

1 **Remobilization of Deep Basin Brine during Exhumation of the Illizi Basin, Algeria**

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1 **Abstract**

2 Understanding the processes that drive fluid flow in sedimentary basins has important
3 implications for models of metallic ore and petroleum migration and accumulation in the
4 subsurface. Potential drive mechanisms include gravity-driven flow, compaction-driven flow,
5 and density-driven convection. In this study, we demonstrate that the brine hosted in
6 Ordovician sandstone in the Illizi Basin in Algeria is genetically linked to Triassic-Liassic
7 evaporites deposited > 400 km to the north in the Berkine Basin. This observation confirms
8 that long distance, lateral, brine migration has occurred within the basin in the past. We
9 assess the hydrogeologic record preserved in aqueous fluid inclusions within the Ordovician
10 sandstone, document a marked increase in formation water salinity during cooling and
11 exhumation, and evaluate the drive mechanisms for late-stage remobilization of deep brines
12 within the basin. It is hypothesized that the release of overpressure during exhumation of the
13 Illizi Basin may have been a critical contributor to updip fluid flux. This model could be
14 applicable to other exhumed basins worldwide.

15

1 **1. Introduction**

2 Understanding the processes that drive fluid flow in sedimentary basins has important
3 implications for models of metallic ore and petroleum migration and accumulation in the
4 subsurface. A number of different mechanisms have been proposed for driving regional fluid
5 flow in sedimentary basins (e.g. Bethke, 1985; 1989; Bjørlykke, 1993, 2015; Cathles and
6 Adams, 2005; Chi and Xue, 2011; Frazer et al., 2014): (1) topographic or gravity-driven
7 flow, (2) sedimentary or tectonic compaction-driven flow, and (3) thermal- or salinity-driven
8 convection. In this study, we present new compositional analysis of formation water hosted in
9 Ordovician sandstone in the Illizi Basin in Algeria, and demonstrate that this brine is
10 genetically linked to Triassic-Liassic evaporites deposited in the Berkine Basin to the north
11 (Fig. 1). Building on previous thermal history modelling of the area (English et al., 2016b,
12 2016c), we assess the hydrogeologic record preserved in aqueous fluid inclusions in order to
13 constrain the timing of brine migration into the Ordovician sandstone in the Illizi Basin, and
14 we evaluate overpressure dissipation during exhumation as a potential drive mechanism for
15 late-stage remobilization of deep brines. This model could be applicable to other exhumed
16 basins worldwide and has implications for the timing and direction of major fluxes of deep
17 basin brines in the geological record.

18 19 **2. Geological history of the Illizi Basin**

20 Following the Late Neoproterozoic Pan-African orogeny, the northern margin of
21 Gondwana was characterized by a vast clastic-dominated Paleozoic sedimentary basin on the
22 edge of the ProtoTethys (Beuf et al., 1971; Stampfli and Borel, 2002; Guiraud et al., 2005).
23 During the collision of Gondwana with Laurasia, the Late Carboniferous-Early Permian
24 Hercynian (Variscan) Orogeny caused extensive uplift and erosion along north–south
25 trending arches in northwest Africa (Aliev et al., 1971; Buroillet et al., 1978; Boote et al.,
26 1998; Acheche et al., 2001). Erosion removed the post-Cambro-Ordovician sequence on
27 some of these uplifted arches, but the majority of the Paleozoic sequence was preserved in the
28 Illizi Basin (Galeazzi et al., 2010; English et al., 2016c; Fig. 2). Renewed deposition of the
29 Mesozoic–early Cenozoic “Tethys Supersequence” (Boote et al., 1998) in North Africa
30 followed the opening of the Tethyan seaway, and a Triassic salt basin developed to the north
31 of the Illizi Basin in the Berkine and Oued Mya basins (Turner and Sherif, 2007; Galeazzi et
32 al., 2010). Mesozoic-Cenozoic deposition in the region was punctuated by Early-Mid Aptian

1 transpression and strike-slip deformation during the Austrian event (Boudjema, 1987;
2 Galeazzi et al., 2010), and by Mid-Late Eocene inversion that caused growth of the Atlas
3 range to the north (Echikh, 1998). Intraplate uplift and magmatism also initiated in North
4 Africa during the Mid-Late Eocene (Wilson and Guiraud, 1992; Wilson et al., 1998; Liégeois
5 et al., 2005) leading to the development of numerous topographic swells and large-scale
6 exhumation of flanking sedimentary basins such as the Illizi and Tim Mersoï basins (English
7 et al., 2016b).

8 The new data in this study comes from an Ordovician sandstone reservoir in a gas-
9 condensate field located in the southern Illizi Basin (Fig. 1). Previous studies have
10 constrained the burial and thermal history of the study area through integration of regional
11 stratigraphy, sonic compaction analysis, biostratigraphy, thermal maturity, fluid inclusion
12 microthermometry, and apatite fission-track data (English et al., 2016b; 2016c). These studies
13 confirmed that the preserved Paleozoic sequence in the Illizi Basin was subjected to elevated
14 temperatures in the past, initially because of additional burial prior to Hercynian exhumation,
15 and again during reburial in the Mesozoic and early Cenozoic (Fig. 2). The available data
16 indicate that maximum burial most likely occurred during the early Eocene, prior to uplift of
17 the Hoggar massif and northward tilting of the Illizi Basin (English et al., 2016b). Based on
18 1-dimensional (1D) modelling, maximum burial depths for the Top Ordovician are estimated
19 at 2.95 km in the northern part of the field (Well A) and 3.37 km in the southern part of the
20 field (Well G), and the corresponding estimates of maximum paleotemperature are 140°C and
21 156°C respectively (English et al., 2016c). The magnitudes of subsequent Cenozoic
22 exhumation are estimated at 1.0 km and 1.4 km for the northern (Well A) and southern (Well
23 G) areas respectively. Based on the modelling of English et al. (2016c), hydrocarbon
24 generation from the lower Silurian source rocks is interpreted to have started during the
25 Carboniferous, ceased temporarily during Hercynian exhumation, and subsequently resumed
26 in the Mesozoic following renewed subsidence (Fig. 2). Trap formation in the study area
27 occurred during the Eocene-Miocene regional tilting of the Illizi Basin, after the primary
28 hydrocarbon generation events, and alternative charging mechanisms have been proposed
29 including (1) long-distance migration from an active source area; (2) remigration of existing
30 hydrocarbons within the basin; (3) phase separation and gas expansion within exhumed
31 preexisting oil and gas accumulations; and/or (4) late-stage exhumation charge during the
32 depressuring of the overlying source rocks (English et al. 2016c).

1

2 **3. Methods**

3 Four samples of formation water were collected at surface during flow testing of the
4 Ordovician sandstone reservoir at two different wells (Well D and Well H) in the study area
5 (Fig. 1). The Ordovician sandstone occurs at a depth of ~2 km in the area present-day. The
6 water samples were sent for compositional analysis at Expro's laboratory in Chandlers Ford,
7 UK. All cations (except ammonium and tetramethylammonium) were determined by
8 inductively coupled plasma (ICP), while ammonium and tetramethylammonium were
9 determined by colorimetry and ion chromatography respectively. Most of the anions were
10 determined by ion chromatography, while the acid anions and bicarbonate concentrations
11 were determined by ion exclusion chromatography, and chloride was determined by
12 potentiometric titration.

13 Fluid inclusion analysis was carried out on 39 Ordovician Unit IV sandstone samples
14 from across seven wells (Wells A through G) in a gas-condensate field in the study area (Fig.
15 1). Aqueous and petroleum inclusions were observed as primary inclusions within quartz
16 overgrowths, along quartz grain boundaries and in paragenetically-late barite cement, and as
17 secondary inclusions in healed microfractures. The homogenization temperature (T_h) of each
18 inclusion was determined optically by incrementally heating the sample until a vapour bubble
19 dissipated into the inclusion (Goldstein and Reynolds, 1994; Walderhaug, 1994). The final
20 melting temperature was measured by initially freezing the samples and recording the
21 temperature of the final disappearance of solid during subsequent heating. The salinity of the
22 aqueous inclusions can then be estimated from the ice-melting temperatures (Oakes et al.,
23 1990; Bodnar, 1993).

