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Solute Co-Production From Small Geothermal Reservoirs – How Little Is Too Little?

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ABSTRACT

How much lithium can we 'gain' by way of solute co-production from geothermal reservoirs in Central Europe? without asking 'at what costs', we examine the generic question of solute co-production solely in terms of fluid turnover and residence times, assuming conservative mixing and transport (while a lithium-depleted fluid is being continually re-fed into the reservoir and recirculated within its available flow-paths, lithium replenishment from adjacent rocks likely remains negligible over the project time scales of consideration). We illustrate this for two typical geothermal reservoir settings based on fluid turnover (liquid-phase only) in Central Europe: a petrothermal system in the N-German Sedimentary Basin, and a more 'aquifer'-like reservoir in the Upper-Rhine Rift valley.

Can tracer tests enable model-independent predictions of georeservoir output? this question is worth raising particularly as concerns thermal drawdown, and solute co-production. For geothermal reservoirs operated by production/re-injection wells, thermal lifetime (TLT) is usually defined in terms of a temperature drop threshold, and estimated as a function of fluid turnover time (FT) and heat exchange surface-area-per-volume, the former (FT) being hopefully measurable by means of a tracer test, whereas the latter is rather difficult to infer from tracer signals alone. Deriving FT from artificial-tracer signals looks model-independent (formally), but is subject to large-time extrapolation uncertainty (which restores model-dependence).

Unlike thermal forecasting, tracer-based prognosis of solute co-production (more precisely, of its lower-bound level, assuming conservative transport by fluid turnover only, non-replenished from adjacent rocks) is not impeded by large-time extrapolation uncertainty, nor by reservoir model and/or parameter ambiguity, since mass output prediction as a function of time requires just knowledge of conservative-tracer fluxes within the forecasting time horizon. Once a tracer test was conducted in accordance with the rules of the art, the reservoir can be treated like a black box with a response function derivable, in a well-defined manner, from the artificial-tracer signal (as long as the flow regime does not change significantly).

This approach is adequate for (conservative) solute co-production, but not for heat transport. Tracer test results from a particular Upper-Jurassic (Malm) carbonate aquifer near Munich illustrate the issue with TLT as a poorly-defined function of FT. Tracer signals available to date yield FT in the range of months (still subject to extrapolation uncertainty), and are compatible with both fracture-dominated and aquifer-like representations of reservoir structure; compatible values for the heat exchange surface area span four(!) magnitude orders.

1. INTRODUCTION

Growing interest in 'geothermal lithium' co-production is nourished by the endeavor to reduce the environmental footprint of lithium extraction from more traditional hydrogeosphere compartments that are closer to the ground surface, along with a perceived or imagined lithium scarcity or even 'crisis', facing what seems to be an ever-growing demand for it. For the geothermal community, additional motivation is spurred by a perceived 'need' to overcome the economic unviability of most geothermal projects in Central/Northern Europe.

How much lithium can we 'gain' by way of solute co-production from geothermal reservoirs in Central Europe? Without asking 'at what costs', we examine the generic question of solute co-production here solely in terms of fluid turnover and residence times, assuming conservative 'mixing' and transport; while a solute-depleted fluid is being (more or less continually) re-fed into the reservoir and recirculated within the 'available' flow-paths, solute 'replenishment' from adjacent rocks likely remains negligible over the project time scales of consideration (given their fluid turnover rhythm). We illustrate this for two typical geothermal reservoir settings based on fluid turnover (liquid-phase only) in Central Europe: a petrothermal system in the N-German Sedimentary Basin, and a more 'aquifer'-like reservoir in the Upper-Rhine Rift valley. Their somewhat contrasting flow-storage characteristics are summarized in fig. 1, where solid lines show a flow distribution by storage according to the synthetic 'reservoir geometry' concept proposed by Shook (2003), and dashed lines show a flow distribution by retardation following a modified concept of Behrens et al. (2010), which enhances the visibility of im-/mobile fluid exchange (in particular, matrix diffusion) effects in reservoir compartments else inaccessible to measurement.

