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To cite this version:

Pauline Harlé, Alexandra Kushnir, Coralie Aichholzer, Michael Heap, Régis Hehn, et al.. Heat flow density estimates in the Upper Rhine Graben using laboratory measurements of thermal conductivity on sedimentary rocks. Geothermal Energy, 2019, $10.1186/s40517-019-0154-3$. hal-02448402

HAL Id: hal-02448402 <https://hal.science/hal-02448402v1>

Submitted on 22 Jan 2020

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RESEARCH

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Heat flow density estimates in the Upper Rhine Graben using laboratory measurements of thermal conductivity on sedimentary rocks

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Abstract

The Upper Rhine Graben (URG) has been extensively studied for geothermal exploitation over the past decades. Yet, the thermal conductivity of the sedimentary cover is still poorly constrained, limiting our ability to provide robust heat flow density estimates. To improve our understanding of heat fow density in the URG, we present a new large thermal conductivity database for sedimentary rocks collected at outcrops in the area including measurements on (1) dry rocks at ambient temperature (dry); (2) dry rocks at high temperature (hot) and (3) water-saturated rocks at ambient temperature (wet). These measurements, covering the various lithologies composing the sedimentary sequence, are associated with equilibrium-temperature profles measured in the Soultz-sous-Forêts wells and in the GRT-1 borehole (Rittershofen) (all in France). Heat flow density values considering the various experimental thermal conductivity conditions were obtained for diferent depth intervals in the wells along with average values for the whole boreholes. The results agree with the previous heat flow density estimates based on dry rocks but more importantly highlight that accounting for the efect of temperature and water saturation of the formations is crucial to providing accurate heat flow density estimates in a sedimentary basin. For Soultz-sous-Forêts, we calculate average conductive heat flow density to be 127 mW/m² when considering hot rocks and 184 mW/m² for wet rocks. Heat flow density in the GRT-1 well is estimated at 109 and 164 mW/m² for hot and wet rocks, respectively. Results from the Rittershoffen well suggest that heat flow density is nearly constant with depth, contrary to the observations for the Soultz-sous-Forêts site. Our results show a positive heat fow density anomaly in the Jurassic formations, which could be explained by a combined efect of a higher radiogenic heat production in the Jurassic sediments and thermal disturbance caused by the presence of the major faults close to the Soultz-sous-Forêts geothermal site. Although additional data are required to improve these estimates and our understanding of the thermal processes, we consider the heat flow densities estimated herein as the most reliable currently available for the URG.

Keywords: Heat flow density, Upper Rhine Graben, Thermal conductivity, Saturation, Temperature, Sedimentary rocks, Geothermal energy, Soultz-sous-Forêts, Rittershofen

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Background

The Upper Rhine Graben (URG) has been known to be a region of high heat flow density for more than 40 years (Cermák and Rybach 2012; Gable 1986; Lucazeau and Vasseur 1989; Rybach 2007; Vasseur 1980, 1982). Tis regional conductive heat fow density anomaly is due to crustal thinning caused by extension during the Cenozoic and facilitated by the basin-wide deep-rooted groundwater circulation that locally enhances the surface heat fow density (Clauser and Villinger 1990; Schellschmidt and Schulz 1992). Due to the heat flow density anomaly, the URG represents an ideal region for geothermal exploitation. Located in the northeastern part of France and southwestern part of Germany (Fig. 1), this continental rift is already home to several operational (e.g., Soultz-sous-Forêts and Rittershofen, both France, and Bruchsal, Insheim and Landau, all in Germany) and planned (e.g., Illkirch, Vendenheim, and Wissembourg, all France) geothermal sites (Aichholzer et al. 2016; Vidal and Genter 2018), but accurate estimates of heat fow density remain crucial for temperature estimations prior to drilling operations and thermodynamic models that inform our understanding of measured temperature profles for continued geothermal exploration (Beardsmore et al. 2001; Flores Marquez 1992; Frone et al. 2015; Schütz et al. 2014).

Thermal profiles of operational wells have revealed conductive heat flow density in the upper 1–1.5 km, as opposed to convective heat fow density common to the geothermal reservoir (Bächler et al. 2003; Baillieux et al. 2013; Clauser and Villinger 1990; Guillou-Frottier et al. 2013; Kohl et al. 2000; Le Carlier et al. 1994; Magnenet et al. 2014; Pribnow and Schellschmidt 2000). When conductive regime dominates in the Earth's crust, conductive heat fow density (also called surface or terrestrial heat fow density) is estimated using Fourier's law of heat conduction $q=\lambda\times\left(\frac{\Delta T}{\Delta z}\right)$, where q is the conductive heat flow density (W/m²), λ is thermal conductivity (W/mK) and $\left(\frac{\Delta T}{\Delta z}\right)$ is the temperature gradient $({\rm {}^{\circ}C/m})$ (Beardsmore et al. 2001). Conductive heat flow density can thus be defned as the product of the thermal gradient and the mean thermal conductivity of the medium. Thermal conductivity describes the capacity of a material to transfer heat in the presence of a thermal gradient and is a major parameter controlling the conductive heat flow density. The thermal conductivity of the rocks depends not only on mineralogical composition and microstructure (porosity, fracture density, texture), but also on pressure, rock temperature and degree of saturation and nature of the fluid. The effect of those parameters on thermal conductivity have been largely investigated by previous studies, which showed that the thermal conductivity of rocks generally decreases with increasing porosity (Woodside and Messmer 1961; Popov et al. 2003; Nagaraju and Roy 2014; Guo et al. 2017; Mielke et al. 2017), whereas pressure tends to reduce porosity, close (micro)cracks and improve heat transport at grain–grain contacts and, thus, increase thermal conductivity (Walsh and Decker 1966; Abdulagatova et al. 2009; Schön and Dasgupta 2015). On the contrary, high temperature tends to decrease the thermal conductivity of rocks due to diferential thermal expansion of the minerals, which may increase contact resistances between the grains and result in thermal cracking creating porosity (Vosteen

and Schellschmidt 2003; Abdulagatov et al. 2006; Abdulagatova et al. 2009; Guo et al. 2017). Finally, thermal conductivity increases with water saturation compared to dry conditions as water conducts heat much better than air (Zimmerman 1989; Popov et al. 2003; Schütz et al. 2012; Nagaraju and Roy 2014; Guo et al. 2017; Albert et al. 2017).

Thus, at prospective geothermal sites where conduction dominates heat transfer in the upper few kilometers of the crust, a combined understanding of the geology and the thermal conductivity of the rocks (preferably at the in situ temperature and fuid saturation conditions) can be used to provide useful conductive heat fow density estimates.

Studies have defned a mean conductive heat fow density between 100 and 120 mW/ m^2 in the URG, with a maximum value of 150 mW/m² at Soultz-sous-Forêts (Baillieux et al. 2013; Kohl et al. 2000), much higher than the mean value of 60 mW/m 2 in Europe (Majorowicz and Wybraniec 2011). Unfortunately, the absence of a large database for thermal conductivities of sedimentary rocks from the region implies that these heat fow density estimates are based on a limited number of thermal conductivity values, often determined for general lithologies that may not be fully representative of the formations within the graben (Flores Marquez 1992). This lack of data for the upper part of the sedimentary cover can lead to unreliable estimations of the local heat fow density (Fuchs and Balling 2016). Moreover, the thermal conductivity values considered for these early heat fow density estimations are for rocks in the dry state at ambient temperature and the absence of URG-specific models makes them difficult to port to in situ conditions. Indeed, while mixing models (Abdulagatova et al. 2009; Fuchs et al. 2013; Vosteen and Schellschmidt 2003) are valuable when no or very few samples from the study area are available, it is always preferable to gather a regional database when possible. Tus, new thermal conductivity measurements under various temperature and saturation conditions for a wide range of sedimentary formations from the URG are required to provide robust heat flow density estimates.

Although geothermal exploitation is well-developed in the URG, the thermal conductivity of the sedimentary cover is still poorly known, mostly due to the lack of core material, in particular in the upper strata (above the *Muschelkalk* series) where conduction predominates.

Here, we analyze the behavior of thermal conductivity of sedimentary rocks sampled in the URG as a function of diferent parameters including porosity, fuid saturation and temperature, with the aim of providing a new thermal conductivity database for the URG. This large database is used to provide more accurate heat flow density estimates for sites where an equilibrium-temperature profile is known. The influence of the various thermal conductivity measurement conditions on the calculated heat fow density is also investigated. Using these laboratory data, we estimate the heat fow density for the Soultz-sous-Forêts and Rittershofen operational industrial wells.

The geology of the Upper Rhine Graben (URG)

To perform this study, we collected analogue rock samples that best represent the geological formations of the Upper Rhine Graben.

In this study, "facies" refers to the lithology (rock type), sedimentary structures and depositional environments characterizing the rock. "Sedimentary formation" refers to a set of homogenous sedimentary strata with the same facies and paleontological content and characterized by obvious lower and upper boundaries. A "series" of formations, like the *Muschelkalk* or the *Buntsandstein* series, refers to a group of sedimentary formations deposited within the same geodynamic context, for example in fuvial systems for the *Buntsandstein*.