24

25 **4. Results**

26 The present-day formation water of the Ordovician sandstone in the study area is a
27 sodium- and calcium-rich (Na-Ca-Cl) brine characterized by little to no sulfate and
28 bicarbonate (Table 1). The total dissolved solids (TDS) content of the samples is uniformly in
29 the 158-161 g/l range. The chloride contents in all four samples are consistently in the 95-98
30 g/l range and constitute > 99% of the anionic species. The sodium content varies between 33-
31 37 g/l, and constitutes 50-60% of the cationic species. Calcium is the second major cation

1 (~30%), and magnesium and potassium are present only in relatively minor concentrations (<
2 5% each). All four samples have low Cl/Br and Na/Br mass ratios (~113 and ~41
3 respectively).

4 The fluid inclusion dataset is dominated by two distinct groups of aqueous inclusions
5 (Table 2; Fig. 3): (1) low-to-high temperature, low-salinity (4-8 wt.%) inclusions, and (2) low
6 temperature, high-salinity (18-25 wt.%) inclusions, with an additional smaller number of
7 inclusions with intermediate temperature and salinity between the two end-member groups.
8 All salinities quoted herein are NaCl equivalent. The salinity of the first group of inclusions
9 predominantly ranges between 4-8 wt.%, and homogenization temperatures extend from
10 95°C up to modal values of 135-155°C. This family of aqueous inclusions co-occurs with
11 single-phase oil inclusions at close to full gas saturation, determined by the similarity of T_h in
12 both aqueous and oil inclusions (Munz, 2001). The second distinct group of aqueous
13 inclusions is characterized by significantly higher salinities of 18-25 wt.% and by cooler
14 modal homogenization temperatures of 90-120°C (Table 2; Fig. 3). Low temperature, high-
15 salinity (~18 wt.%) inclusions are also hosted in barite cement (Fig. 3A) and are associated
16 with phase-separated oil and associated gas inclusions. Further details on the petroleum
17 inclusions from this dataset can be found in English et al. (2016c).

18 It is noteworthy that the high-salinity inclusions are not observed to coincide with
19 maximum burial temperatures indicating that these inclusions record either early (pre-
20 maximum burial) or late (post-maximum burial) fluid compositions. Petrographic evidence
21 indicating that the high-salinity aqueous inclusions post-date the low-salinity aqueous
22 inclusions is summarized in Figure 4.

- 23 1. Primary aqueous inclusions trapped in barite cement in Well A yield
24 homogenization temperatures (98–115°C) and salinities (18–19 wt.%) that are
25 relatively similar to the present-day reservoir temperature (~95°C) and
26 formation water salinity (~14.5 wt.%). Barite occurs as late-stage pore-filling
27 or replacive cement and generally encloses and post-dates all the quartz
28 cement phases (Fig. 4A); the lower salinity primary fluid inclusions in Well A
29 are exclusively hosted in these earlier quartz cements.
- 30 2. In Well B, all of the primary aqueous inclusions are hosted in quartz
31 overgrowths and are from the lower salinity group. However, secondary
32 aqueous oil-associated inclusions in a healed microfracture are observed to

1 cross-cut a quartz overgrowth and detrital grain (Fig. 4B) and therefore post-
2 date the initial quartz overgrowths. These secondary inclusions have higher
3 salinity (14–15 wt.%) and lower homogenization temperatures (108–130°C)
4 than the primary quartz inclusions, which again suggests that the higher
5 salinity inclusions are younger.

- 6 3. The clearest evidence for the relative timing of the two distinct fluid inclusion
7 groups comes from Well D (Fig. 4C). Higher salinity (17.2 wt.%) secondary
8 aqueous inclusions, with homogenization temperatures of 122–124°C, are
9 observed within a healed microfracture that cuts across a quartz cement
10 overgrowth (and detrital quartz grain) that contains primary inclusions of
11 lower salinity (7–8 wt.%) formed at higher temperatures (145–170°C).

12

13 **5. Origin of formation water in Ordovician sandstone**

14 The high salinity of brines is often associated with halite intervals within sedimentary
15 basins. The brines may form either initially through the progressive evaporation of seawater
16 or through dissolution of marine evaporites such as halite (Hanor, 1994). Na-Cl-Br
17 systematics of basinal brines can be used to shed light on their genesis (Rittenhouse, 1967;
18 Carpenter, 1978; Walter et al., 1990; Kesler et al., 1995). Fluid Cl/Br and Na/Br ratios
19 remain constant during progressive evaporation of seawater to the point of halite
20 precipitation, but these ratios start to progressively decrease during subsequent halite, Mg-salt
21 and K-salt precipitation as Br is preferentially retained in the fluid phase (Fig. 5). Conversely,
22 the dissolution of halite will act to increase these ratios because halite is relatively deficient in
23 Br. These ratios are not impacted by subsequent dilution with meteoric water (Kesler et al.,
24 1995).

25 The formation water in this study is characterized by low Cl/Br (~113) and Na/Br
26 (~41) mass ratios relative to seawater (292 and 162 respectively; Carpenter, 1978), and this is
27 consistent with genesis from the progressive evaporation of seawater beyond the point of
28 halite precipitation (Fig. 5). The Paleozoic sequence of the Illizi Basin is dominated by clastic
29 sediments deposited within fluvial, deltaic and marine conditions, and evaporitic sequences
30 were not deposited in the region until the Triassic–Lower Jurassic (Turner and Sherif, 2007).
31 This evaporitic basin was situated across the Berkine and Oued Mya basins, and did not
32 extend as far south as the study area in the southern Illizi Basin (Fig. 1). A comparison with

1 published water analyses from the Triassic reservoir sandstones of the El Borma Field in the
2 Berkine-Ghadames Basin (Morad et al., 1994) confirms that the Ordovician formation waters
3 in the study area have a similar Na-Cl-Br signature to the Triassic brines (Fig. 5). Hence, it is
4 probable that the highly saline formation waters present in the Ordovician sandstones in the
5 Illizi Basin are originally derived from the younger Triassic section in the Berkine Basin,
6 deposited over 400 km to the north (Fig. 1). Calcium-rich brines with salinities in excess of
7 25 wt.% have also been reported from Ordovician rocks in the Hassi Messaoud area by
8 Chiarelli (1973), where the Triassic sequence, including halite, is situated directly on top of
9 Cambro-Ordovician strata (Fig. 6; Bacheller and Peterson, 1991).

10 Abnormally high pressures originate in undercompacted shales sealed by the
11 impermeable salt in the present-day Hassi Messaoud region, and the Oued Mya and Berkine
12 basins (Chiarelli, 1978; Yahi et al., 2001). Mesozoic sediments were deposited directly on
13 Cambro-Ordovician rocks in parts of the basin characterized by deep Hercynian incision (Fig.
14 1; Zeroug et al., 2007; Galeazzi et al., 2010). High-salinity brines may, therefore, have
15 originally infiltrated into the Cambro-Ordovician sequence at these locations (north of the
16 study area; Fig. 6) by density-driven brine reflux (e.g. Bein and Dutton, 1993; Ceriani et al.,
17 2002) during or after the deposition of the evaporitic sequence or by relative overpressuring
18 in the Mesozoic subsalt formations adjacent to Ordovician subcrops (Chiarelli, 1978).

19 20 **6. Temporal evolution of Ordovician formation water**

21 Fluid inclusions are commonly utilized in thermal history, diagenetic and
22 petrographic studies to elucidate the temperature and salinity record of trapped liquid and/or
23 vapour inclusions (Roedder, 1984; Walderhaug, 1994; Goldstein, 2001; Blanchet et al.,
24 2003). It has been demonstrated the fluid inclusion dataset is dominated by two distinct
25 groups of aqueous inclusions (Figs. 3 and 4). The low salinity of the first group of inclusions
26 (4-8 wt.%), and associated homogenization temperatures, record cementation from the onset
27 of quartz diagenesis at 95°C up to likely maximum burial temperatures of 135-155°C (Figs. 2
28 and 4; English et al., 2016c; English et al., in review B). This family of aqueous inclusions
29 co-occurs with single-phase oil inclusions indicating that oil generation also took place in the
30 basin during this time. In contrast, the higher salinities (18-25 wt.%) and cooler modal
31 homogenization temperatures (90-120°C) of the second distinct group of secondary aqueous
32 inclusions (Table 2; Fig. 3) occur within microfractures that crosscut quartz overgrowths

1 containing low-salinity inclusions (Fig. 4). Consequently, the high-salinity (and lower
2 temperature) inclusions are interpreted to post-date the low-salinity (and higher temperature)
3 inclusions (Figs. 2 and 4). A subset of inclusions with intermediate salinities (10-18 wt.%)
4 and homogenization temperatures (Table 2; Fig. 3) may record the transition from one
5 formation water type to the other.