2. TRACER-BASED FORECAST APPROACH

Co-produced solute depletion during fluid re-circulation is roughly governed by a 'mixing' relationship,

$$C_{\mathrm{mix}}(\mathrm{t}) \;=\; C_{\mathrm{ini}}\;(\,1 - R(\mathrm{t})\,) \;+\; C_{\mathrm{resid}}\;R(\mathrm{t}) \tag{1}$$

with $R(\mathrm{t})$ denoting the re-circulated water fraction as a function of time, to which a residual (non-extractable) solute level $C_{\mathrm{resid}} > 0$ may persist in the re-injected water, but hopefully significantly lower than the reservoir's native level C_{ini} . In the sequel, for 'geothermal lithium' co-production, C_{resid} is assumed as a non-dynamic (gradient-independent) level of, say, 15 mg/L, whereas C_{ini} is site-specific.

We posit that $R(\mathrm{t})$ can be estimated from artificial-tracer signals, as the cumulative-recovery ratio of a conservative artificial tracer being transported in the reservoir under similar flow conditions, based on a deconvolved tracer signal corresponding to a single, short-time, flux-mode input of a well-defined tracer quantity (mass or activity) A_{tracer} .

We thus get for the co-produced solute mass as a function of time, cumulatively:

$$M_{\mathrm{out}}(\mathrm{t}) \quad = \quad (\, C_{\mathrm{ini}} - C_{\mathrm{resid}} \,) \quad [\, VOL_{\mathrm{out}}(\mathrm{t}) - \int_{\mathrm{o}}^{\mathrm{t}} \int_{\mathrm{o}}^{\mathrm{t'}} Q(\mathrm{t'})\, Q(\mathrm{t''})\, g(\mathrm{t''})\, \mathrm{dt''}\, \mathrm{dt'} \,] \qquad (2)$$

where g as a function of time is obtained from a flux-mode sampled conservative-tracer signal, normalized by the tracer's total input quantity A_{tracer} . This tracer-based g, with physical dimensions of 1/*volume*, is in turn used like a Green's kernel to represent 'mixing losses' due to depleted-fluid re-injection.

2.1 Solute Co-Production Estimates for Tiny Single-Well Heat Extraction Schemes in the N-German Basin

Dissolved lithium levels in the range of 150 – 250 mg/L have been reported, from various sources, for native formation waters encountered at the most popular geothermal project sites in the N-German sedimentary basin. We apply relationship (2) for a tracer-based appraisal of 'geothermal lithium' co-producible from a former gas exploration well called Horstberg-Z1, in the Südheide area. The rationale of the **GenESys** project (innovative single-well techniques for heat extraction from tight sediments), its lessons and accomplishments have been reported extensively by its original initiators and developers (Jung et al. 2005), and will not be reproduced here. Here, we only refer to artificial-tracer signals, from tracer tests conducted upon hydraulic fracturing (the most successful segment of a rich bundle of reservoir development and stimulation measures, back in 2004). Details of tracer (non-trivial) behavior were provided by Behrens et al. (2006). Tracer-test findings and their bearings on the maximum expectable thermal lifetime of such single-well operation schemes were summarized by Ghergut et al. (2013, 2016).

It is yet hard to tell which of the two conservative tracers, tritiated water (HTO), or naphthalene-1,5-disulfonic acid salt (NDS), should be taken as 'more representative' of lithium transport in the Horstberg formation (subject to hydraulic fracturing), since a lower ion size can be more or less compensated by a larger hydration radius, the latter being however limited by 'competitors' within the overall ionic strength, which in turn might get (temporarily) lowered by massive water injection. In the target formation, native water salinity is known to be 'over-saturated'. Sporadic fluid sample analyses, physico-chemical fluid monitoring, electrical conductivity records from past stimulation/fracturing experiments are yet inconclusive. Back in 2004, hydraulic fracturing has been performed successfully, in selected claystone/sandstone layers, using 'plain water' with a minimum of physico-chemical conditioning, without proppants; at certain stages during prior (less successful) stimulation experiments, also some acid had been added (in moderate quantities) at selected well-screen intervals. On the other hand, from simulations shown in the sequel (figs. 2 – 4), it turns out that it actually 'doesn't matter' which of the tracers (HTO or NDS) is taken as a lithium 'proxy', despite the marked difference in their measured breakthrough signals.

