the temperature range of the stored groundwater spans over more than 50°C. Thermal waters discharged by boreholes whose pay-zones occurred within regions of correspondingly contrasting temperatures displayed Ca^{2+} and HCO_3^- concentrations which were reversely correlated with the inflow temperatures. Since calcite ($CaCO_3$) has the distinctive property of becoming increasingly soluble as the aqueous solution cools down ("retrograde solubility"), the noticed reverse correlation could indicate: either calcite dissolution along upflowing plumes of deeply-originating groundwater subject to progressive cooling; or calcite deposition along dowflowing plumes of meteoric recharge water that experienced progressive heating. The coexistence of such adjacent, "warm" and "cold" plumes – each of them several kilometres wide – is in accordance with the overall temperature distribution within the aquifer.

That model could explain as well why the geothermal aquifer displayed, over short lateral distances (a few kilometres), productivity variations of more than one order of magnitude. Where reflection seismic records were available, significant lateral variations were also noticed to be displayed by the velocity of the seismic waves which propagated across the thick carbonate stack: such variations appeared to be correlated with the aquifer productivity – namely, increased seismic velocities corresponded to poor productivity, and vice-versa. The low-productivity, compact zones could be ascribed to pore clogging in response to calcite precipitation, as the meteoric water recharge – supplied from the reservoir outcrop area – became progressively heated while descending deep into the aquifer; while the less compact zones could be due to calcite dissolution, ensuing to cooling of the ascending, deeply-originating groundwater.

Keywords: Carbonate Reservoir, Retrograde Solubility, Convective Heat Transfer, ReflectionSeismic Survey, Interval Velocity, Geothermal

INTRODUCTION

A calcite characteristic designated as “retrograde solubility” is a key factor in carbonate reservoirs diagenesis, and, implicitly, for the extractability of stored fluids. It refers to the fact that as the temperature of an aqueous solution increases, the latter tends to become supersaturated in calcite, which eventually precipitates within the rock pores and fractures; conversely, a cooling solution becomes increasingly aggressive toward the carbonate host rock, resulting in porosity enhancements.

The possibility that such processes developed within an aquifer stored in the Late Jurassic-Early Cretaceous carbonate rock-stack which extends beneath the Moesian Platform (Romania) has been addressed in the present study. There was considered to this purpose the chemical composition (Ţenu et al., 1994) and the temperature (Danchiv et al., 1992; Mitrofan & Tudor, 1992; Crăciun et al., 2000; Niculae et al., 2019) of thermal water sampled from several deep (~3000 m) wells drilled for geothermal exploration.