6 The homogenization temperatures of the high-salinity inclusions are most similar to
7 present-day temperatures within the Ordovician sandstone in the study area (95-100°C),
8 although the present-day formation water salinity is a little lower, at ~14.5 wt.% (Table 1;
9 Figs. 3 and 4). Additionally, the low temperature, high-salinity aqueous inclusions hosted in
10 paragenetically-late barite have salinities of ~18 wt.%, and hence manifest the closest
11 characteristics to the present-day formation water (Fig. 4A). In conclusion, the fluid inclusion
12 dataset from the Ordovician sandstones of the Illizi Basin are interpreted to record a
13 significant increase in formation water salinity after, or during, Cenozoic exhumation and
14 cooling (Fig. 2). Phase-separated oil and associated gas are observed in petroleum inclusions
15 in the late-stage barite cement (English et al., 2016c) indicating both gas and oil were present
16 in the pore system during exhumation and cooling, unlike the higher temperature petroleum
17 inclusions that are characterized by single-phase oils.

18 Comparing the connate (original) and present-day salinities in a given stratigraphic
19 unit can provide important insights for bulk transport fluid flow models. The Lower
20 Paleozoic section of the Illizi Basin was deposited in fluvial, deltaic and normal marine
21 environments, and it can be surmised that the original connate water in the Ordovician
22 sandstone during time of deposition was unlikely to be significantly in excess of normal
23 seawater salinity (~35 g/l). This is in sharp contrast to the present-day formation water
24 salinity of ~160 g/l (Table 1) indicating that large-scale vertical or lateral transport of brine
25 into the study area occurred at some point during the history of the basin.

26 Formation water salinity-depth trends have been described in many different
27 sedimentary basins around the world (e.g. Dickey, 1966; 1969; Hanor, 1979; 1994; Gran et
28 al., 1992). Figure 7 illustrates two such trends from (1) Jurassic to Cretaceous strata in
29 southern Arkansas and northern Louisiana (Dickey, 1969), and (2) the northern North Sea
30 (Gran et al., 1992). There is a pronounced increase in salinity with depth in the Gulf Coast
31 example, and salinities approach values of 350,000 ppm at a depth of ~3 km (Fig. 7). The
32 high-salinity brines in this region are interpreted to be derived in part from an underlying
33 evaporitic unit (Moldovanyi and Walter, 1992; Hanor, 1994). In contrast, there is no

1 significant increase in salinity with depth in the northern North Sea example (Fig. 7), where
2 no underlying evaporites are known to occur (Gran et al., 1992; Bjørlykke and Gran, 1994).

3 In comparison, the high temperature, low-salinity fluid inclusions from the Illizi Basin
4 study area lie along the northern North Sea trend (Fig. 7), which suggests that no significant
5 mixing with evaporitic brines occurred during burial or at maximum burial conditions. As per
6 the northern North Sea example, no evaporites were present in the study area because the
7 depositional edge of the Triassic salt basin was situated much further north (Figs. 1 and 6). In
8 contrast, the low temperature, high-salinity fluid inclusion group from the study area plots on
9 the southern Arkansas-northern Louisiana trend (Fig. 7), consistent with the proposed
10 derivation from an evaporitic source, as demonstrated for the present-day formation water
11 (Fig. 5). This dramatic change in the salinity-depth relationship recorded by the fluid
12 inclusion dataset confirms that a significant lateral transport of brine occurred within the
13 basin, and the thermal record of the fluid inclusions confirms that this fluid flow event
14 occurred during or after exhumation (Figs. 2, 4 and 7).

16 **7. Updip migration of deep brine during exhumation**

17 Formation water in the subsurface may be characterized by static or dynamic
18 conditions. Hydrodynamic flow in sedimentary basins is driven by spatial variations in water
19 potential (Hubbert, 1953; England et al., 1987), and can occur as a result of (e.g. Bethke,
20 1989; Bjørlykke, 1993, 2015): (1) gravity-driven flow of meteoric water downwards into the
21 basin caused by an elevated water table on the uplifted basin margins; (2) sedimentary or
22 tectonic compaction-driven flow upward and outward from the basin center caused by fluid
23 overpressure generated during burial by compaction, clay mineral diagenesis and petroleum
24 generation; (3) density-driven flow caused by thermal or salinity gradients which may initiate
25 convection. Potentiometric maps from the Illizi and Berkine basins in Algeria indicate the
26 presence of both an active gravity-flow hydrodynamic regime on the southern flank of the
27 Illizi Basin, and an overpressured system beneath the Triassic salts of the Berkine Basin
28 (Figs. 1 and 6; Chiarelli, 1978).

29 Although present-day potentiometric maps for the Cambro-Ordovician in the Illizi
30 Basin suggest northward hydrodynamic flow of meteoric water from elevated outcrops in the
31 south (Chiarelli, 1978), the fluid inclusion data presented here indicate that exhumation of the
32 Illizi Basin was associated with a major flux of highly saline brines that most likely

1 originated from the salt-rich section to the north of the study area (Fig. 6). Triassic deposits
2 directly overlie Cambro-Ordovician strata along major Hercynian structures (Fig. 1), and the
3 high-salinity brines may have originally infiltrated the Cambro-Ordovician sequence at these
4 locations via density-driven brine reflux during or after the deposition of the Triassic
5 evaporitic sequence. Lateral flow within the Cambro-Ordovician sequence is interpreted to
6 have been focused between the underlying crystalline basement and the overlying compacted
7 Lower Silurian shales, and may also have been facilitated by natural fracture systems within
8 the sandstone-dominated sequence. Compaction-driven lateral flow is ruled out on the basis
9 that the Illizi Basin was undergoing exhumation during precipitation of the cements
10 containing the lower temperature/higher salinity fluids (Fig. 2), and deposition had largely
11 ceased in the Berkine Basin to the north. Rayleigh convection is ruled out on the basis that it
12 requires thick sequences (100-300 m) of homogeneous and porous sandstones with no
13 interbedded thin shales or cemented intervals (Bjørlykke et al., 1988). Additionally, the
14 apparent updip migration of higher salinity brine, displacing less dense and lower salinity
15 formation water, indicates that density contrast was not a driving factor for the fluid flow.

16 Instead, we propose that breaching of a closed, relatively overpressured system during
17 basin exhumation is an alternative mechanism for regional updip flow of formation water.
18 New discharge sites were created along the newly exposed southern margin of the basin
19 during the Cenozoic (Fig. 6) and opening of the basin margin to normal hydrostatic
20 conditions during exhumation could have generated the required regional lateral gradient in
21 fluid potential (i.e. relatively overpressured conditions to the north and normal hydrostatic
22 conditions to the south). Alternatively, if the original overpressure in the basin was laterally
23 discontinuous and compartmentalized by faulting, it is possible that some of these faults may
24 have become critically-stressed (Barton et al., 1995) during the removal of overburden during
25 exhumation (e.g. Corcoran and Doré, 2002; English et al., in review A). This would lead to
26 the progressive release of overpressured fluid through frictional sliding along optimally-
27 oriented pre-existing faults (Sibson, 2000; Finkbeiner et al., 2001) during exhumation until
28 the system regains equilibrium. Recent geomechanical analysis in the study area has
29 confirmed that the primary N-S and NW-SE fault systems in this part of the Illizi Basin are
30 likely to be critically stressed under the present-day *in situ* stress conditions (English et al., in
31 review A).