It is furthermore hard to tell to what extent the native lithium levels at Horstberg might have become 'diluted', over a more or less significant radius around the borehole, by the various stimulation treatments, particularly by the massive hydraulic fracturing operations. Two limiting scenarios are therefore considered here, first (fig. 2) assuming the undisturbed native levels, taken to be ~210 mg/L based on LBEG (Lower Saxony's State Authority for Mining, Energy and Geology) appraisals, next (fig. 3) assuming these levels would become roughly halved at post-fracturing stages.

With the cyclic production scheme envisaged by GenESys (yearly three-month production stages during winter, shut-in for the remainder of ~9 months, allowing for thermal 'recovery' to a yet uncertain extent), each 'next' cycle would resume, in terms of solute co-production, from a lowered in-situ level – cf. the r.-h. s. end-levels of blue/green curves in figs. 2 and 3; it seems rather unlikely that this could be replenished by significant amounts, during shut-in stages, through 'fresh' (geological-age) lithium ion release from adjacent rocks. The system is deemed 'tiny' given its flow rates, decreasing from ~9 to ~3 L/s over one 'free-outflow' stage. 'Free' outflow at the upper well-screen is based on prior pressure buildup by water injection at the lower well-screen, without using a production pump (actually, a pressure regulator and/or flow controller device needs to be working at the upper screen).

2.2 Solute Co-Production Estimates for Mid-Sized Inter-Well Loops (like Soultz-sous-Forêts) in the Upper-Rhine Rift Valley

Dissolved lithium levels of ~173 mg/L have been reported for native formation waters in the geothermal reservoir at Soultz-sous-Forêts (France) by Sanjuan et al. (2020), along with more or less similar values for further sites on both (Western/French and Eastern/German) sides of the Upper-Rhine Rift valley. Relationship (2) can be used to forecast 'geothermal lithium' output for the main loop (fluid production from well GPK2, 'spent'-fluid re-injection into well GPK3) using artificial-tracer signals obtained during a relatively stable flow regime, back in 2005, with a six-month stage of fluid turnover at almost constant rate ~12 L/s (Blumenthal 2007).

Unlike the previous section's single-well operation setting, fluid/heat/solute turnover in inter-well systems is usually supposed to be continuous and monotonous. However, the forecast time horizon in figs. 2 – 4 was kept similar to the duration for which a conservative-tracer signal, undisturbed and virtually 'complete', was available for the GPK3 → GPK2 (partial) loop at Soultz-sous-Forêts.