In the URG, the deep Paleozoic basement is covered by Permian clastic formations. Above these Permian sedimentary rocks are the fuvial to fuvio-deltaic sandstones and conglomerates of the lower Triassic *Buntsandstein* (B) series, overlain by marine-to-lagoonal sediments of the middle Triassic *Muschelkalk* (M) series (Aichholzer et al. 2016). The *Muschelkalk* formations are mainly composed of interbedded marlycalcareous rocks, shelly sandstones, evaporitic deposits, and dolostones (Duringer et al. 2019; Ménillet 2015). The carbonate-shelf environment that formed the upper *Muschelkalk* series was followed by the evaporitic episode of the *Keuper* (K), upper Triassic. The *Keuper* is characterized by massive clays, marly clays and dolomites that contain evaporitic deposits (gypsum and anhydrite). Some sandstone marker beds are also encountered in the lower, middle and upper *Keuper* formations (Aichholzer et al. 2016; Duringer et al. 2019; Ménillet 2015). The depositional environment became fully marine from the Jurassic, with marls/limestones alternations during Hettangian, followed by clays and clayey marls alternations with occasional calcareous banks up to the base of Bajocian. Bajocian was characterized by carbonate-shelf episodes (Duringer et al. 2019; Ménillet 2015). The sedimentary formations described above were deposited before the rift episode of the graben and can be found beyond the borders of the URG.

The Tertiary rocks were unconformingly deposited on top of the eroded upper Jurassic sediments (Berger et al. 2005; Duringer et al. 2019; Hinsken et al. 2007, 2011; Roussé 2006; Sittler 1965, 1992). Eocene and lower Oligocene formations are composed, from base to top, by lacustrine and evaporitic clays, marls, limestones and a few sandstones in the central part of the graben and conglomerates on the borders (*Zone Dolomitique, Couche Rouge* and *Couches de Pechelbronn* in the North and *Zone Salifère inférieure, moyenne* and *supérieure* in the South (see Duringer et al. 2019). Middle and upper Oligocene deposits are formed by marine clays and marls intercepted by some poorly cemented sandstones (*Série Grise*). The retreat of the sea in the graben at the end of the Rupelian (upper Oligocene) led to the deposition of the thick formation of the *Couches de Niederroedern* also called *Série Bariolée*. Tis formation is a lacustrine-to-fuvial variegated marly formation attributed to the Chattian (Berger et al. 2005; Hinsken et al. 2007, 2011; Roussé 2006; Sittler 1965). The upper Oligocene is eroded in some parts of the rift and the Miocene does not exist in the whole graben due to erosion (Aichholzer et al. 2016; Berger et al. 2005; Roussé 2006). The uppermost part of the sedimentary cover is characterized by a continental episode made of fuvial to ephemeral lake deposits that were formed during the Pliocene. Loess deposited during the Quaternary ends the sedimentary cover (Duringer et al. 2019; Ménillet 2015).

Case study: the Soultz‑sous‑Forêts geothermal site

Started in the 1980s, the Soultz-sous-Forêts (northern Alsace, France) (Fig. 1) project was the frst Enhanced Geothermal System (EGS) developed in the world (Baria et al. 1999; Genter et al. 2010; Gérard et al. 2006; Kappelmeyer 1991). The geology at the Soultz-sous-Forêts site is well known thanks to an old exploratory oil borehole, EPS-1, that was deepened for the needs of geothermal exploration and fully cored from 933 (middle *Muschelkalk*) to 2227 m (Paleozoic basement) (Aichholzer et al. 2019; Dezayes et al. 2005) e.g., in the zone where convection predominates (Genter and Traineau 1992). The Permian and Triassic sedimentary rocks of the *Buntsandstein* are described in Aichholzer et al. (2016, 2019) and Vidal et al. (2015) and physical properties of the EPS-1 cores studied by Géraud et al. (2010), Grifths et al. (2016), Hafen (2012), Hafen et al. (2013, 2017), Heap et al. (2017, 2018, 2019), Kushnir et al. (2018), Surma and Geraud

(2003), Schellschmidt and Clauser (1996), Flores Marquez 1992 and Schellschmidt and Schulz (1992).

In total, four boreholes have been drilled at the Soultz-sous-Forêts site: GPK-1, GPK-2, $GPK-3$, and $GPK-4$, the last three of which extend to a depth of 5 km. The stratigraphy of the GPK-1 and 2 boreholes was described in detail by Aichholzer et al. (2016) and is composed of 1400 m (measured depth, MD) of sedimentary cover. These boreholes have provided precious data including temperature profles that inform on regional heat transfer mechanisms (conduction and convection). At Soultz-sous-Forêts, the typical equilibrium-temperature profles measured in the 5-km-deep wells revealed a temperature of 200 °C at a depth of 5 km. Importantly, the temperature profle can be divided into three parts (Genter et al. 2010, 2015; Vidal et al. 2015). First, the uppermost part of the well (between 0 and 1 km-depth, down to the top of the *Muschelkalk* series), composed of sedimentary formations (Tertiary, Jurassic, and Upper Triassic), features a geothermal gradient of 110 $\rm{C/km}$, which, in the URG, indicates a conductive regime (Pribnow and Schellschmidt 2000). The section between a depth of 1 and 3.5 km, comprising the sedimentary rocks of the *Buntsandstein* series (down to a depth of about 1400 m) and fractured granitic basement, is characterized by a very low thermal gradient of 5 °C/km, interpreted as convective regime in the URG. Finally, the deepest part of the thermal profle, below a depth of 3.5 km, is composed exclusively of crystalline basement and features a thermal gradient of 30 °C/km, indicating another conduction zone. At the Soultz-sous-Forêts site, the previously estimated heat fow densities range between 140 and 150 mW/m² (Baillieux et al. 2013; Flores Marquez 1992; Pribnow and Clauser 2000; Le Carlier et al. 1994; Schellschmidt and Clauser 1996).

Case study: the Rittershofen geothermal site (France)

The Rittershoffen geothermal site, located 6 km southeast of Soultz-sous-Forêts (Fig. 1), is the second EGS project developed in northern Alsace (Baujard et al. 2017). To date, no heat fow density has been calculated for the Rittershofen geothermal site. However, a heat flow density of 121 mW/m² has been calculated for a nearby borehole (SAN 1 (SANDMUHLE 1) borehole, about 3.5 km from the GRT-1 well) (Lucazeau 2019). The drilled GRT-1 and GRT-2 boreholes extend to a depth of 2580 and 3196 m MD, respectively, and target the geothermal reservoir located at the sediment–basement interface. Temperature measurements in the wells indicate a constant thermal gradient of 87 °C/km (conduction) in the sedimentary cover between the surface and the top of the *Muschelkalk* (1661 m depth MD), and an average temperature of 160 °C at the top of the *Muschelkalk* cap rock (Baujard et al. 2017). The very low gradient (3° C/km) below this depth threshold indicates a convective regime in the *Muschelkalk* and *Buntsandstein* sedimentary formations, and in the fractured granitic basement.

Stratigraphy of the 2200-m-thick sedimentary cover in GRT-1 has been recently updated by Duringer et al. (2019) and correlated with stratigraphy in GPK-1 and GPK-2 in Aichholzer et al. (2016), which revealed several variations in the sedimentary cover between the two sites and even between the two wells at Soultz-sous-Forêts. Diferences include missing layers due to erosion, lateral thickness variations in sedimentary formations and thinning or absence of some formations due to faulting.

Methods

Selecting representative rock samples

We collected 45 samples (typically $15 \times 15 \times 15$ cm) from 27 different locations in the east of France (Alsace, Lorraine and Franche-Comté) and in Southwest Germany (Baden-Württemberg) (Figs. 1, 2, 3) that we consider provide a representative suite of rocks for the sedimentary cover of the URG. The choice of samples collected was guided by the formations encountered in the wells at Rittershofen (Duringer et al. 2019) and the wells at Soultz-sous-Forêts (Aichholzer et al. 2016). But for completeness, we sampled additional formations that are widespread in the region including, for instance, the *Grande Oolithe* [J-GO (Fig. 2)] formation, which is absent in the northern part of the URG due to erosion, but encountered to the south of Haguenau. We did not only sample the thickest formations, but also formations that represent outliers, in terms of lithology and texture, that may be characterized by very diferent thermal conductivities. Most samples were collected from rock outcrops, those not available at outcrops were collected at the potash mines in Wittelsheim (France) (O-ZSS samples) or sourced from drill cores from the DP202 borehole in Pulversheim (France) (O-CN and O-CM samples). We note that poorly consolidated formations were not sampled because it was not possible to prepare a sample suitable for measurement in the laboratory. The sampled

of the sampling site (see Fig. 1 for location on the map), nature of the sampled rock and location of the sampling site are indicated

Fig. 3 List of *Keuper*, *Muschelkalk* and *Buntsandstein* samples classed in stratigraphic order. Lithostratigraphic unit, number of the sampling site (see Fig. 1 for location on the map), nature of the sampled rock and location of the sampling site are indicated

formations that most closely represent those that could not be sampled were used as proxies.

Our method to estimate heat flow density is designed to be used at prospective geothermal sites for which there are no boreholes, therefore, in the absence of site-specifc rock samples. Although measurements on analogue samples should be used with care (Bauer et al. 2017), we have chosen to measure samples taken from rock outcrops for several reasons. First, when boreholes do exist (such as at Soultz-sous-Forêts and Rittershofen), the recovered rock cuttings are often too small to make reliable laboratory measurements. Further, despite having cores from exploration well EPS-1 (at the Soultzsous-Forêts site), these cores are only available from a depth of 933 m and therefore do not sample the conduction zone. Finally, not all regionally signifcant formations are encountered in the boreholes at Rittershofen and Soultz-sous-Forêts (e.g., such as the *Grande Oolithe,* J-GO). Therefore, the accurate estimation of heat flow density is reliant on the establishment of a comprehensive thermal conductivity database developed using analogue geological materials that are representative of the local and regional geology, including outcrop material.