32 The fluid flow resulting from pressure release may be restricted to a geologically
33 short time period, depending on distance and effective permeability, because equilibration of

1 unsteady state pressure distributions is one of the quickest mixing processes for subsurface
2 fluids (e.g. Smalley and Muggeridge, 2010). The volume of fluid flux may also be somewhat
3 limited because of the low compressibility of water (Bjørlykke, 1993, 2015). However, if
4 overpressure release results in increased effective stress in deeper, less exhumed parts of the
5 basin, additional compaction and pore-volume loss may supplement the updip fluid flow.
6 Additionally, volumetric expansion of *in situ* gaseous hydrocarbons during basin exhumation
7 (e.g. Doré and Jensen, 1996; Doré et al., 2002; English et al., 2016c), exhumation charge
8 from petroleum source rocks (English et al., 2016a), and methane exsolution from formation
9 water may also serve to displace significant additional volumes of formation water updip if
10 these incremental hydrocarbon volumes are trapped within the basin.

11 Following the removal of the lateral gradient in fluid potential due to the bleed-off of
12 excess pressure, subsequent fluid-flow reversal by gravity-driven flow of meteoric water
13 from the basin margins may have become more well established (Fig. 6). This could account
14 for the decreased salinity in the present-day formation water (~14.5 wt.%) compared to the
15 higher salinity of the migrated basinal brines (low-temperature aqueous inclusions >18 wt.%;
16 Figs. 3A and 4). Additionally, the mixing of barium-rich brines with sulfate-rich meteoric
17 waters may also account for the precipitation of late-stage barite cement found in permeable
18 beds (>50 md) within the Upper Ordovician (Fig. 4A; English et al., 2016c; in review B).
19 Chiarelli (1973, 1978) has also noted the deep invasion of relatively freshwater in shallower
20 stratigraphic zones in the Illizi Basin with salinities < 0.5 wt.% being present in Devonian
21 sandstones up to 200 km north of their southern outcrops.

22

23 **8. Conclusions**

24 Existing drive mechanisms for fluid flow in sedimentary basins include gravity-driven
25 flow, compaction-driven flow, and density-driven convection. Interrogation of fluid inclusion
26 and present-day water chemistry data from Ordovician sandstones confirms that long
27 distance, lateral, brine migration and mixing have occurred within the Illizi Basin, Algeria.
28 The Na-Cl-Br characteristics of the Ordovician sandstone formation water in the study area is
29 consistent with derivation from the Triassic-Liassic halite-bearing sequence deposited in the
30 Berkine Basin, which is located > 400 km to the north. The hydrogeologic record preserved
31 by fluid inclusions within the Ordovician sandstone suggests that an early stage, low-salinity
32 water was displaced by a higher salinity fluid during, and subsequent to, exhumation from

1 maximum burial depth and temperature. It is proposed that the release of fluid overpressure
2 during exhumation of the Illizi Basin may have been a critical contributor to updip fluid flux.
3 Fluid flux during basin exhumation has not been considered widely in the literature, and this
4 model may have implications for the timing and direction of major fluxes of deep basin
5 brines and associated fluids in the geological record.

6

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1 **List of Figures and Tables**

2 **Figure 1:** Location of the study area in the Illizi Basin, Algeria. Extent of Triassic-Jurassic
3 evaporitic basin and clastics adapted from Turner and Sherif (2007) and Galeazzi et al.
4 (2010). Regions of present-day high pressured Triassic and Ordovician systems from
5 Chiarelli (1978). Transect line corresponds to Figure 5.

6 **Figure 2:** 1D burial history model (Well G) for the study area in the Illizi Basin (modified
7 from English et al., 2016c).

8 **Figure 3:** Salinity [A] and homogenization temperature [B] distributions from aqueous fluid
9 inclusions in the Upper Ordovician sandstones from the study area in the Illizi Basin.
10 Salinities are in wt.% NaCl equivalent.

11 **Figure 4:** Petrographic evidence indicating that the higher-salinity, lower temperature
12 aqueous fluid inclusions post-date the lower-salinity, higher temperature aqueous fluid
13 inclusions. A: Lower temperature, higher-salinity aqueous inclusions trapped in late-stage
14 barite cements. B: Higher-salinity secondary inclusions observed in a microfracture that cuts
15 across a detrital quartz grain and quartz overgrowth. C: Higher-salinity secondary inclusions
16 observed in a microfracture that cuts across a quartz overgrowth containing lower salinity,
17 higher temperature primary inclusions. Salinities are in wt.% NaCl equivalent.

18 **Figure 5:** Na-Cl-Br systematics for basinal brines (based on Kesler et al., 1995). Solid line
19 shows the compositional change during progressive seawater evaporation (data from
20 McCaffrey et al., 1987 and Carpenter, 1978). The Ordovician formation water in this study is
21 characterized by relatively low Cl/Br and Na/Br ratios that are consistent with residual brines
22 generated during progressive evaporation of seawater beyond the point of halite precipitation.
23 The Triassic formation waters in the El Borma Field (Fig. 1, Morad et al., 1994) also have a
24 similar signature. These systematics are not impacted by subsequent dilution with meteoric
25 water, and exchange reactions involving Na and Ca would result in data plotting off of the
26 defined seawater evaporation and halite dissolution trends (Kesler et al., 1995).

27 **Figure 6:** Schematic cross-section of the Illizi, Berkine and Oued Mya basins (modified from
28 Galeazzi et al., 2010). Triassic deposits directly overlie Cambro-Ordovician strata along
29 major Hercynian structures north of the study area. High-salinity brines may have originally
30 infiltrated the Cambro-Ordovician sequence at these locations via density-driven brine reflux
31 during or after the deposition of the Triassic evaporitic sequence. Aqueous fluid inclusions in

1 Ordovician sandstones record the migration of a high-salinity brine into the study area during
2 cooling and exhumation of the Illizi Basin. Lateral flow within the Cambro-Ordovician
3 sequence is interpreted to be focused between the underlying crystalline basement and the
4 overlying Lower Silurian shales. Present day, the sub-salt area around Hassi Messaoud and
5 Hassi R'Mel is characterized by an overpressured system while an active gravity-driven
6 hydrodynamic flow system has been interpreted at multiple stratigraphic levels on the
7 southern margin of the exhumed Illizi Basin (Chiarelli, 1978). The burial history of the study
8 area in the Illizi Basin is shown in Figure 2 and illustrates the major exhumation of the Illizi
9 Basin during the Cenozoic. Section line is shown on Figure 1.

10 **Figure 7:** Formation water salinity-depth trends from (1) Jurassic to Cretaceous strata in
11 southern Arkansas and northern Louisiana (Dickey, 1969; data from Hawkins et al., 1963),
12 and (2) the northern North Sea (Gran et al., 1992; including data from Egeberg and Aagaard,
13 1989). The higher salinity formation waters in the Gulf Coast example are interpreted to be
14 derived from an underlying evaporitic unit (Moldovanyi and Walter, 1992; Hanor, 1994),
15 whereas no underlying evaporites are known to occur in the northern North Sea example
16 (Gran et al., 1992; Bjørlykke and Gran, 1994). The high temperature, low-salinity fluid
17 inclusions in the Illizi Basin study area lie along the northern North Sea trend, whereas the
18 low temperature, high-salinity fluid inclusions plot along the southern Arkansas-northern
19 Louisiana trend. This dramatic change in the salinity-depth relationship recorded by the fluid
20 inclusion dataset indicates that a significant lateral transport of brine likely occurred within
21 the basin at some point during its history.