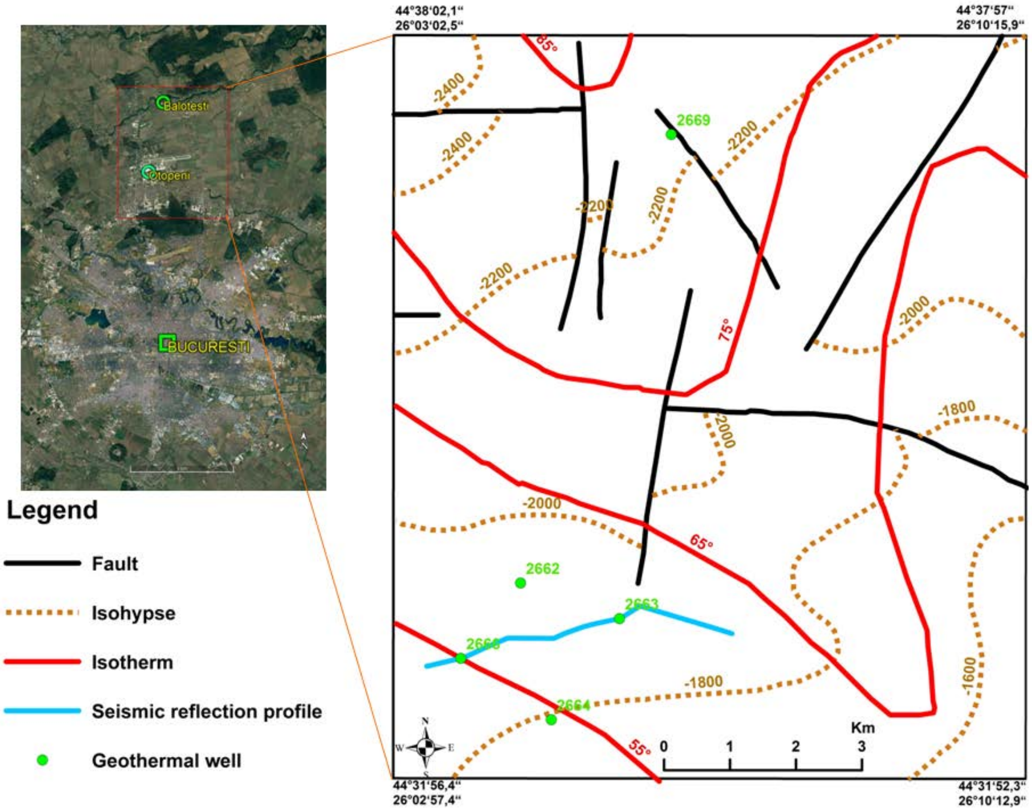

Figure 1 – Contour map at the top of the J_3-K_1 carbonate reservoir (isohypses with respect to sea level – modified from Crăciun et al., 2000), superimposed on a map (modified from Niculae et al., 2019) of temperature distribution at the same geological boundary. The represented domain is indicated by the rectangle within the Google Earth index map

THERMO-CHEMICAL SIGNATURES DISPLAYED BY THE AQUIFER

The concerned aquifer extends laterally over hundreds of kilometers, having a thickness of at least several hundred meters, possibly exceeding even 1 km. The carbonate reservoir dips gently, so that from its outcrop area, it eventually reaches a known depth in excess of 3 km.

If the carbonate reservoir was subject to a laterally uniform basal heat flow, and if thermal conductivity values were similar, one would expect a gradual increase of the temperature at the top of the aquifer, as the latter deepened beneath the overlying impervious formations. Obvious departures from such a thermal regime are however recorded: (i) between various domains displaying the same depth of the aquifer top, the latter exhibits (Figure 1) highly contrasting temperatures; (ii) over its entire thickness, of almost 1000 m, the aquifer is subject to a virtually isothermal regime (Danchiv et al., 1992; Mitrofan & Tudor, 1992; Crăciun et al., 2000). The possible cause of such distortions of a normal thermal field could be forced convective heat transfer, similar to that modeled by Woodbury & Smith (1985). A corresponding setting implies that plumes of meteoric water flowing down the reservoir dip are laterally juxtaposed – and partly interact – with adjacent plumes of deep-origin water that flows updip the reservoir.

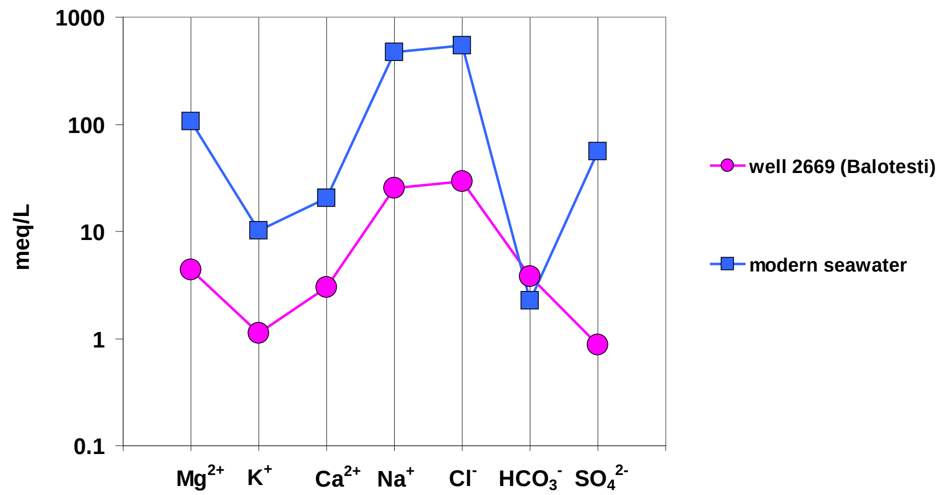

Figure 2 – Schoeller diagram illustrating that the groundwater sample collected from the well 2669 Baloteşti is chemically similar to modern seawater.