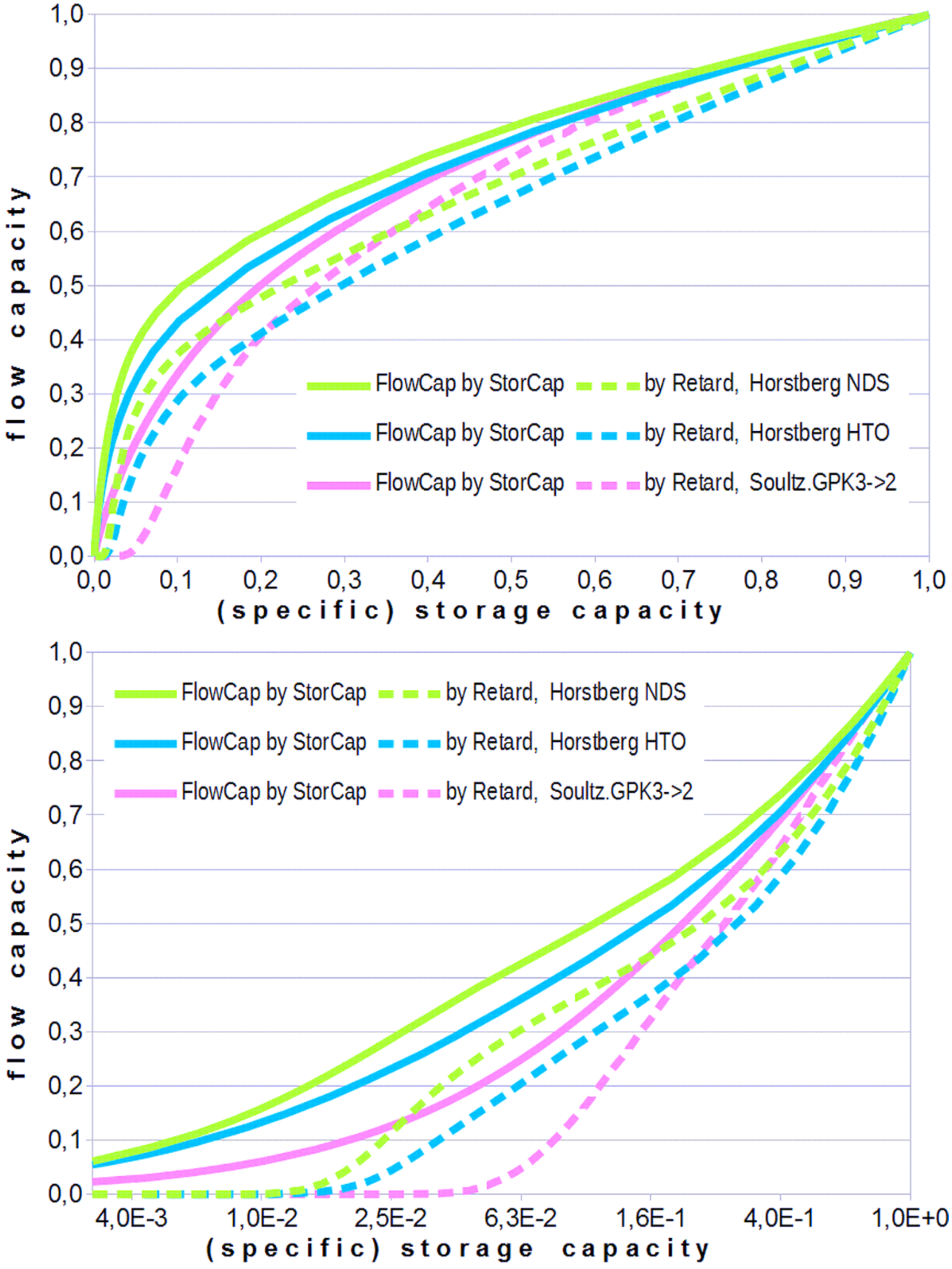


Figure 1: Horstberg versus Soultz-sous-Forêts: petrothermal vs. more aquifer-like reservoir characteristics. Different weighting of im-/mobile fluid exchange (in particular, matrix diffusion) effects by different tracers (NDS, HTO) within the same reservoir (Horstberg), whose visibility is enhanced by the logarithmic time scale (lower plot) in flow-retardation (and to a lesser extent also in flow-storage) diagrams. Solid lines follow the concept of Shook (2003), dashed lines the modified concept of Behrens et al. (2010).