22 **Table 1:** Compositional analysis of formation water in the Ordovician sandstone of the Illizi
23 Basin

24 **Table 2:** Summary of fluid inclusion data from Upper Ordovician sandstone in the Illizi
25 Basin

Figure 1

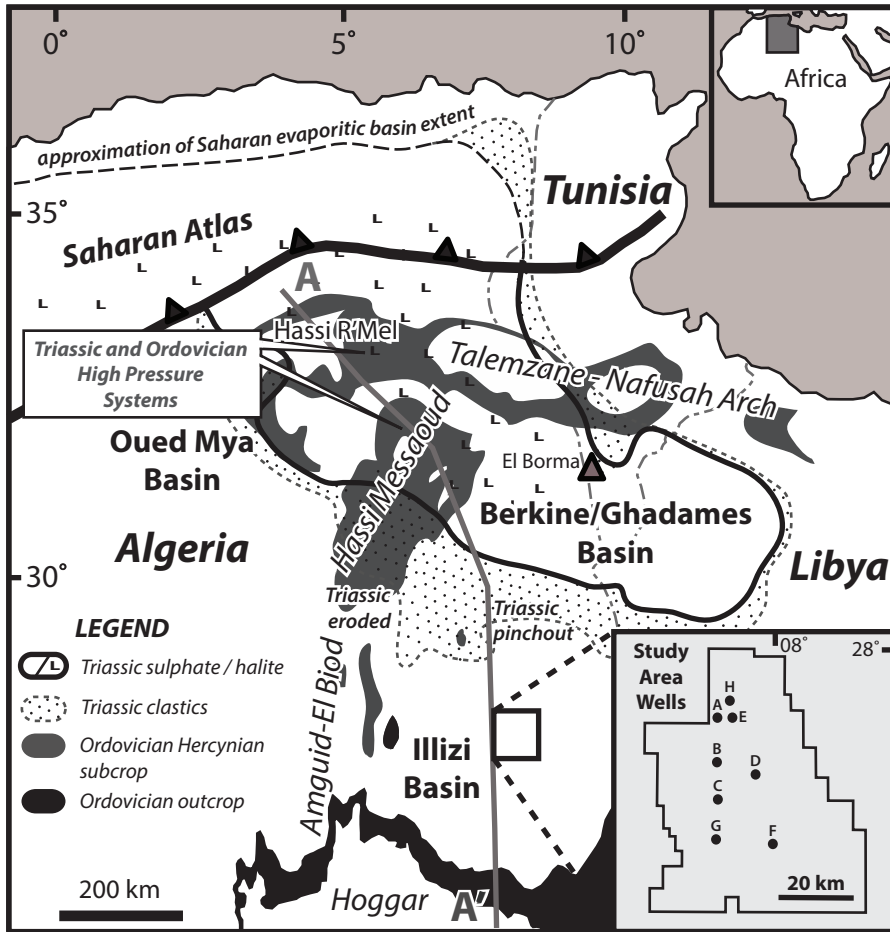


Figure 2