One such upflowing plume has likely been intercepted by the well 2669 Balotesti (Figure 1): the saline solution discharged by that borehole was rather concentrated (~2.15 g/L) and its relative contents of Cl^-, Na^+, Ca^{2+}, K^+ and Mg^{2+} were quite similar to those of modern seawater (Figure 2). A significant contribution of marine fluids initially trapped in the basin sediments can therefore be conjectured, such an inference being in accordance also with the fact that within the same carbonate reservoir - although large distances away - hydrocarbon accumulations are encountered (Paraschiv, 1979; Anastasiu et al., 2002).

Alternatively, it is probable that the wells 2662 Otopeni, and especially 2664 Otopeni (Figure 1), reflect progressive interaction with groundwater plumes having a larger fraction of meteoric water, as indicated by the gradual reduction of the Cl^- and Na^+ concentrations (Figure 3).

At the same time, the temperatures measured in each of the three considered wells at the top of the carbonate reservoir progressively decrease from Baloteşti toward Otopeni (Figure 4), i.e., along a NNE-SSW direction (Figure 1). The correspondingly recorded temperature contrasts are far larger than those to be expected by assuming a strictly conductive heat transfer, subject to reasonable values of the geothermal gradient (Figure 4). Hence, also this temperature distribution pattern likely mirrors a gradually increasing contribution of cold freshwater toward the well 2664 Otopeni - an inference that is as well substantiated by Cl^- and Na^+ concentrations diminishing along with the reduction of the temperature recorded the top of the carbonate reservoir (Figure 5).

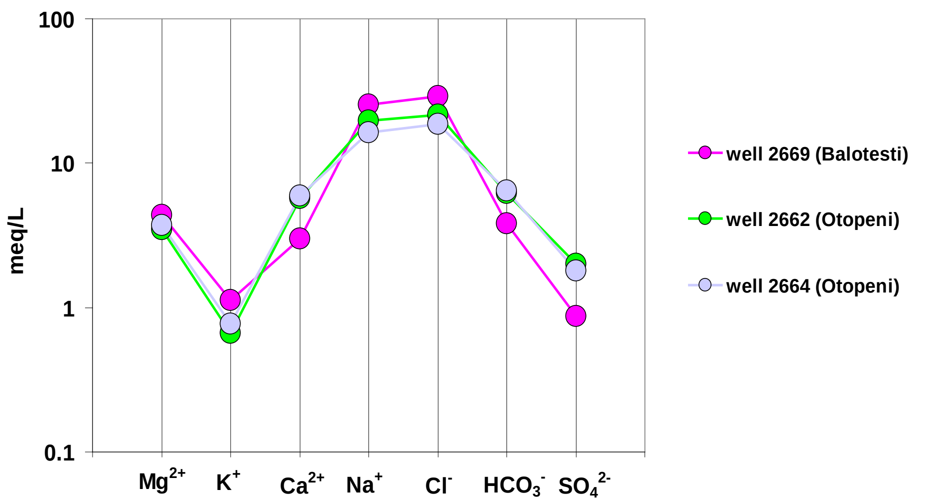

Figure 3 – Schoeller diagram constructed for groundwater samples collected from wells tapping the aquifer hosted by the J_3-K_1 carbonate reservoir.