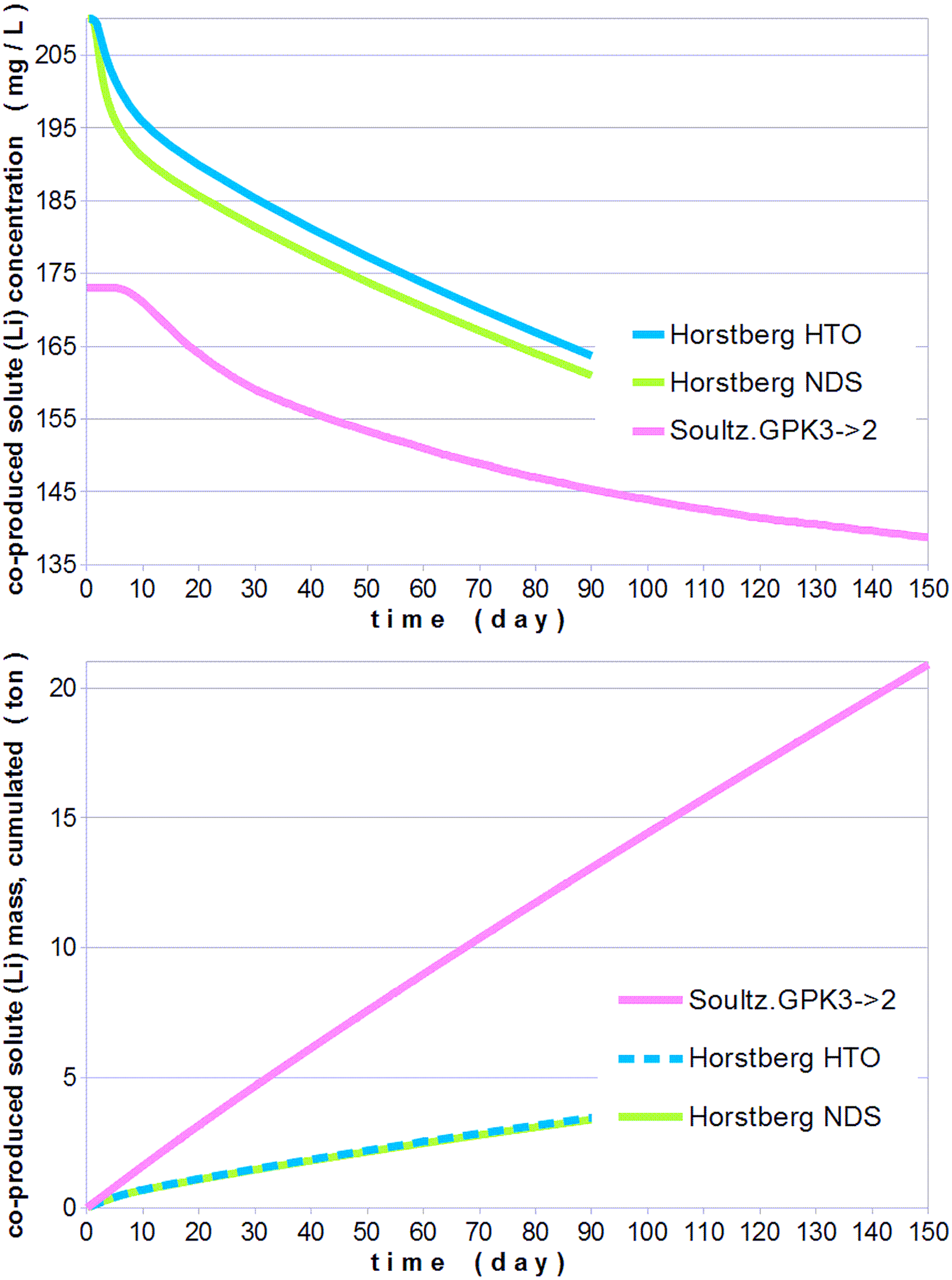


Figure 2: Horstberg versus Soultz-sous-Forêts: solute co-production output assuming initial solute levels were undisturbed by massive fracturing operations. Horstberg estimates span a (hypothetical) three-month production cycle, as originally endeavored by the GenESys scheme. Soultz-s.-F. estimates span the duration for which 'fully representative' tracer test signals were available. – Caveat: such seemingly 'linear' evolution trends should not be taken as a basis for long-term lithium output forecast (cf. fig. 5 in the sequel), for neither site; early slopes 'insinuate' long-term prospects that are tremendously overestimated (unrealistically optimistic).