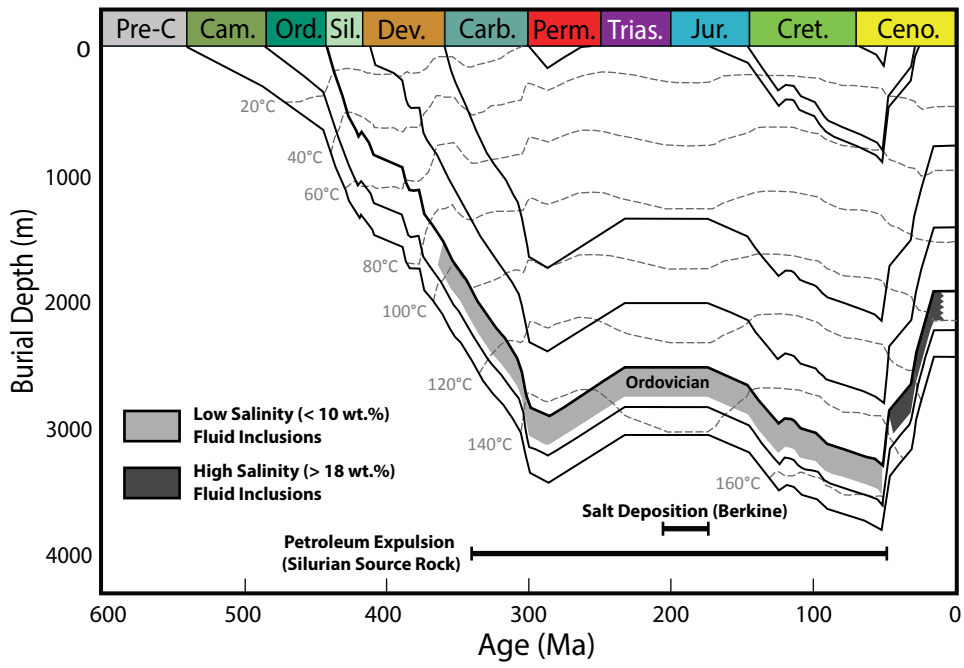
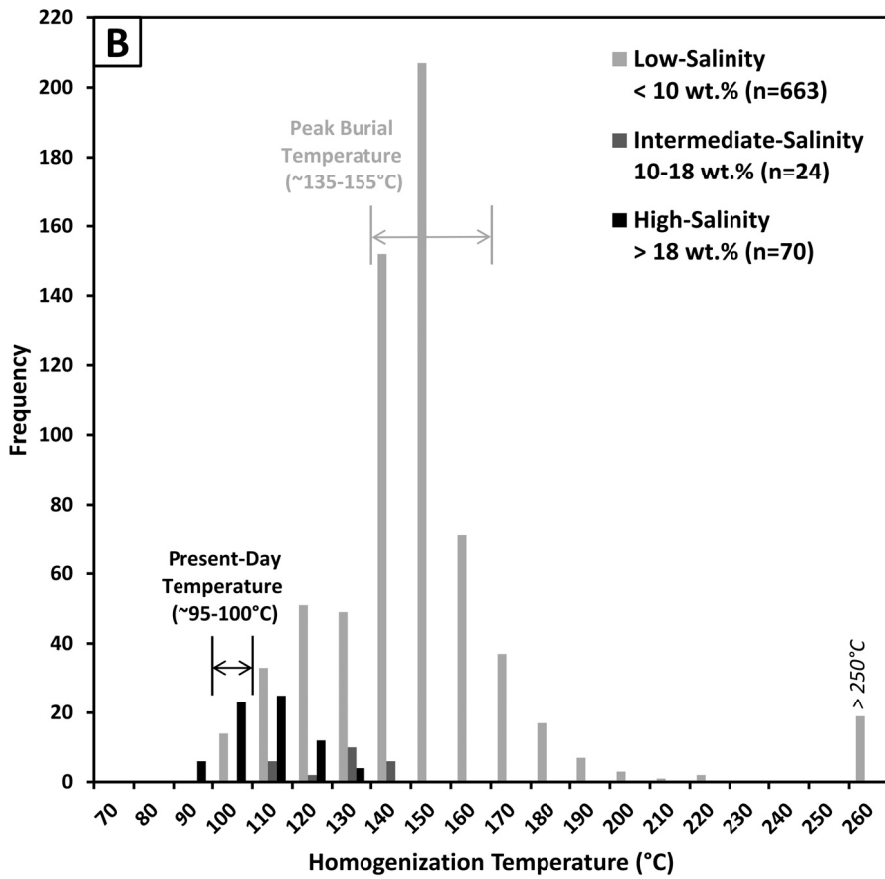
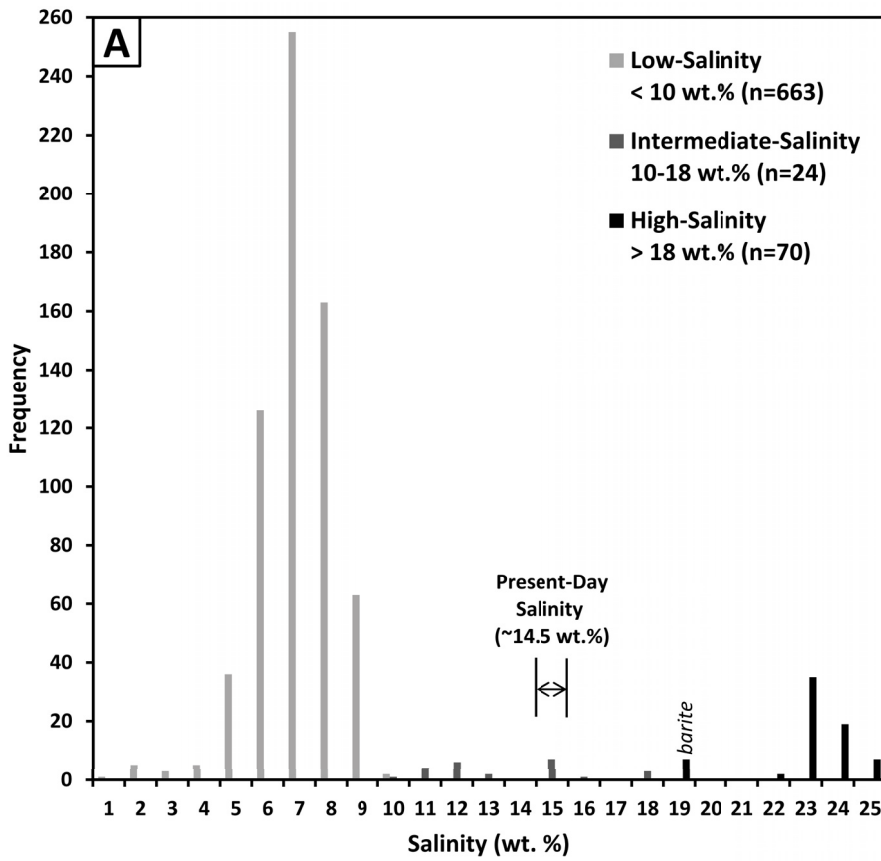
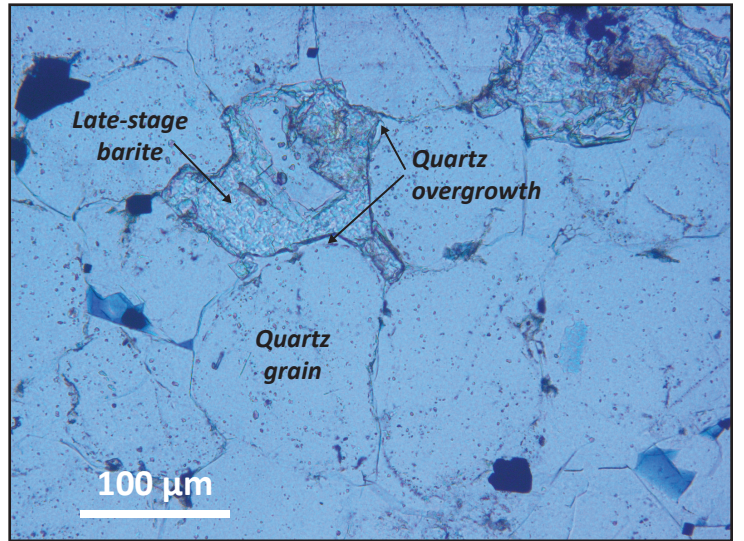
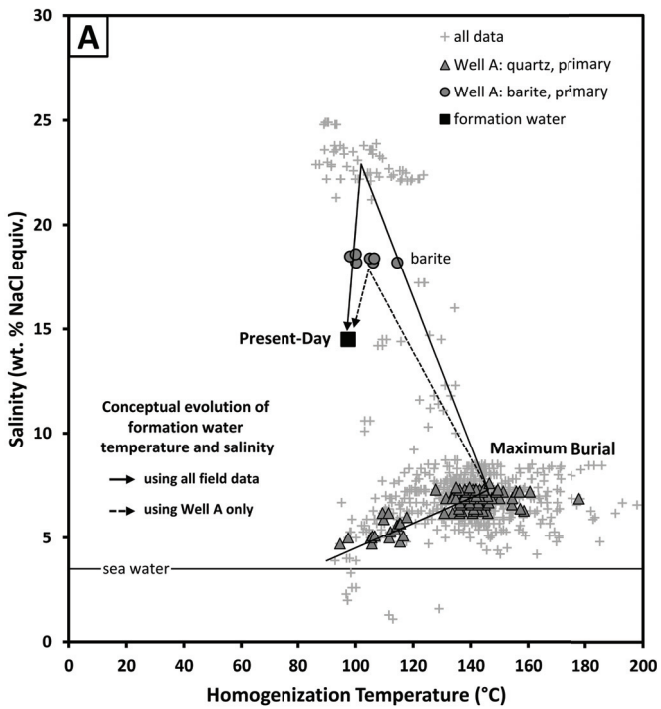
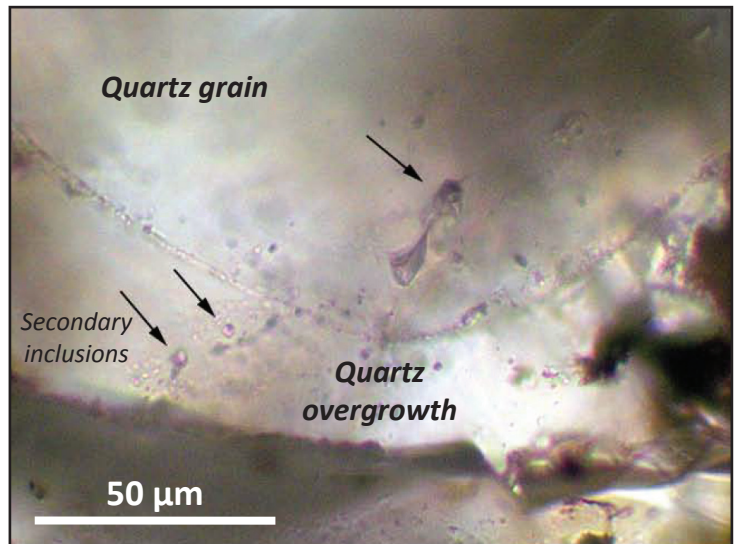
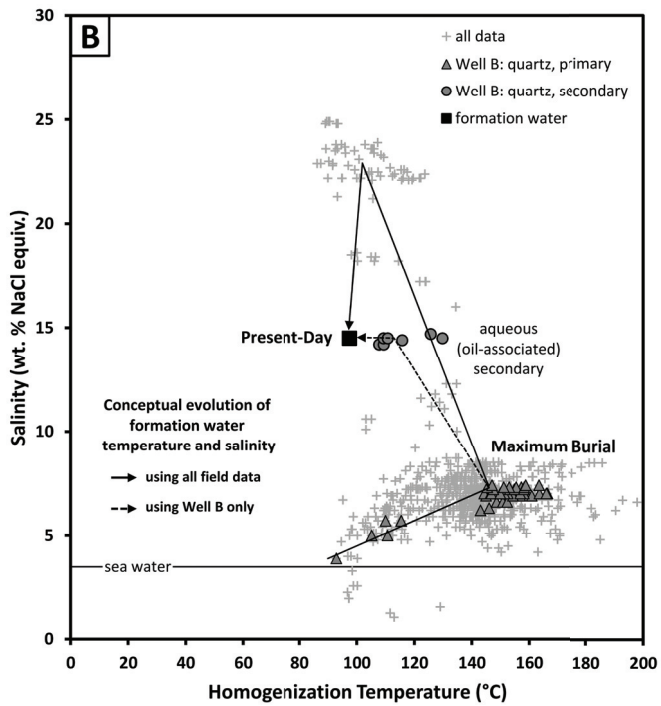