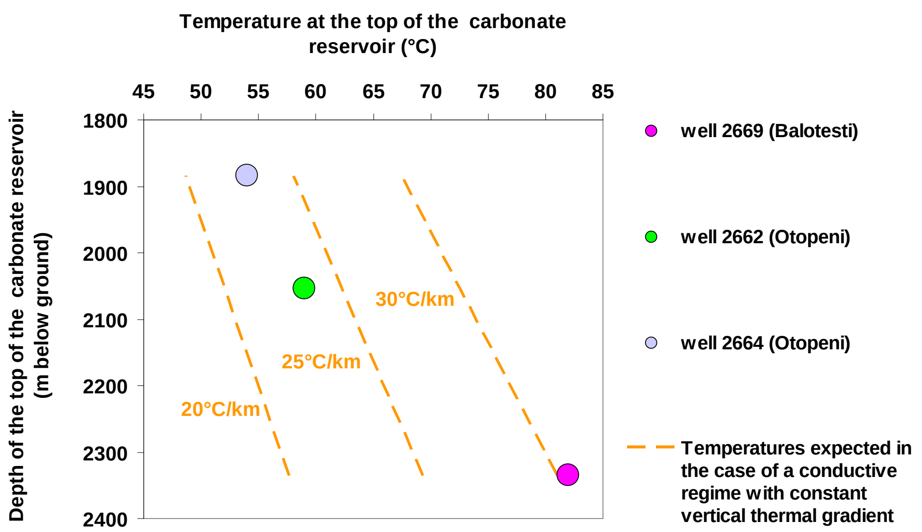

Figure 4 – Temperatures recorded at the top of the J_3-K_1 carbonate reservoir in the three considered wells. Under a strictly conductive thermal regime and subject to a lateral uniformity of the heat flow and of the thermal conductivities, the concerned temperature values should have plotted approximately on a common linear profile - similar to those

illustrated by each of the dashed lines (where the labels indicate the geothermal gradient which was used for constructing each corresponding hypothetical profile).

It is on the other hand noticeable that as the aqueous solution becomes more dilute (as indicated by the progressive lowering of its Cl^- contents, Figure 6), the corresponding Ca^{2+} and HCO_3^- concentrations are steadily increasing. This instance likely indicates interaction with the host rock, implying that each of the two following complementary processes could, separately, operate: (i) the aquifer carbonate host rock was dissolved by upflowing plumes of deep origin groundwater that experienced progressive cooling (while being, in addition, diluted by freshwater); (ii) calcite was deposited from meteoric recharge water, which advanced as dowflowing plumes and, accordingly, underwent progressive heating (while being concomitantly "contaminated" with connate water). The accordingly conjectured behaviour is expected to occur as a consequence of the calcite specific property of becoming increasingly less soluble, as the aqueous solution heats up ("retrograde solubility" – e.g., Andre & Rajaram, 2005). Such an inference is consistent with the instance that the Ca^{2+} and HCO_3^- concentrations of the water samples collected from the considered wells are reversely correlated (Figure 7) with the corresponding temperatures recorded at the reservoir top.

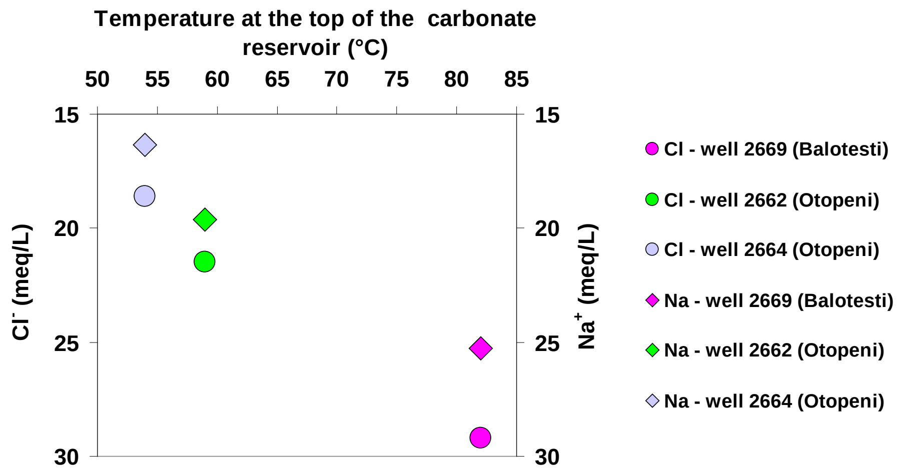

Figure 5 – Cl^- and Na^+ concentrations in thermal water sampled from the indicated wells, plotted against temperatures recorded in the same wells at the top of the J_3-K_1 carbonate reservoir (notice that vertical scales have a downward-increasing values-order).