Sample preparation

Each rock sample was cut and polished to obtain two planer and parallel surfaces (Fig. 4a). When possible, two pieces of sample for each studied formation were prepared. The thickness of the prepared samples was typically between 2.5 and 4 cm to allow a probing depth sufficient for the measurement and so that the samples could fit in the experimental jig (Fig. 4b, c). As a precaution, samples considered sensitive to water, such as the potash and marls, were cut and polished without using water to minimize water– mineral reactions that could afect the physical state of the samples.

Before characterization, all samples were vacuum-dried at 40 °C overnight.

Porosity measurements

The connected porosity of the samples was measured to inform on the relationship between thermal conductivity and porosity. We used the triple-weight water saturation method (Guéguen and Palciauskas 1992). The dry samples were vacuum-saturated with water and the saturated mass $\left(m_{_{\rm W}}\right)$ and the immersed saturated mass $\left(m_{i}\right)$ of the samples were measured. The samples were then vacuum-dried again and the dry mass (m_d) of each sample was measured. The connected water porosity, ϕ , was calculated using the following equation (units: unitless, value given as %):

$$
\phi = \frac{m_w - m_d}{m_w - m_i}.\tag{1}
$$

The porosities of the gypsum and the marl samples were not assessed for the samples on which thermal conductivity measurements were made; a separate sample set was dedicated to porosity measurements for these rocks. Although the thermal conductivities of these samples were measured in the wet condition (explained below), we wanted to avoid wet–dry cycles that may have infuenced the physical state of these samples prior to measuring their thermal conductivity. The porosity of the potash samples could not be measured due to their dissolution upon exposure to water.

Thermal conductivity measurements

The thermal conductivity measurements were conducted in the laboratory using a thermal analyzer (model TPS 500 by Hot Disk) based on the Transient Plane Source (TPS) method with a Kapton-plastic coated sensor (Gustafsson 1991; Gustavsson and Gustafsson 2005).

All the measurements were carried out at ambient pressure. The measurements were frst conducted on oven-dry samples at room temperature ("dry"), then on water-saturated samples at room temperature ("wet") and, fnally, on oven-dry samples at high temperature ("hot"; up to 160 °C). Samples could not be measured in the saturated state at high temperature nor at higher pressure conditions due to equipment constraints. The dry, wet, and hot measurements were all carried out on the same suite of rock samples. Whenever bedding was visible, the measurements were conducted such that the bedding was parallel to the sensor. Anisotropy of the samples could not be studied since the thermal analyzer used for the measurements averages the thermal properties of the investigated volume of rock.

Dry measurements

The dry measurements were carried out first because they are quick to perform and there is no risk of damaging the rock samples. To perform a measurement, the prepared, planer surfaces of two pieces of rock sample were placed either side of the sensor and the samples clamped together using a metal screw at the top of the jig, ensuring that the sample surfaces were fush with the sensor (Fig. 4b, c). During the measurement, an electrical current was passed through the sensor for a specifed duration to increase sample temperature; the sensor recorded the temperature increase of the sample as a function of time. The Hot Disk software used these data to determine the thermal conductivity and thermal difusivity (the specifc heat capacity was calculated using these measured data). The temperature of the sample before the measurement (monitored using a hand-held thermometer) was also used in the calculation performed by the software. The measurement parameters (output power and measurement duration) depended on the thermal properties of the sample. For our measurements, we used output powers between 35 and 440 mW and measurement durations between 2.5 and 40 s. We performed four measurements on each set of samples using the four permutations of sample confguration (since each sample has two plane parallel faces). The measurements were performed at least 5 min apart and once the sample had cooled back to room temperature following each measurement. For some formations, only one piece of sample could be prepared. To measure these samples, we used a piece of insulating foam of known thermal properties in place of the second piece of sample; the foam has a thermal conductivity, measured using the Hot Disk Analyzer, at least 10 times lower than that of the measured sample.

Wet measurements

We performed wet measurements on at least two samples per lithology (limestone, sandstone, dolostone, gypsum, and marl). The first sample was the sample closest to the mean dry thermal conductivity for that lithology. The second was the sample with either the lowest or highest dry thermal conductivity for a given lithology, depending on the distribution of dry conductivity for that lithology. Wet measurements on the marls could only be conducted on the K-MISm marls from the *Marnes Irisées supérieures* (K-MIS; *Keuper*) formation (Fig. 3), which remained coherent following saturation with water. The wet measurements were performed on samples vacuum-saturated with water using the same Hot Disk method described above. To ensure the samples remained water-saturated during the measurement, the entire sample jig was submerged in a water-flled container. Once the measurements were complete, the samples were air-dried and then vacuum-dried in an oven at 40 °C, as described above.

Hot measurements

The hot measurements were performed last in case exposure to high temperature resulted in any changes to the rock microstructure (such as thermal microcracking) or mineral assemblage (e.g., the dehydroxylation of clays). Although the marls and gypsums may have been afected by exposure to water during the wet measurements, we decided to perform the measurements in the same order for consistency. Hot measurements were performed on the same samples as used for the wet measurements. Additionally, the thermal conductivities of the O-ZSSpo potash and the J-MO (Fig. 2) marls, which could not be measured wet, were measured at high temperature. The hot measurements were performed at temperatures between 50 and 160 °C (Additional fle 1). The hot measurements were performed on oven-dry samples using the same Hot Disk method described above (using a sensor designed to withstand high temperatures). To perform the measurements, the entire sample jig was placed inside an oven. The temperature was then increased at a heating rate of $5 \degree C/\text{min}$ until the target temperature was reached. After the oven had reached the programmed temperature, the stability of the sample temperature was monitored using the dedicated function of the Hot Disk and the measurements were only performed once the sample was at thermal equilibrium with temperature in the oven. A waiting time of at least 30 min between two consecutive measurements was used for the hot measurements.

The chosen target temperatures for each lithology depended on the temperature expected for that lithology in the subsurface and based on temperatures measured in the GPK-2 (Soultz-sous-Forêts) (Genter et al. 2010) and GRT-1 (Rittershofen) wells (Baujard et al. 2017). For the formations not encountered in these wells, such as the *Grande Oolithe* (J-GO), the target temperatures were chosen based on the likely temperatures at the expected depths for these formations.

Thermal conductivity results

The measured thermal conductivities ("dry" for all samples and "wet" and "hot" for select samples) and connected porosities are given in Additional file 1. The connected porosity of our samples varies between 5 and 18% for the dolostones, between 0.5 and 22% for the limestones, between 2 and 36% for the sandstones, between 14 and 44% for the marls, and between 1 and 4% for the gypsum (Additional fle 1; Fig. 5).

Thermal diffusivity and heat capacity of the samples are given in Additional file 1, but will not be discussed in this study.

Dry‑state samples at ambient temperature

Dry thermal conductivity is plotted as a function of porosity in Fig. 6. This figure shows that dry thermal conductivity decreases as porosity is increased. In our dataset, the range of thermal conductivity for a given porosity increased as porosity decreased. Despite the trend of decreasing thermal conductivity with increasing porosity, thermal conductivity also varies as a function of lithology. For example, the thermal conductivities of sandstone, limestone, and gypsum of similar porosities ($\phi \sim 2\%$) are ~ 4 , ~ 2.5 to 3, and \sim 1.5 W/mK, respectively. For the low-porosity samples (<20%), our results suggest

yellow polygon indicates the general trend of evolution of conductivity as a function of porosity

that the dolostones show the highest thermal conductivities, followed by the sandstones, the limestones and marls, and finally the gypsums. The dry thermal conductivity of the potash from the O-ZSS formation (not shown in Fig. 6) is 5.25 W/mK (Additional fle 1), the highest dry thermal conductivity of the entire sample suite.

Wet‑state samples at ambient temperature

The wet thermal conductivities of the samples are compared with their corresponding dry thermal conductivities in Fig. 7a. The latter shows that the wet thermal conductivities of our sample suite are higher than their dry thermal conductivities for all but four of the samples. Only four samples (the M-GC shelly sandstone, the J-FSg calcareous sandstone, the limestone from the J-CE formation and the K-MISgy gypsum), which have low (ϕ < 10%) porosities, have thermal conductivities higher in the dry state than in the saturated state (Fig. 7a). We examine the variation in thermal

saturated conductivity values for the same sample and arrows indicate the direction of variation from dry to saturated. Colors indicate the lithology. **b** Efect of water saturation on thermal conductivity as a function of porosity of individual samples. Trendline and corresponding equation and coefficient of determination, R^2 , are given. **c** Comparison of thermal conductivities measured under dry state (λ_{div}) and water-saturated state (λ_{wet}). Trendline and corresponding equation and coefficient of determination, R², are given. The equation of the trendline is used for wet thermal conductivity estimates for samples measured only in the dry state

conductivity between the dry, λ_{dry} , and saturated, λ_{wet} , state by defining an "effect of water saturation", $(\lambda_{wet} - \lambda_{dry})/\lambda_{dry} \times 100$. The effect of water saturation is plotted as a function of porosity in Fig. 7b. This figure shows that the influence of water saturation on the thermal conductivity increases as porosity increases and that the data can be well described by a linear trend. Increases in thermal conductivity are small for low-porosity samples (less than 5%), high for high-porosity samples (up to 90%) and appear independent of lithology (Fig. 7b). We further note that, regardless of lithology, the thermal conductivity of the saturated samples is linearly related to their dry thermal conductivities (Fig. 7c, samples with thermal conductivity higher in the dry state than in the wet state are not included).