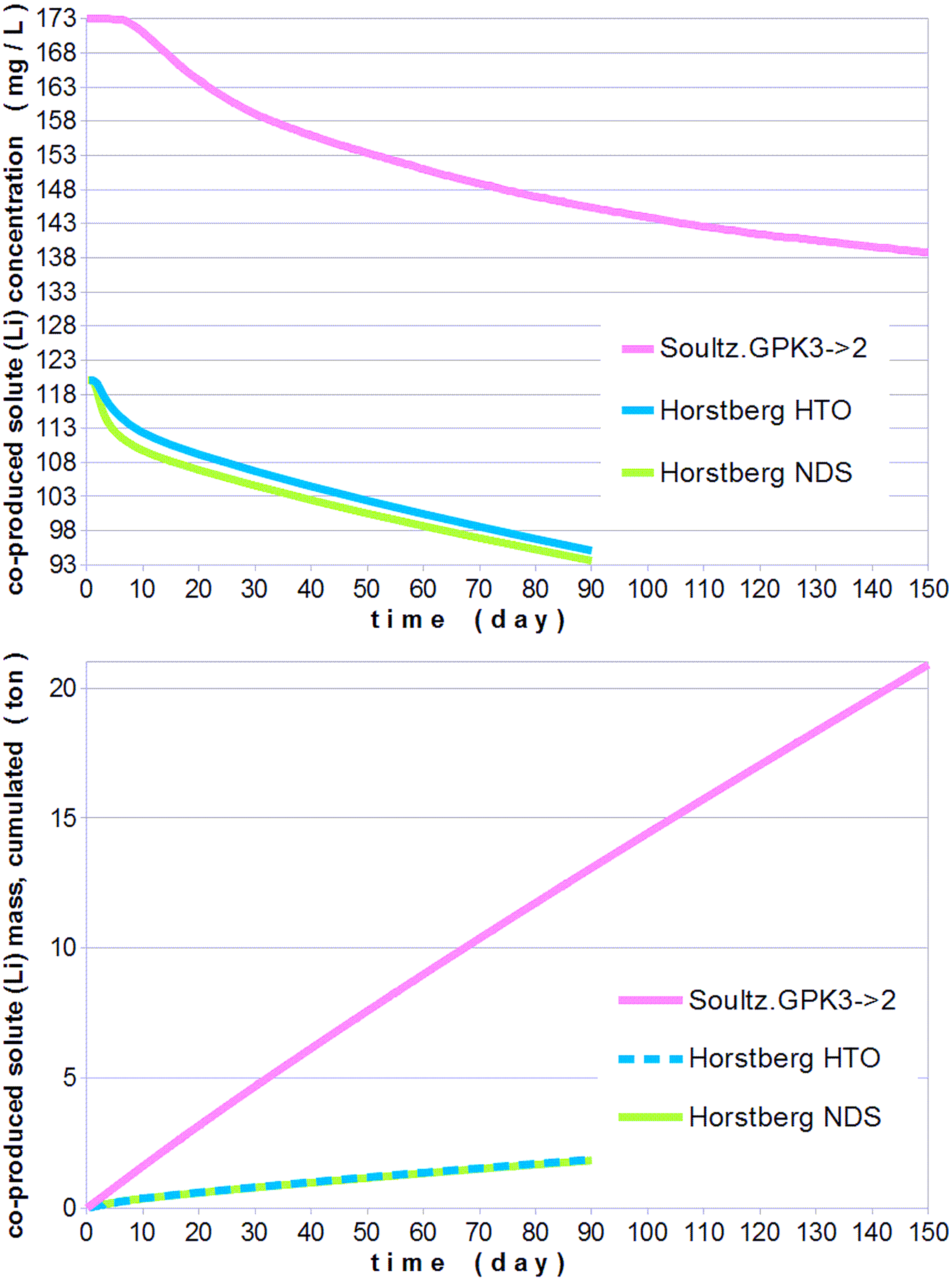


Figure 3: Horstberg versus Soultz-sous-Forêts: solute co-production output assuming initial solute levels were roughly halved by massive fracturing operations – at the Horstberg site only, whose cumulative mass output (lower plot, blue/green curves) slope now appears approx. halved, compared to fig. 2. – Caveat: such seemingly 'linear' evolution trends should not be taken as a basis for long-term lithium output forecast, for neither site; such early slopes would considerably overestimate mid-term trends and final (total) output prospects (cf. fig. 5 in the sequel), even when the prior 'dilution' by reservoir stimulation treatments is taken into account.