IMPLICATIONS FOR RESERVOIR PRODUCTIVITY

Most likely, the so far illustrated results (Figures 3 to 7) mirror processes which develop along an upflowing plume intercepted by the wells 2669, 2662 and 2664. It is on the other hand reasonable to expect that within downflowing plumes – which convey meteoric recharge water toward higher temperature regions at depth – the reservoir pores are clogged by calcite precipitation, which results in productivity reductions in those aquifer domains. In particular, poor productivity has been displayed by the well 2666, drilled at Otopeni just 1 km west of the wells 2662 and 2664 (Figure 1). In the latter boreholes, as well as in the nearby well 2663 (located ~2 km further east, Figure 1), bottomhole specific yields of the order of 40 L/(s bar) have been recorded. In contrast, the well 2666 displayed a bottomhole specific yield of only 3 L/(s bar).

It is also worth mentioning that a reflection seismic survey has shown (Mitrofan & Surdulescu, 1990) that the recorded productivity contrasts were correlated with rapid lateral changes (Figure 8) in the interval velocity computed for the ~1 km thick carbonate reservoir: specifically, increased interval velocities corresponded to poor productivity, and vice-versa.

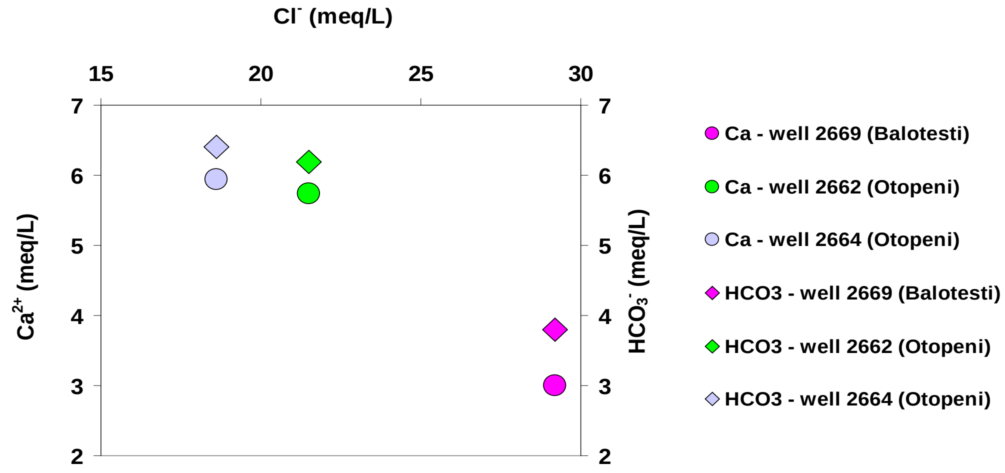

Figure 6 – Ca^{2+} vs. Cl^- and HCO_3^- vs. Cl^- reciprocal concentration plots constructed for thermal water samples collected from the indicated wells.

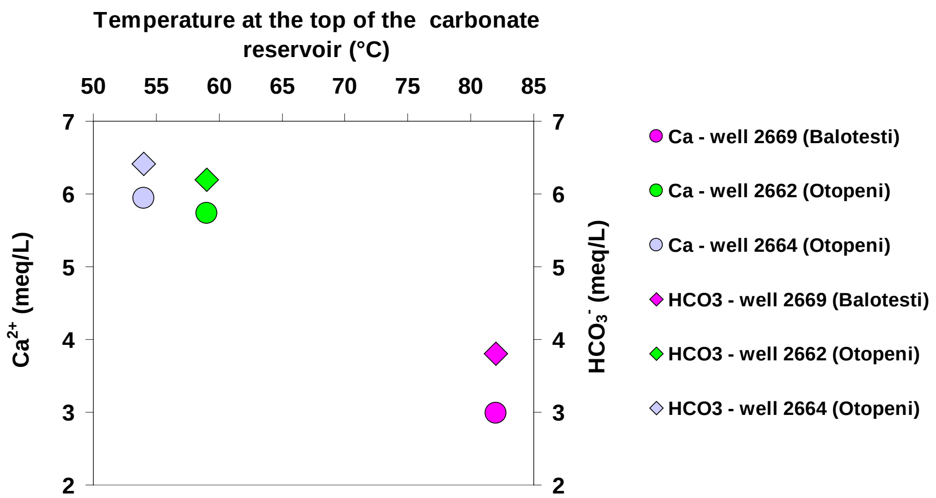

Figure 7 – Ca^{2+} and HCO_3^- concentrations in thermal water sampled from the indicated wells, plotted against temperatures recorded in the same wells at the top of the J_3-K_1 carbonate reservoir.