Dry‑state samples at high temperature

Thermal conductivity is plotted as a function of temperature in Fig. 8, which shows that, for all lithologies, thermal conductivity decreases with increasing temperature.

graphs for clarity

Thermal conductivity as a function of temperature is well described by a linear trend (with coefficients of determination, R^2 , above 0.91; see Additional file 2), apart from gypsum sample K-MISgy above 80 °C (R^2 = 0.34; see Additional file 2). Indeed, the K-MISgy gypsum sample shows a decrease similar to that of the other samples below 80 °C but reveals a sharp decrease in thermal conductivity above that limit, from 1.30 W/mK at 80 °C to 0.36 W/mK at 100 °C. Thermal conductivity of the sample slightly increases above 100 °C.

The thermal conductivities of the samples with the lowest thermal conductivities at ambient temperature appear to be infuenced less by an increase in temperature. For example, the high-conductivity O-ZSSpo potash sample is more afected (12% conductivity decrease) by a 43 °C temperature increase than the low-thermal-conductivity K-GR sandstone sample is by a 121 °C temperature increase (4% conductivity decrease).

To emphasize the infuence of the dry thermal conductivity, the slopes of the linear trends of thermal conductivity as a function of temperature (see Additional fle 2 for equations) are plotted as a function of thermal conductivity at ambient temperature in Fig. 9. The K-MISgy gypsum sample is not represented in this graph due to its atypical temperature dependence. This figure shows that the influence of temperature on the thermal conductivity of our sample suite depends on the dry thermal conductivity and that the data can be well described by a linear trend.

Discussion

The large thermal conductivity and porosity ranges measured in this study suggest that these values are the result of interaction between several parameters. The variability in measured porosities, from near-zero to 44% depending on lithology, indicates that no characteristic value exists for a given lithology. Therefore, a precise measurement of the porosity of all rocks is required to properly quantify the infuence of this property on thermal conductivity of rocks.

Thermal conductivity

Dry‑state samples at ambient temperature

Thermal conductivity ranges are large for each rock type, highlighting that thermal conductivity does not only depend on lithology and there is no characteristic conductivity for a given lithology.

The measurements allowed us to observe a first-order relation between the thermal conductivities of the samples and their porosities for all lithologies: thermal conductivity decreases with increasing porosity as observed in previous studies (Popov et al. 2003; Nagaraju and Roy 2014; Guo et al. 2017; Mielke et al. 2017).

As shown by Fig. 5, the range of thermal conductivity values is small for porous rocks (φ>25%), regardless of lithology and, thus, composition, which indicates that porosity is a predominant factor controlling thermal conductivity of porous rocks. On the contrary, for less porous rocks (ϕ < 14%), conductivity values show more variability and depend more on the lithology, which suggests a higher efect of mineralogical composition of the sample.

Saturated‑state samples at ambient temperature

Many studies have shown that measurements on samples in the dry state often underestimate thermal conductivity of porous rocks due to the low conductivity of air in the pores (Nagaraju and Roy 2014). Furthermore, measurements on saturated samples are often more representative of the in situ conditions since the pore volume is generally flled with water or other liquids that have higher thermal conductivities than air $(λ_{water}=0.6 W/mK)$ (Nagaraju and Roy 2014; Vosteen and Schellschmidt 2003). The results show an influence of saturation for all the investigated samples. Thermal conductivity in the saturated state is generally higher than in the dry state (Fig. 7a). For water-saturated samples, the contrast in thermal conductivity between the rock matrix and the fuid in the pores is smaller than for dry rocks, hence the observed trend for the sedimentary rock samples of this study. However, this trend does not apply to the M-GC shelly sandstone (*Grès Coquillier, Muschelkalk*), the J-FSg calcareous sandstone (*Formation de Schalkendorf*, Jurassic), the J-CE limestone (*Calcaire d'Ettendorf*, Jurassic) and to the K-MISgy gypsum (*Marnes Irisées supérieures*, *Keuper*). These samples show a low porosity (ϕ < 10%) and thermal conductivity higher in the dry state than in the saturated state as observed in previous studies by Nagaraju and Roy (2014) and Albert et al. (2017) for sedimentary rocks with porosity smaller than 3%. According to Albert et al. (2017), some rocks react to the fuid used for saturation, resulting in dissolution of some mineral and/or modifcation of the structure of the rock matrix of the sample and thus of its thermal conductivity. It is plausible that this phenomenon could have impacted our

samples. The very low porosity of these rocks, and therefore its very low influence of the pore space on conductivity, would have allowed us to observe the decrease in thermal conductivity of the rock matrix due to saturation. For more porous samples, the efect of porosity on conductivity is more signifcant and could have masked the impact of interaction between water and the rock matrix.

As shown by Fig. 7b, the effect of water saturation strongly depends on the porosity of the sample and very little on the sample's mineralogy. This relationship has already been observed in sedimentary rocks by Popov et al. (2003) and Nagaraju and Roy (2014).

However, thermal conductivity measured on saturated samples could not be correlated with the porosity of the samples. According to Popov et al. (2003), this can be explained by the variability of thermal conductivity of the rock matrix of the studied samples and in particular by the infuence of the clay content. In the saturated state, the contrast between thermal conductivity of the matrix and that of the fuid flling the pores is smaller than for dry rocks. It is thus possible that the infuence of composition of the rock matrix on thermal conductivity in the saturated state is more important than that of porosity.

Finally, thermal conductivity of the saturated samples is linearly proportional to thermal conductivity of the dry samples (Fig. 7c): thermal conductivity in the saturated state increases with increasing conductivity in the dry state, as observed by Guo et al. (2017) for sandstones. This relationship allows us to estimate conductivity of a sample in the water-saturated state at ambient temperature using the dry-state measurement at room temperature (Eq. 8).

Dry‑state samples at high temperature

Many studies have shown that thermal conductivity of dry rocks tends to decrease with increasing temperature (between −80 and 100 °C by Guo et al. 2017; in the temperature range from 25 to 500 °C by Kant et al. 2017 and between 0 and 300 °C by Vosteen and Schellschmidt 2003). According to Guo et al. (2017), temperature is one of the factors which has the most impact on thermal conductivity of a solid. These studies also showed that the decrease in thermal conductivity with increasing temperature is nonlinear. Therefore, performing measurements at relevant in situ temperatures is essential and the experimental conditions should be as close as possible to in situ conditions. Tis choice is even more important since, in this study, we cannot perform measurements on water-saturated samples at high temperature nor can we study the efect of pressure (closer to in situ conditions) due to equipmental constraints.

The results of high-temperature measurements show an approximate negative linear correlation between thermal conductivity and increasing temperature (Fig. 8 and Additional fle 2). It should, however, be emphasized that the temperature ranges chosen for this study (between 18 and 160 °C) are less extensive than the ones considered for the studies previously mentioned. These trends can be used to interpolate/extrapolate thermal conductivity at a given temperature for these dry samples and those for which they are used as proxies.

We fnd that thermal conductivity of dry rocks decreases with increasing temperature, as shown previously by Kant et al (2017), Guo et al. (2017), Vosteen and Schellschmidt (2003) and Clauser and Huenges (1995). Since, at ambient pressure, the thermal conductivity of air only increases from 0.025 to 0.036 W/mK as temperature is increased from 20 to 150 °C (Kadoya et al. 1985), we attribute this decrease to changes in the thermal conductivity of the rock matrix with increasing temperature. We also cannot rule out decreases in thermal conductivity as a result of increases in porosity due to thermal microcracking (Chaki et al. 2008; Grifths et al. 2018; Kant et al. 2017) and/or chemical reactions such as the dehydroxylation of clays (Earnest 1991a, b; Mollo et al. 2011), especially at temperatures in excess of $100 \degree C$. The sharp decrease in thermal conductivity of the K-MISgy gypsum above 80 °C can be easily explained by the progressive transformation of gypsum (CaSO₄ 2H₂O) to bassanite (hemihydrate CaSO₄ ½H₂O) (i.e., partial dehydration of gypsum) (Sadeghiamirshahidi and Vitton 2019). The slight increase in thermal conductivity with increasing temperature above this limit can be caused by transformation of the small amount of gypsum that had been left un-transformed in the sample into bassanite.

Yet we observe that some samples are more affected by temperature increase. Thermal conductivity of the high-conductivity O-ZSSpo potash decreases more than that of the K-GR sandstone despite a greater temperature increase for the latter. We propose that the higher the initial thermal conductivity measured on a dry sample at room temperature, the more it is afected by temperature increase (Figs. 8 and 9) as observed by Vosteen and Schellschmidt (2003). Tis relationship, which can be satisfactorily characterized by a linear trendline $(R^2=0.87)$, could be used to calculate an estimated thermal conductivity at a given temperature using the dry-state measurements at room temperature (Eq. 5).

Heat fow density estimations

Methods for heat fow density estimations

Based on the various thermal conductivity measurement conditions, three types of heat fow density were calculated for each site: (1) dry rocks at ambient temperature ("dry" heat fow density); (2) dry rocks at in situ temperature ("hot" heat fow density), and (3) saturated rocks at ambient temperature ("wet" heat flow density).