Figure 3

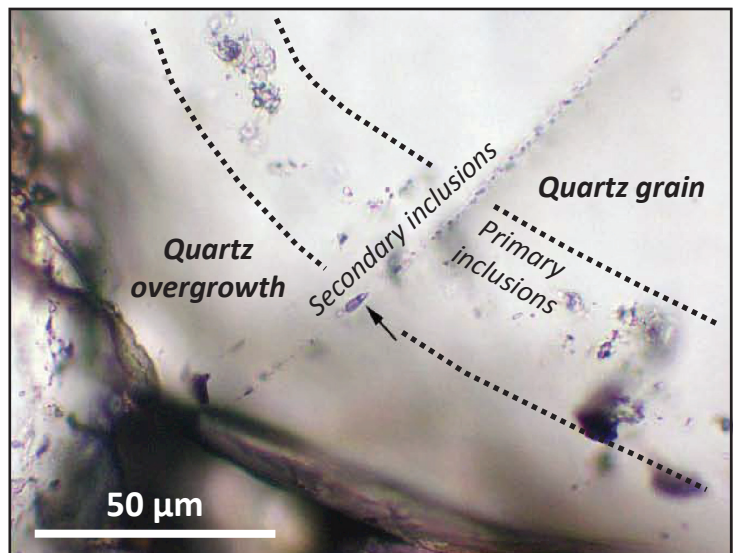
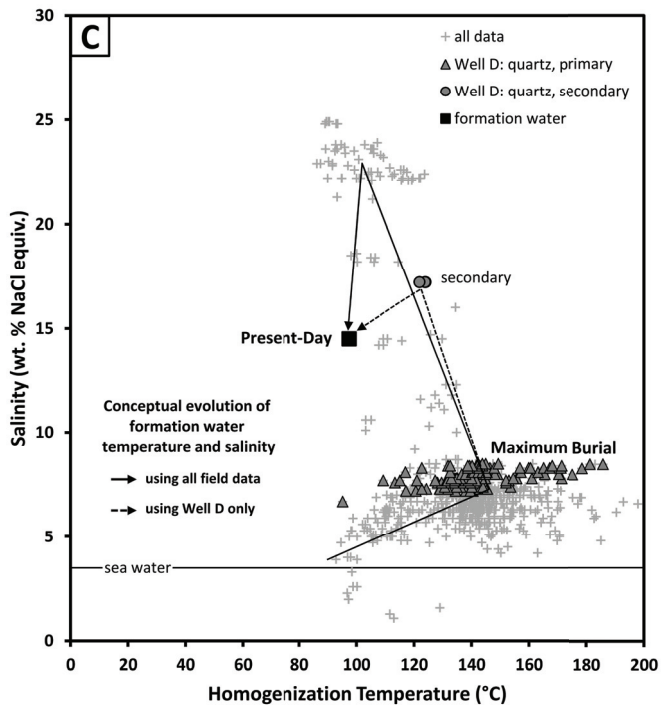