At the same time, despite the fact that a larger meteoric component was recorded in well 2666 than in the wells 2664 and 2662 (the corresponding Cl^- concentrations being 17.6, as compared to 18.6 and 21.5 meq/L, respectively), the 2666 well sample contents of

Ca^{2+} and HCO_3^- were smaller than the corresponding concentrations recorded in the wells 2664 and 2662: this instance supports the inference that the well 2666 water sample mirrors calcite precipitation occurring within a distinct, downflowing plume, which is adjacent to the upflowing plume intercepted by the wells 2664, 2662 and 2669.

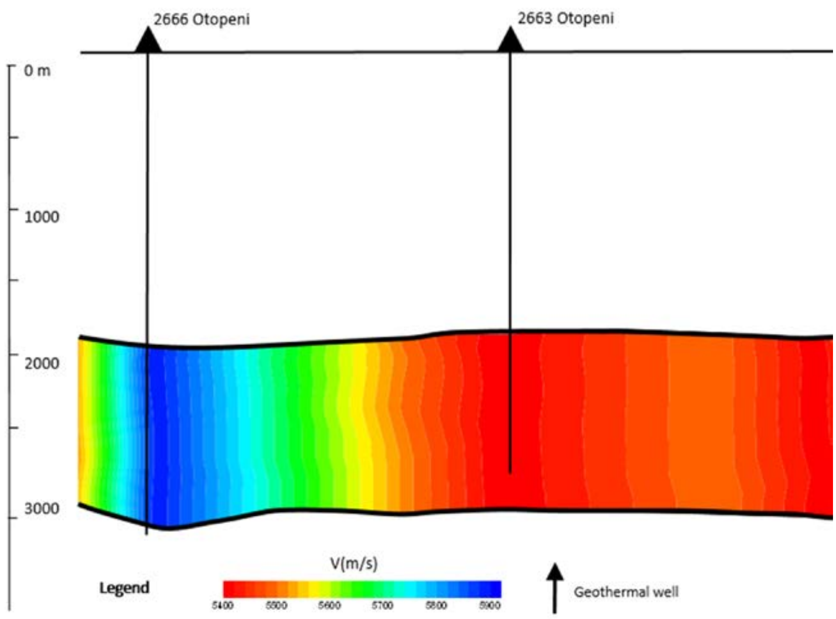

Figure 8 – Cross section illustrating the position and the interval velocities lateral variations which were determined for the carbonate reservoir, based on a seismic reflection survey (modified from Mitrofan & Surdulescu, 1990).

CONCLUSIONS

Thermal groundwater sampled from the aquifer hosted by the Late Jurassic-Early Cretaceous carbonate reservoir of the Moesian Platform (Romania) displays chemical characteristics indicating the contribution of two distinct fluid types: one of deep origin, akin to modern seawater, and the other consisting of meteorically-derived freshwater. The overall temperature distribution within the aquifer suggests that ascending plumes of deep-origin connate water are intercalated with descending plumes of meteoric-origin water. Such adjacent plumes – each of them several kilometres wide – interact in fact with each other, as indicated by the fact that all thermal water samples displayed signatures of both types of fluids.

Because of the progressive deepening of the carbonate reservoir, the temperature range of the stored groundwater spans over more than 50°C. Chemical analyses of waters with such highly contrasting temperatures have indicated Ca^{2+} and HCO_3^- concentrations which gradually decreased toward regions of hotter water. This circumstance could be a result of the calcite so-called “retrograde solubility”, and it accordingly could explain why the exploration for the geothermal resources of the concerned aquifer has outlined large productivity contrasts (in excess of one order of magnitude) between boreholes located just 2-3 km away from one another: the low-productivity, compact zones could be ascribed to pore clogging in response to calcite precipitation, as the meteoric water recharge – supplied from the reservoir outcrop area – became progressively heated while descending deep into the aquifer; while the less compact zones could be due to calcite dissolution, ensuing to cooling of the ascending, deeply-originating groundwater.

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