For the dry heat fow density estimates, the thermal conductivities measured on the samples in the laboratory were considered without corrections for in situ temperature or water saturation.

For the hot heat fow density estimates, the thermal conductivities of the samples were either taken directly from the data (if the sample was measured at the in situ temperature) or the thermal conductivity at ambient temperature was corrected for the in situ temperature. To perform this correction, we interpolate/extrapolate the existing data using the empirical relationships defned in this study. For samples measured under dry, hot conditions, the decrease in thermal conductivity with temperature can be described by the following linear equation:

$$
\lambda(T) = aT + b,\tag{2}
$$

where $\lambda(T)$ is the thermal conductivity (in W/mK) at temperature T (in \degree C), and empirical constants a and b are given by the best-fit linear regression of the data provided in Fig. 8 (equations provided in Additional file 2). The a coefficient of each of the linear trendlines is plotted in Fig. 9 as a function of the dry thermal conductivity at ambient temperature, λ_{dry} , of the associated sample. The best-fit linear regression of this plot provides the following equation:

$$
a = -0.0026 \times \lambda_{\rm dry} + 0.0022. \tag{3}
$$

Thus giving:

$$
\lambda(T) = \left(-0.0026 \times \lambda_{\text{dry}} + 0.0022\right) \times T + b. \tag{4}
$$

The mean measuring temperature of the dry measurements is applied as a correction coefficient for T to average b as measured λ_{drv} . The thermal conductivity λ_{hot} at the in situ temperature $T_{\text{in situ}}$, can therefore be approximated from the dry thermal conductivity at ambient temperature, using the following relation:

$$
\lambda_{\text{hot}} = \left[\left(-0.0026 \times \lambda_{\text{dry}} \right) + 0.0022 \right] \times (T_{\text{in situ}} - 25) + \lambda_{\text{dry}}. \tag{5}
$$

For the gypsum-bearing formations, two equations (corresponding to equations of the best-ft linear regressions, Fig. 8 and Additional fle 2) were considered to estimate hot thermal conductivity of the gypsum:

Below 80 °C:

$$
\lambda_{\text{hot}} = -0.0024 \times T_{\text{in situ}} + 1.4852. \tag{6}
$$

Above 80 °C:

$$
\lambda_{\text{hot}} = 0.001 \times T_{\text{in situ}} + 0.2337. \tag{7}
$$

For the wet heat fow density estimates, the thermal conductivities of the samples were either taken directly from the data (if the sample was measured in the saturated state) or the thermal conductivity at ambient temperature was corrected for water saturation using the relationship defined by all our wet data (Fig. 7c). The wet thermal conductivity, λ_{wet} (in W/mK), can be estimated from the dry thermal conductivity at ambient temperature using the following empirical relationship (Fig. 7c):

$$
\lambda_{\text{wet}} = (0.8496 \times \lambda_{\text{dry}}) + 0.7524. \tag{8}
$$

Samples with a thermal conductivity higher in the dry state than in the water-saturated state are not included since thermal conductivity is expected to increase with water saturation.

For the three types of heat fow density, a mean thermal conductivity was calculated for each sedimentary formation observed in the borehole. The composition of the formations (proportions of sandstone, marls, dolostone, etc.) was estimated based on the GRT-1 masterlog (Duringer and Orciani (2015), internal report). The mean thermal conductivity of the formation, λ_{form} , was calculated as the weighted harmonic mean of the thermal conductivities of the samples composing the formation (Beardsmore et al. 2001).

For the formations that were not sampled, the mean thermal conductivity of a formation with similar geological characteristics was used.

The three types of heat flow density were calculated using the equilibrated thermal profles measured in the wells (Baujard et al. (2017) for Rittershofen and Genter et al. (2010) for Soultz-sous-Forêts) and the mean thermal conductivities of the formations λ_{form} . The depths of the limits (top $z_{\text{top form}}$ and bottom $z_{\text{bot form}}$) of the formations were taken from Duringer et al. (2019) for the Rittershofen site and from Aichholzer et al. (2016) for the Soultz-sous-Forêts site. The heat flow densities were calculated for each formation between the surface and the top of the *Muschelkalk* using Fourier's law:

$$
q_{\text{form}} = \lambda_{\text{form}} \times \frac{T_{\text{bot form}} - T_{\text{top form}}}{z_{\text{bot form}} - z_{\text{top form}}},
$$
\n(9)

where $q_{\rm form}$ is the heat flow density of the formation (in W/m²), $\lambda_{\rm form}$ is the mean thermal conductivity of the sedimentary formation (in W/mK), and $T_{\text{top form}}$ and $T_{\text{bot form}}$ are the temperatures measured in the well at the top and bottom of the formation, respectively.

For all sites, we define mean heat flow densities for four intervals (1) the whole sedimentary cover in the conduction zone (between the surface and the top of the *Muschelkalk*, "global" heat fow density), considered as the "equilibrium" heat fow density and for the (2) Tertiary, (3) Jurassic and (4) the *Keuper* intervals to study the evolution of heat flow density with depth.

For each interval, a mean thermal conductivity $\lambda_{\text{interval}}$ (in W/mK) was calculated as the harmonic mean of the thermal conductivities of the formations within the interval weighted by the thickness of the formations. Tis mean thermal conductivity was used to calculate $\overline{q_{\text{interval}}}$ the mean heat flow density (in W/m²) of the considered interval, such as:

$$
\overline{q_{\text{interval}}} = \overline{\lambda_{\text{interval}}} \times \frac{T_{\text{bot interval}} - T_{\text{top interval}}}{z_{\text{bot interval}} - z_{\text{top interval}}},
$$
\n(10)

where $T_{\text{bot interval}}$ and $T_{\text{top interval}}$ are the temperatures (in \degree C) at the top and bottom of the interval, respectively, and $z_{bot interval}$ and $z_{top interval}$ are the depths (in m) of the top and bottom limits of the considered interval, respectively. A detailed description of the equations and error calculation are provided in Additional fle 3.

The first meters of the boreholes were filled with air when the equilibrium-temperature profiles were assessed in the geothermal wells. Therefore, the temperature was considered only where measurements were performed in brine, from depths of 50 and 40.3 meters in GPK-1 and GPK-2, respectively. For GRT-1, the frst 149.8 m were not included in the heat flow density calculations.

Estimated heat fow densities

Conductive heat fow density was calculated for each geothermal well (GPK-1 and GPK-2 in Soultz-sous-Forêts and GRT-1 in Rittershofen) from the equilibrium-temperature profles measured in the wells and from the thermal conductivities measured on (1) dry rocks at ambient temperature ("dry"), (2) dry rock at in situ temperature ("hot") and (3) saturated rocks at ambient temperature ("wet"). The results are

presented in Figs. 10, 11 and 12 for GPK-1, GPK-2 and GRT-1, respectively. All the mean values are compiled in Table 1.

The temperature gradient is higher $(3-23%)$ than the global gradient (mean temperature gradient of the whole sedimentary column, between the surface and the top

of the *Muschelkalk*) in the Tertiary in GRT-1 and in the Jurassic in all the wells. Gradients in the other intervals are lower (3–17%) than the global gradients.

For all the wells and all conditions considered, thermal conductivity in the Tertiary interval is always inferior (by 8%) or equal to the global thermal conductivity. The

hot thermal conductivities of the *Keuper* interval in GRT-1 and GPK-1 are also lower (2–4%) than the global thermal conductivity. Values for the other intervals are higher (4–37%) than the global thermal conductivity.

Well	Interval	Mean T $(^{\circ}C)$	$\Delta T/\Delta z$ $(^{\circ}C/100 \text{ m})$	Dry		Hot		Wet	
				Mean λ (W/ mK)	Mean q (mW/m ²)	Mean λ (W/ mK)	Mean q (mW/m ²)	Mean λ (W/ mK)	Mean q (mW/ m ²
GRT-1	Global	92.54	9.07	1.35 ± 0.20	122 ± 19	1.20 ± 0.18	109 ± 18	1.81 ± 0.19	164 ± 20
	Tertiary	71.59	9.32	1.25 ± 0.24	$117 + 24$	1.20 ± 0.24	112 ± 24	1.71 ± 0.23	$159 + 24$
	Jurassic	132.30	9.69	1.49 ± 0.04	144 ± 18	1.22 ± 0.05	118 ± 16	1.90 ± 0.05	184 ± 22
	Keuper	153.25	7.03	1.77 ± 0.07	$125 + 31$	1.15 ± 0.07	81 ± 22	2.27 ± 0.08	$160 + 39$
GPK-1	Global	64.49	10.13	1.40 ± 0.24	141 ± 26	1.25 ± 0.21	126 ± 23	1.85 ± 0.23	$188 + 26$
	Tertiary	46.02	10.08	1.25 ± 0.30	126 ± 32	1.23 ± 0.30	124 ± 32	1.71 ± 0.29	172 ± 32
	Jurassic	83.07	12.46	1.50 ± 0.05	$187 + 22$	1.32 ± 0.06	165 ± 21	1.91 ± 0.05	238 ± 25
	Keuper	101.54	8.37	1.85 ± 0.06	155 ± 24	1.22 ± 0.06	102 ± 17	2.32 ± 0.09	$194 + 31$
GPK-2	Global	62.34	10.14	1.33 ± 0.26	135 ± 29	1.26 ± 0.25	$127 + 27$	1.78 ± 0.25	181 ± 28
	Tertiary	49.00	9.78	1.25 ± 0.31	123 ± 33	1.23 ± 0.32	120 ± 33	1.71 ± 0.31	$167 + 33$
	Jurassic	87.60	11.82	1.50 ± 0.05	177 ± 21	1.32 ± 0.06	$157 + 20$	1.90 ± 0.05	225 ± 25
	Keuper	100.95	8.81	1.82 ± 0.10	160 ± 63	1.31 ± 0.07	$115 + 45$	2.26 ± 0.12	$199 + 77$