Well A: 1916.6 m



Well B: 1914.5 m



Well D: 1917.1 m

Figure 5

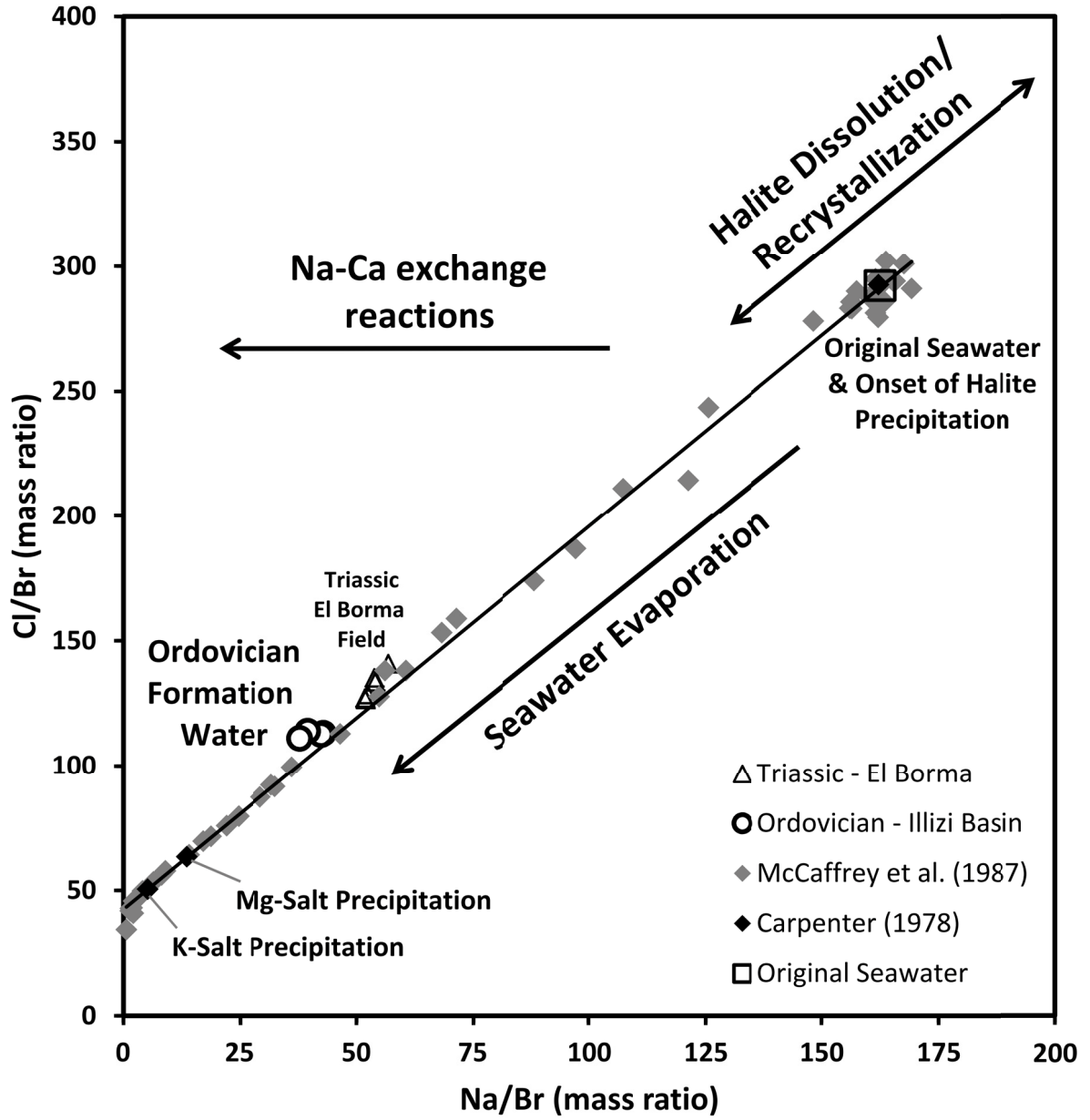


Figure 6

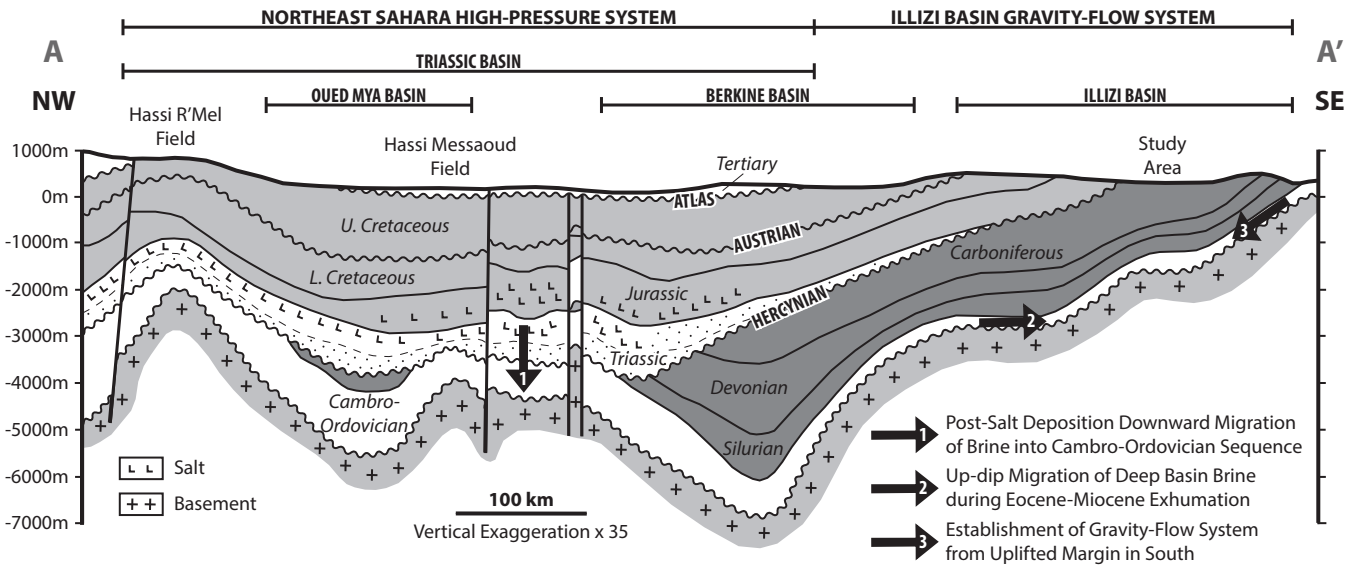


Figure 7

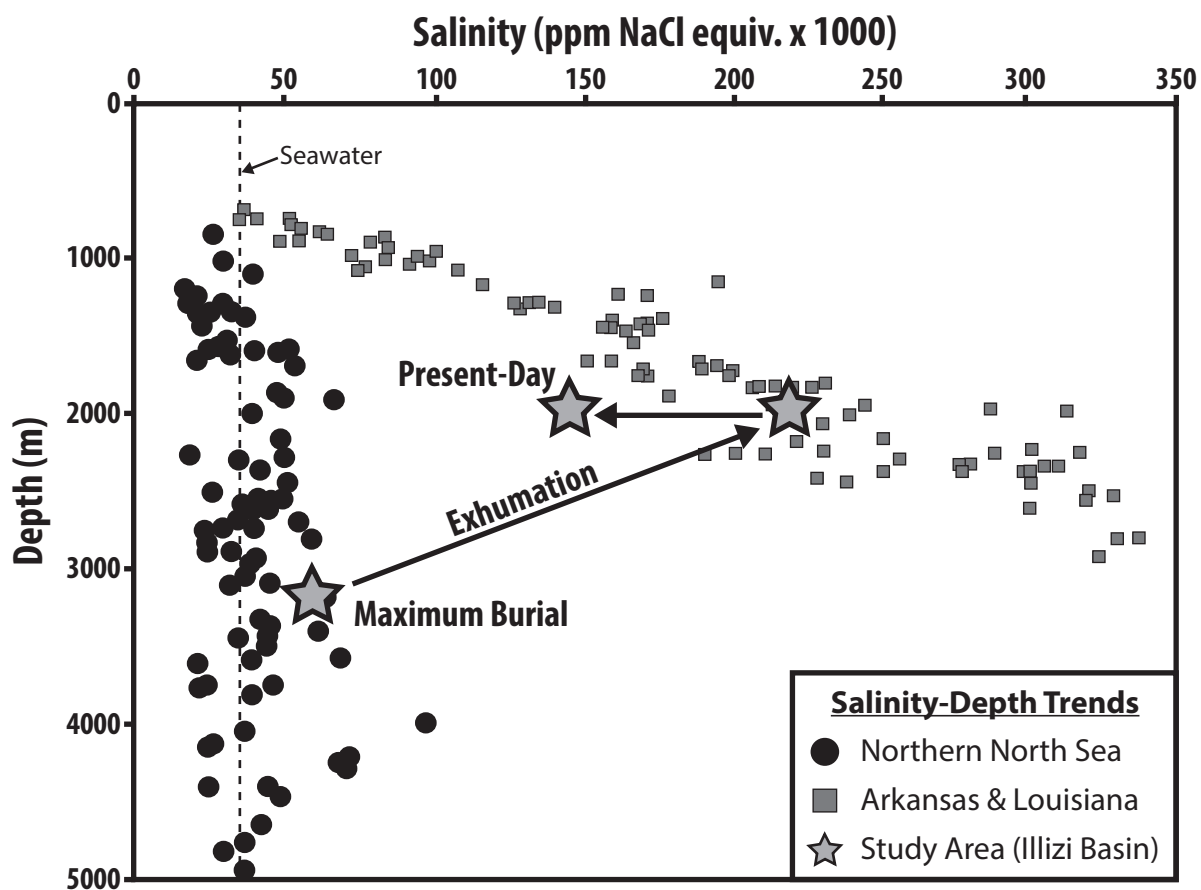


TABLE 1: Compositional analysis of formation water in the Ordovician sandstone of the Illizi Basin

By Volume (mg/L)

Field / Well Reference	Well H (pre-frac)	
Sample Reference	1.11	1.15
Physicochemical Parameters		
pH @ 19.9 °C (pH units)	3.95	4.18
Resistivity @ 20 °C (ohm.m)●	0.0650	0.0643
Density @ 20.03 °C (kg/L)*	1.1098	1.1115
Anionic Species (mg/L)		
Chloride	96,398	97,835
Sulphate	3	2
Bromide	846	881
Nitrate	3.0	2.6
Phosphate	-	-
Iodide	< 3	< 3
Bicarbonate	< 11	< 11
Carbonate	0	0
Hydroxide	0	0
Formate	28.2	20.7
Acetate	155	156
Propionate	12	12
Butyrate	2.6	2.3
iso -Valerate	< 2	< 2
Cl:Br	114	111
<i>HCl Preserved</i>		
Sample Reference	1.13	1.17
Density @ 20.06 °C (kg/L)*	1.1104	1.1121
Boron (as Tetraborate $B_4O_7^{2-}$)	22	23
Cationic Species (mg/L)		
Lithium	52	55
Barium	523	677
Strontium	695	731
Calcium	20,449	21,395
Magnesium	1,880	1,953
Sodium	33,496	33,352
Potassium	2,203	2,082
Iron	520	458
Copper	< 1	< 1
Zinc	< 2	< 2
Manganese	41	44
Aluminium	< 2	< 2
Ammonium	87.9	88.7
Tetra Methyl Ammonium	< 22	< 22
Total Species (mg/L)		
Total Barium	529	678
Total Iron	508	485
Elements (mg/L)		
Phosphorus	< 6	< 6
Silicon	13	15
Sulphur	23	19
Total Cl ⁻ equivalent (mg/L)	96,910	98,356
Total Na ⁺ equivalent (mg/L)	63,095	64,134
Total NaCl equivalent (mg/L)	160,004	162,490
TDS (calculated) (mg/L)	158,467	160,947
Cation/Anion Balance	100.40%	100.55%
Cation/Anion Bias	0.40%	0.55%

By Volume (mg/L)

Field / Well Reference	Well D (post-frac)	
Sample Reference	2-26	2-27
Physicochemical Parameters		
pH @ 19.4 ± 1.6 °C (pH units)	5.87	5.65
Resistivity @ 20 °C (ohm.m)●	0.0678	0.0671
Density @ 20.07 ± 0.11 °C (kg/L)*	1.1108	1.1126
Anionic Species (mg/L)		
Chloride	95,358	96,926
Sulphate	< 1	< 1
Bromide	843	861
Nitrate	2.4	2.9
Phosphate	< 2	< 1
Iodide	3.9	4.0
Bicarbonate	39	33
Carbonate	0	0
Hydroxide	0	0
Formate	3.7	3.2
Acetate	86	87
Propionate	4.2	4.3
Butyrate	< 1	< 1
iso -Valerate	< 1	< 1
Cl:Br	113	113
<i>HCl Preserved</i>		
Sample Reference	2-26	2-27
Density @ 20.06 °C (kg/L)*	1.1104	1.1121
Boron	20	20
Cationic Species (mg/L)		
Lithium	75	77
Barium	2,994	3,122
Strontium	1,404	1,449
Calcium	18,337	18,436
Magnesium	1,494	1,519
Sodium	36,183	36,556
Potassium	1,025	1,044
Iron	73	78
Copper	< 2	< 2
Zinc	< 1	< 1
Manganese	10.1	10.2
Aluminium	< 11	< 11
Ammonium	115	121
Tetra Methyl Ammonium	197	182
Total Species (mg/L)		
Total Barium	2,959	3,059
Total Iron	158	161
Elements (mg/L)		
Phosphorus	< 11	< 11
Silicon	18	19
Sulphur	< 21	< 21
Total Cl ⁻ equivalent (mg/L)	95,847	97,420
Total Na ⁺ equivalent (mg/L)	62,914	63,534
Total NaCl equivalent (mg/L)	158,760	160,954
TDS (calculated) (mg/L)	157,974	160,250
Cation/Anion Balance	101.22%	100.57%
Cation/Anion Bias	1.22%	0.57%

TABLE 2: Summary of Fluid Inclusion Data from Upper Ordovician Sandstone in the Illizi Basin

Category	n	Ave. T_h (°C)	±	T_{ice} (°C)	±	Salinity (wt.%)†	±
Low Salinity (<10 wt.%)	663	142.6	25.6	-3.9	1.7	6.6	1.2
Intermediate Salinity (10-18 wt.%)	24	121.6	10.4	-9.2	2.2	13.1	2.3
High Salinity (>18 wt.%)	70	102.9	10.0	-20.1	2.2	22.5	1.6

† Salinities are in wt.% NaCl equivalent