Table 1 Mean calculated thermal conductivities *λ* **and heat fow densities q for the various intervals in the GRT-1, GPK-1 and GPK-2 geothermal wells**

Mean measured temperatures and temperature gradients are given for the various intervals [global (from surface to top of the *Muschelkalk*), Tertiary, Jurassic and *Keuper*]. Data for dry rocks at ambient temperature (dry), dry rocks at in situ temperature (hot) and saturated rocks at ambient temperature (wet)

In the GRT-1 well, the calculated mean global heat flow densities are 122 mW/m² $(\pm 16\%)$, 109 mW/m² ($\pm 16\%$) and 164 mW/m² ($\pm 12\%$) for dry, hot and wet rocks, respectively. In GPK-1, the global heat flow densities are $141 \text{ mW/m}^2 \ (\pm 18\%)$, 126 mW/m² (\pm 18%) and 188 mW/m² (\pm 14%) for dry, hot and wet rocks, respectively. In GPK-2, the global heat flow densities are 135 mW/m² (\pm 21%), 127 mW/m² (\pm 21%) and 181 mW/m² (\pm 16%), for dry, hot and wet rocks, respectively.

Heat fow density calculated for the Jurassic in all the wells and for all conditions is higher $(9-32%)$ than the global heat flow density. The same is observed for the hot heat fow density in the Tertiary (3%) and dry heat fow density in the *Keuper* at Rittershofen (2% higher than global heat fow density) and for the dry and wet heat fow densities in the *Keuper* for the GPK-1 and GPK-2 wells (3–10% higher). Heat fow densities calculated for all the other intervals are lower (2–26%) than global heat fow density.

In summary, at both sites and under all measurement conditions considered, the mean heat fow densities calculated for the Tertiary always fall within the range of variability of the mean global heat flow densities. The same applies for the *Keuper* in GRT-1 in the dry and wet conditions and for the mean hot and wet heat fow densities in the Jurassic. The mean value calculated for the Jurassic at Rittershoffen for dry rocks is too high to be in the range of variability of the global dry heat fow density. Hot heat fow density in the *Keuper* is too low to be in the range of variability of the hot mean value.

At Soultz-sous-Forêts, the mean heat fow densities calculated in the Jurassic are systematically out of the interval of the global heat flow densities in GPK-1 and GPK-2. The hot heat fow density calculated in the *Keuper* in GPK-1 is also too low to be within the range of variability of the hot global value. The other values are within the interval of confidence of the global heat flows.

Infuence of the thermal‑conductivity‑measurement conditions

The dry heat flow densities calculated using the new thermal conductivities values measured in this study on samples from the URG are similar to the previously published heat fow densities for the Soultz-sous-Forêts site and close to Rittershofen. The heat flow at Soultz-sous-Forêts was estimated in previous studies to be between 140 and 150 mW/m² (Flores Marquez 1992; Le Carlier et al. 1994; Schellschmidt and Clauser 1996; Pribnow and Clauser 2000; Baillieux et al. 2013) and we estimate here heat flow densities of 141 ± 18 mW/m² for GPK-1 (Fig. 10 and Table 1) and 135 ± 21 mW/m^2 for GPK-2 (Fig. 11 and Table 1). The heat flow density estimated near the Rittershoffen site, 121 mW/m², compares well with our estimated value of 122 ± 16 mW/m² for GRT-1 (Fig. 12 and Table 1).

Our results also show that heat fow density depends strongly on thermal conductivity measurement conditions. Heat fow densities calculated using the thermal conductivities of water-saturated rocks at ambient temperature can be up to 43% higher than heat fow densities calculated for dry rocks (dry rocks at ambient temperature) (Table 1), and heat fow densities calculated using values measured on hot rocks (dry rocks at in situ temperature) can be up to 11% lower than "dry" heat fow densities (Table 1). Since the "dry" rock scenario is likely the least realistic, these results probably do not represent the true heat fow density values. Further, dry heat fow densities were already discussed for Soultz-sous-Forêts in previous studies, henceforth we only discuss the hot and wet heat fow density models determined by our study.

It is still unclear whether the in situ rocks are saturated or not; verifying the rock saturation at depth in the geothermal wells is particularly complicated. We therefore cannot conclude on the validity of the wet heat fow densities but, as stated, the efect of saturation of the rocks on the calculated heat flow density should not be ignored. The effect of temperature increase down the borehole, however, is undeniable. Temperature increases with depth, and as shown before, thermal conductivity of rocks decreases with increasing temperature. Consequently, we cannot assert that considering hot rocks is more representative than considering wet rocks since it is very plausible that some formations are both hot and saturated while some others are still efectively dry at the in situ temperature conditions. Although the efect of pressure could not be accounted for, we conclude that the hot and wet heat fow densities calculated for the URG using the thermal conductivity data presented herein are more representative of the in situ conditions than the dry heat fow densities previously calculated.

Evolution of the heat fow density with depth

In a sedimentary basin in which heat transfer is mostly driven by conduction, heat fow density should remain approximately constant with depth (Andrews-Speed et al. 1984; Schütz et al. 2014). Therefore, examining the vertical heat flow density variations with depth at Soultz-sous-Forêts and Rittershofen should enable us to investigate conductive equilibrium down the geothermal wells. Heat fow density is considered constant if the intervals of confdence of the heat fow density values in the various intervals (Tertiary, Jurassic and *Keuper*) overlap those of the global heat fow densities (over the whole conduction zone). Variations for the dry heat fow densities will not be discussed here, because, as stated earlier, dry conditions were already investigated by previous studies (Baillieux et al. 2013; Flores Marquez 1992; Pribnow and Clauser 2000; Le Carlier et al. 1994; Schellschmidt and Clauser 1996; Pribnow and Schellschmidt 2000).

In the GRT-1 well, the error bars of the hot or wet heat fow densities over the Tertiary, Jurassic and *Keuper* intervals all overlap the interval of confdence of the calculated global hot or wet heat flow densities (Table 1 and Fig. 12). Thus, we consider the conductive heat fow density in GRT-1 to be constant for the whole sedimentary cover between the surface and the top of the *Muschelkalk*; equilibrium thermal conduction of heat controls the evolution of temperature with depth over this interval. At Soultz-sous-Forêts, with the exception of the wet value for the Jurassic in GPK-2, the error bars of the hot or wet heat fow densities over the Tertiary, the Jurassic and the *Keuper* intervals all overlap the confdence interval of the global hot or wet heat fow densities (Table 1 and Figs. 10, 11). However, the calculated absolute values of the Jurassic interval in the hot and wet conditions are systematically higher (2–11%) than the upper boundary of the global heat fow densities. Tis anomaly is only observed for the Soultz-sous-Forêts site, in both GPK-1 and GPK-2. Even though errors on the calculated heat fow densities values are large (about 12% in the Jurassic), the diference between the heat fow densities in the Jurassic and the global heat fow densities are probably signifcant.

It is necessary here to understand if this recurrent heat fow density anomaly in the Jurassic intersected by the Soultz-sous-Forêts wells is real or the result of a systematic error. The method used in this study is the victim of many sources of errors. First, it is possible that when temperature logs were performed in the wells, the systems were not at thermal equilibrium with the surrounding geological formations (Bullard 1947). The temperature log from GPK-1 used to calculate the heat fow density was measured in June 1997, more than 7 months after hydraulic circulation tests in the wells. In GPK-2, temperature was assessed in July 1999, 3 months after the end of the deepening drilling operations in the granitic basement, the sedimentary cover was already fully cased at that time. These periods without perturbations in the wells should have allowed temperature in the boreholes to equilibrate with the host rocks. Temperature logs in Soultzsous-Forêts are thus considered to be as close as possible to equilibrium (Genter et al. 2010).

Second, the local anomaly in the Jurassic could be induced by the rock sampling method. The temperature logs in Soultz-sous-Forêts indeed reveal a higher gradient in the Jurassic than in the Tertiary or in the *Keuper* intervals (Table 1 and Figs. 10, 11) (Pribnow and Schellschmidt 2000). Maintaining a constant heat fow density over the three intervals would require lower thermal conductivity values than those measured in the laboratory on the Jurassic samples. There are several possible sources of error due to sampling method. (1) Some formations cannot be cored in a way that allows thermal conductivity measurements, especially in the Jurassic, due to friability of the material. We therefore chose to sample analogue rocks from accessible outcrops, which can feature (thermal) properties diferent from that of the in situ rocks (Bauer et al. 2017). (2) Not all samples could be measured saturated or at in situ temperature; instead, the hot and wet thermal conductivities are interpolated/extrapolated using the here-defned empirical relationship (Figs. 7c, 9 respectively). These empirical relationships may introduce signifcant errors when estimating hot or wet thermal conductivities and more measurements are required to further refne these equations. Further, the efect of pressure could not be studied on our samples. Based on previous studies (Walsh and Decker 1966; Abdulagatova et al. 2009; Schön and Dasgupta 2015), we would expect pressure to reduce porosity and close microcracks and thus increase thermal conductivity, but the extent of that efect coupled to that of temperature and saturation remain to be quantifed for the rocks of the URG. (3) For formations that could not be sampled, the sampled formations were used as proxies. For example, the J-MO sample was sourced from a quarry and was used as an analogue for almost all the marls from the Jurassic; this choice to use an analogue rock may induce errors in thermal conductivity estimates. Yet, we note that the thermal conductivity of the marls at the base of the Jurassic (J-CMGm, *Calcaires et Marnes à Gryphées*) is not very diferent from that of the J-MO marls (Additional fle 1). We thus consider that the use of these analogues materials as a proxy for all the marls of the Jurassic can be justifed.

Tird, the composition of the formations (proportions of lithology) was estimated from the GRT-1 masterlog (Duringer and Orciani (2015) , internal report). The masterlog itself can bear errors if part of the material in the rock cuttings was lost during cleaning (for example, a greater proportion of marls in the in situ formation than observed in the washed cuttings). Also, the masterlog from the Rittershofen well might not be completely applicable to the Soultz-sous-Forêts geology due to lateral variation of facies in the formations (Fuchs 2018). However, it is very unlikely that facies of the Triassic and Jurassic formations drastically change over 6 km of horizontal distance (between Rittershofen and Soultz-sous-Forêts) as shown by Aichholzer et al. (2016).

While direct downhole measurements of thermal conductivity under in situ conditions are the most applicable and would allow us to verify the reliability of the values measured on outcrop samples, such measurements are rare and more expensive than laboratory measurements (Garibaldi 2010; Harcouët 2005).

Further, paleoclimatic and/or topographic corrections are usually required for heat fow density estimates (Beck 1977; Blackwell et al. 1980; Guillou-Frottier et al. 1998; Jaeger and Sass 1963; Lucazeau and Vasseur 1989; Luijendijk et al. 2011; Majorowicz and Wybraniec 2011; Mareschal et al. 2000; Vasseur 1980) and uncorrected data can yield errors in estimated heat flow densities.

Nevertheless, the gradient in the Jurassic interval of the Soultz-sous-Forêts wells is noticeably higher than in the Rittershoffen well, which leads us to believe that this anomaly cannot only be induced by variations in rock thermal conductivities between the two sites. If the methods are not a source of error, then the heat fow density anomaly observed in the Jurassic interval at Soultz-sous-Forêts could be explained by at least four processes: radiogenic heat production, non-equilibrium conduction of heat, heat fow density refraction or transfer of heat by nonconductive processes.

Radiogenic heat production can contribute signifcantly to surface heat fow density in sedimentary basins (Frone et al. 2015; Guillou-Frottier et al. 2010; Schütz et al. 2012; Waples 2002). In sediments, radiogenic heat production can be high in some shales especially in those rich in organic matter, like in the black shales of the Toarcian (Jurassic) encountered in the URG (Böcker et al. 2017; Böcker and Littke 2016; Waples 2002). Radiogenic heat production can be estimated from gamma-ray (GR) logs performed down boreholes (Bücker and Rybach 1996; Waples 2002). Gamma-ray logging is the continuous quantitative measurement of the natural radioactivity of the formations crossed by a borehole (Serra and Serra 2000). These logs can be performed during or after drilling operations in a tubed or open hole, however the signal (unit: gAPI) measured through casing is attenuated compared to a signal measured in an uncased borehole. GR logs in the GPK-1, GPK-2 and GRT-1 wells are presented in Figs. 10, 11 and 12, respectively (Aichholzer et al. 2016; Duringer et al. 2019). In GPK-1, the GR value in the Tertiary is about 85 gAPI, 100 gAPI in the Jurassic and 81 gAPI in the *Keuper*. GR was measured through casing in the GPK-2 well in the Tertiary until about 575 m and is therefore attenuated by the casing and will not be taken into account for comparison. The GR value for the Jurassic in GPK-2 is about 97 gAPI and about 72 gAPI in the *Keuper*. In GRT-1, the mean GR value in the uncased Tertiary rocks is 80 gAPI and radioactivity is higher in the Jurassic (around 118 gAPI) and in the *Keuper* (92 gAPI, on average). The natural radioactivity in all three wells is highest in the Jurassic, but not high enough to explain the heat fow density anomaly observed at Soultz-sous-Forêts. Further, if radiogenic heat production was the only source of the locally higher heat fow density, the same anomaly would be observed in Rittershofen. Tus, while natural radioactivity in the Jurassic formations may contribute to enhancing the thermal gradient over this interval of the borehole, it cannot account for the entire thermal anomaly.

According to Andrews-Speed et al. (1984), conductive thermal equilibrium may not be maintained in a sedimentary basin due to rapid sedimentation and subsidence or to uplift and erosion episodes. High sedimentation rates (\gg 1 mm/year) can prevent thermal equilibrium between the deposited sediments and the bedrock (England et al. 1980). Rapid sedimentation can thus lead to a transient temperature gradient and heat fow density in the sedimentary sequence until the cold upper sediments warm up and equilibrate with the hot bedrock (Andrews-Speed et al. 1984). In the URG, sedimentation rates during the Tertiary are less than 1 mm/year and so the Tertiary and Jurassic sequences should be at thermal equilibrium. Despite this, an episode of uplift and erosion can enhance the conductive heat flow density in the near-surface layers by bringing up deep hot material (Andrews-Speed et al. 1984; England et al. 1980). However, in the URG, the most recent episode of uplift is not recent enough to have a continued infuence on conductive heat fow density (Sissingh 1998). Moreover, if uplift and erosion were responsible for the heat fow density anomaly, the heat fow density in the Tertiary would also be afected, which is not the case.

Heat refraction can occur in the case of a thermal conductivity contrast between geological structures (Andrews-Speed et al. 1984; England et al. 1980; Fulton and Safer 2009; Guillou-Frottier et al. 1996, 2010; Lee and Henyey 1974). In faulted areas, as in the URG, where rock types of diferent thermal conductivities are brought side by side, refraction of heat can occur and temperature and heat fow density can locally increase towards the most conductive material (Andrews-Speed et al. 1984; Lee and Henyey 1974; Wang et al. 1985). The ratio between thermal conductivities of theoretical mediums 1 and 2 can be expected to be equal to the corresponding heat flow density ratio ($\lambda_1/\lambda_2 = q_1/q_2$) (Lee and Henyey 1974). The Soultz-sous-Forêts wells are located near two major faults (the Kutzenhausen and Soultz-sous-Forêts

faults, Fig. 13) and cross several faults in the sedimentary cover (Figs. 10 and 11). The Kutzenhausen major fault, imaged using seismic methods and located near the GPK-1 well, has 330 m of cumulated ofset (Aichholzer et al. 2016) and juxtaposes the Jurassic and the Tertiary sediments (Baujard et al. 2017; Place et al. 2010; Vidal et al. 2015). The Jurassic sediments have a higher thermal conductivity than the Tertiary sediments, thus a higher heat fow density can be expected in the Jurassic on the eastern side of the fault. The displacement along the major faults near the Rittershoffen well are not as large as at Soultz-sous-Forêts (Baujard et al. 2017) and GRT-1 only crosses faults in the basement (Duringer et al. 2019), which could imply no or very little heat refraction near the GRT-1 well. A 2D or 3D study of the vertical heat fow density near the Soultz-sous-Forêts and Rittershofen areas could verify if heat refraction occurs on either side of these major faults. However, the contrast in thermal conductivities between the Tertiary and the Jurassic might not be large enough to fully explain the Jurassic anomaly.

Finally, nonconductive heat transfer might be responsible for the Jurassic temperature anomaly. The GPK wells cross many faults in the sedimentary cover of which some are open fractures, particularly in the Jurassic (Aichholzer et al. 2016). Indeed, the GPK-1 well crosses a fracture zone at 639 m below surface with fractures with apertures on the order of millimeters (oil was detected within these fractures). The GPK-2 well crosses a fracture zone accompanied by a natural gas anomaly at 686 m and the Soultz-sous-Forêts major fault, also associated with a gas anomaly, at 708 m. If fuid migration (such as water or hydrocarbons) through these fractures occurred, or still occurs, and if the rates of migration exceed a few millimeters per year, the circulation could have induced a local thermal perturbation where the fractures cross the sedimentary formations, generating discontinuities in the heat fow density profles (Andrews-Speed et al. 1984; Guillou-Frottier et al. 2010). If these fractures reach the basement (which is the case for the Kutzenhausen major fault), the thermal efect of fuid circulation in the faults could be non-negligible (Waples 2002). Further, Le Carlier et al. (1994) have identifed the major Soultz-sous-Forêts and the Kutzenhausen faults and the Jurassic formations at Soultz-sous-Forêts as possible fuid pathways. Even if, at present, circulation is not ongoing but stopped recently, the system could be in a transient relaxation phase and may not yet have reached thermal equilibrium and the efect could still be observed in the vertical heat fow density profles.

We consider nonconductive heat transfer as the process most likely to cause the heat fow density anomaly in the Soultz-sous-Forêts wells. Indeed, faults in the sedimentary cover are only crossed in the GPK wells where the heat fow density anomaly is observed. Faults in the Rittershoffen well, where the vertical heat flow density can be considered stable, were only encountered in the basement.

In summary, a systematic error in estimating thermal conductivities of the formations still needs to be discounted and more measurements are required to refne the URG thermal properties database. The method used to calculate the heat flow densities does not include topographic or paleotemperature corrections, which could also result in error in heat fow density estimates. Yet the heat fow density anomaly observed in the Jurassic at the Soultz-sous-Forêts wells could be the result of a coupled infuence of a higher radiogenic heat production in the Jurassic, heat refraction due to the signifcant displacement caused by the major faults near the Soultz-sous-Forêts geothermal site, and local thermal disturbance caused by (paleo)circulation of hot fuids in the fracture zones crossed by the GPK wells.

Conclusions

Thermal conductivity and accurate heat flow densities estimates are essential thermal parameters in geothermal studies. This study shows an important influence of the experimental conditions when measuring the thermal properties of rocks. For dry rocks at ambient temperature, conductivity strongly depends on porosity and decreases with increasing porosity. An infuence of the mineralogical composition of the rock matrix is also noticeable but is less pronounced. Most samples have higher conductivities in the water-saturated state than in the dry state. Moreover, the efect of water saturation increases with increasing porosity and conductivity in the saturated state is proportional to conductivity in the dry state. Furthermore, results show that thermal conductivity of dry rocks decreases with increasing temperature. The decrease is higher for samples with high conductivity in the dry state at ambient temperature. If the efect of water saturation and temperature are not considered, heat fow density estimates can lead to unreliable values. Tus, a good characterization of thermal conductivities of sedimentary formations from the URG crossed by the deep geothermal boreholes combined with the temperature measurements in the wells can provide better estimates of the heat fow density.

The heat flow densities previously reported for the Soultz-sous-Forêts geothermal site relied on a limited dataset, often far from in situ conditions and, until present, no heat fow density has been estimated for the Rittershofen geothermal borehole. Heat fow density was calculated for both sites using the thermal properties of dry rocks at ambient temperature (dry), dry rocks at in situ temperature (hot) and saturated rocks at ambient temperature (wet). The estimated values illustrate the influence of experimental conditions. Dry heat fow density values calculated using the new thermal conductivity data are consistent with previously published heat fow density values in the Upper Rhine Graben. But dry conditions are considered the least representative of the in situ conditions. Our new thermal properties database now allows us to estimate heat flow density using measurements performed on hot or wet rocks, closer to in situ conditions. The updated heat flow densities calculated for the Soultz-sous-Forêts site are 127 ± 25 mW/m² when considering hot rocks and 184 ± 15 mW/m² when using data for wet rocks. Heat flow density in the GRT-1 well is estimated at 109 ± 18 mW/m² for hot rocks and 164 ± 20 mW/m² for wet rocks. However, although we are sure that temperature increases with depth, the level of saturation of the deep formations is very difficult to assess. We therefore cannot recommend the use of hot conductivity values more than wet values when calculating heat fow densities using the available laboratory data. To improve these estimates, the efect of pressure should also be investigated, and the combined efect of saturation, temperature and pressure needs to be quantifed to provide heat fow density estimates as close as possible to the real values.

Variations of vertical heat fow density were studied in the Soultz-sous-Forêts and Rittershofen wells. Heat fow density in the GRT-1 (Rittershofen) well is considered stable, but unstable down the GPK-1 and GPK-2 wells (Soultz-sous-Forêts), which both show a positive heat fow density anomaly in the Jurassic. We consider that this anomaly is either the result of systematic error in the methods applied or a combined efect of a higher radiogenic heat production in the Jurassic sediments and thermal disturbance caused by the presence of the major faults close to the Soultz-sous-Forêts geothermal site.

The empirical equations built on the new dataset can provide a good approximation of the wet and hot thermal conductivity of a rock sampled in the URG when its dry thermal conductivity is known. In addition, the new thermal conductivities provided in this study constitute a valuable database that informs our understanding of the existing geothermal sites, the modeling of the thermal feld and temperature estimation before drilling operations at prospective sites. Such indications inform the prioritization of future geothermal projects at the basin scale.

Unfortunately, although we consider our heat fow density estimates as the most reliable values currently available for the URG, it is still challenging to comment on the reliability of these estimates using the data (laboratory and feld) available to us at this present time. Since heat fow density estimates are important for geothermal site prospection in the URG, we consider it important that future studies build on the new insight provided herein.

Supplementary information

Supplementary information accompanies this paper at [https://doi.org/10.1186/s40517-019-0154-3.](https://doi.org/10.1186/s40517-019-0154-3)

Additional fle 1: Appendix A. Equations of the trendlines of thermal conductivity as a function of temperature shown in Fig. 8. Coefficient of determination R^2 are also given. The samples are ordered by lithology.

Additional fle 2: Appendix B. Detailed description of the equations presented in the paper and associated error calculation.

Additional fle 3: Supplementary materials. Thermal properties database for samples of the Upper Rhine Graben in the dry state at ambient temperature (black), water-saturated state at ambient temperature (blue), dry state at high temperature (red). Samples are in stratigraphical order. Data include lithology, measured porosity, measuring temperature, thermal conductivity and associated standard deviation, thermal difusivity and associated standard deviation, calculated volumetric heat capacity and associated standard deviation, efect of water saturation and slope of the trendline of thermal conductivity as a function of temperature (Fig. 8).

Abbreviations

B: *Buntsandstein*; Dry: dry rocks at ambient temperature; *e*: thickness; E: Eocene; EGS: Enhanced Geothermal System; gAPI: gamma-ray, American Petroleum Industry (unit); GR: gamma-ray; Hot: dry rock at in situ temperature; J: Jurassic; *λ*: thermal conductivity; *K*: *Keuper*; *λ*air: thermal conductivity of air; *λ*dry: thermal conductivity of the sample in the dry state at ambient temperature; *λ_{form}*: thermal conductivity of the sedimentary formation; *λ*_{hot}: thermal conductivity of the sample in the dry state at the in situ temperature; λ_{sample} average thermal conductivity of the sample; λ_{water} ; thermal conductivity of water; *λ*_{wet}: thermal conductivity of the sample in the saturated state at ambient temperature; *M: Muschelkalk*; *m_d*: dry mass of the sample; m_i : immersed mass of the sample; m_{w} : wet mass of the sample; O: Oligocene; $\pmb{\phi}$: porosity of the sample; *q*: conductive heat flow density; *R²*: determination coefficient of the linear regression; *T*: temperature; 7_{bot form}: temperature at the bottom of the sedimentary formation; *T*_{in situ}: in situ temperature; TPS: Transient Plane Source; *T*_{top} form: temperature at the top of the sedimentary formation; URG: Upper Rhine Graben; Wet: saturated rocks at ambient temperature; *x*_{samp}: proportion of the sample in the formation; *z*: depth; $z_{hot form}$: depth of the bottom of the sedimentary formation; $z_{top form}$: depth of the top of the sedimentary formation.

Acknowledgements

The authors would like to thank John Moine and Bertrand Renaudié for sample preparation. We acknowledge in particular the contribution of Frederick Delay, Laurent Guillou-Frottier and Thierry Reuschlé for their help in the interpretation of the experimental data. The authors are grateful to the Electricité de Strasbourg, ECOGI (Rittershofen), the GEIE EMC (Soultz-sous-Forêts), and ES-Géothermie for providing access to geological and geophysical log data and core or cutting samples. We also acknowledge the Bureau de Recherches Géologiques et Minières (BRGM) for providing the Pulversheim DP202 borehole cores. Also, the authors would like to thank the anonymous reviewers for their contributions and the improvement of the earlier version of this article.

Authors' contributions

PH and PhD collected all the samples. PH performed the thermal property and porosity measurements, data analysis and heat fow density modeling and analysis; and wrote the manuscript, with contributions from all authors. ARLK assisted with the measurements and data analysis and contributed to the interpretation of the data and preparation of the manuscript. CA contributed to feldwork and preparation of the manuscript. PhD and CA provided geological expertise and contributed to interpretation of the data and fgures preparation. PhD led all the feld excursions and contributed to paper writing. MJH and PB contributed to paper writing and interpretation of the data. AG, AR, RH and VM provided unpublished data and contributed to interpretation of the data. All authors read and approved the fnal manuscript.

Funding

This study was funded by LABEX Grant ANR-11-LABX-0050_G-EAU-THERMIE-PROFONDE (this research, therefore, benefted from state funding managed by the Agence National de la Recherche (ANR) as part of the "Investissements d'avenir" program) and ANR Grant CANTARE (ANR-15-CE06-0014-01). Funding for open access was generously ofered by the Helmholtz Centre for Environmental Research—UFZ, Helmholtz Centre Potsdam—GFZ German Research Centre for Geosciences, the Karlsruhe Institute of Technology (KIT), and LABEX Grant ANR-11-LABX-0050_G-EAU-THERMIE-PRO-FONDE. A part of this work was conducted in the framework of the EGS Alsace project, which was co-funded by ADEME.

Availability of data and materials

The datasets generated and analyzed during this study are included in this published article in Table 1, Additional fle 1 and Additional fle 2.

The following datasets are not in the public domain: Masterlog of the GRT-1 well, Soultz-sous-Forêts and Rittershofen temperature logs (contact: geothermie@es.fr)

Competing interests

The authors declare that they have no competing interests.

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Received: 8 November 2019 Accepted: 29 November 2019 Published online: 28 December 2019

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