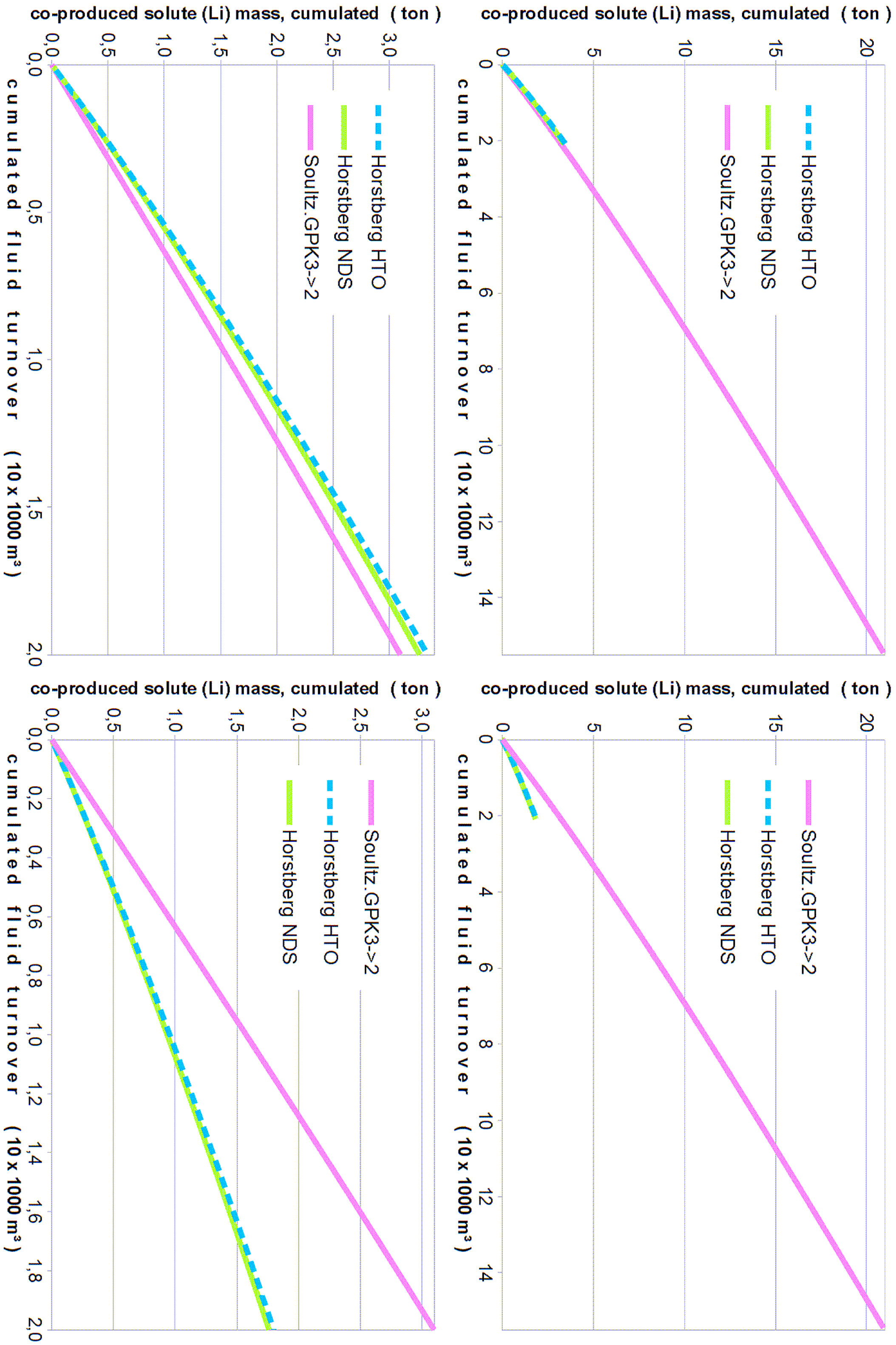


Figure 4: Horstberg vs. Soultz-sous-Forêts: same scenarios as in figs. 2 and 3, mass output now plotted against fluid turnover, rather than time. Slope similarity of Soultz-s.-F. with the 'undisturbed' Horstberg scenario is a mere coincidence of numbers, without theoretical relevance. – Caveat: such seemingly 'linear' evolution trends should not be taken as a basis for long-term lithium output forecast, for neither site (cf. next figure, and comments to previous figures).

Figures 2 – 4 were meant to provide a rapid early appraisal of how solute co-production in a mid-sized 'hydrothermal' (more aquifer-like) setting performs compared to a small petrothermal system, but slope 'trends' seen at early times may act misleadingly; that these slopes are bound to 'vanish' (i. e., drop to negligible levels) within finite time spans, was already inherent in relationship (2). For inter-well loops like at Soultz-sous-Forêts, it might happen within 1–2 decades. For a 'tiny' single-well petrothermal scheme, a significant drop might occur within a matter of months. In fig. 5, however, curve 'flattening' is partly due to the chosen logarithmic scale.

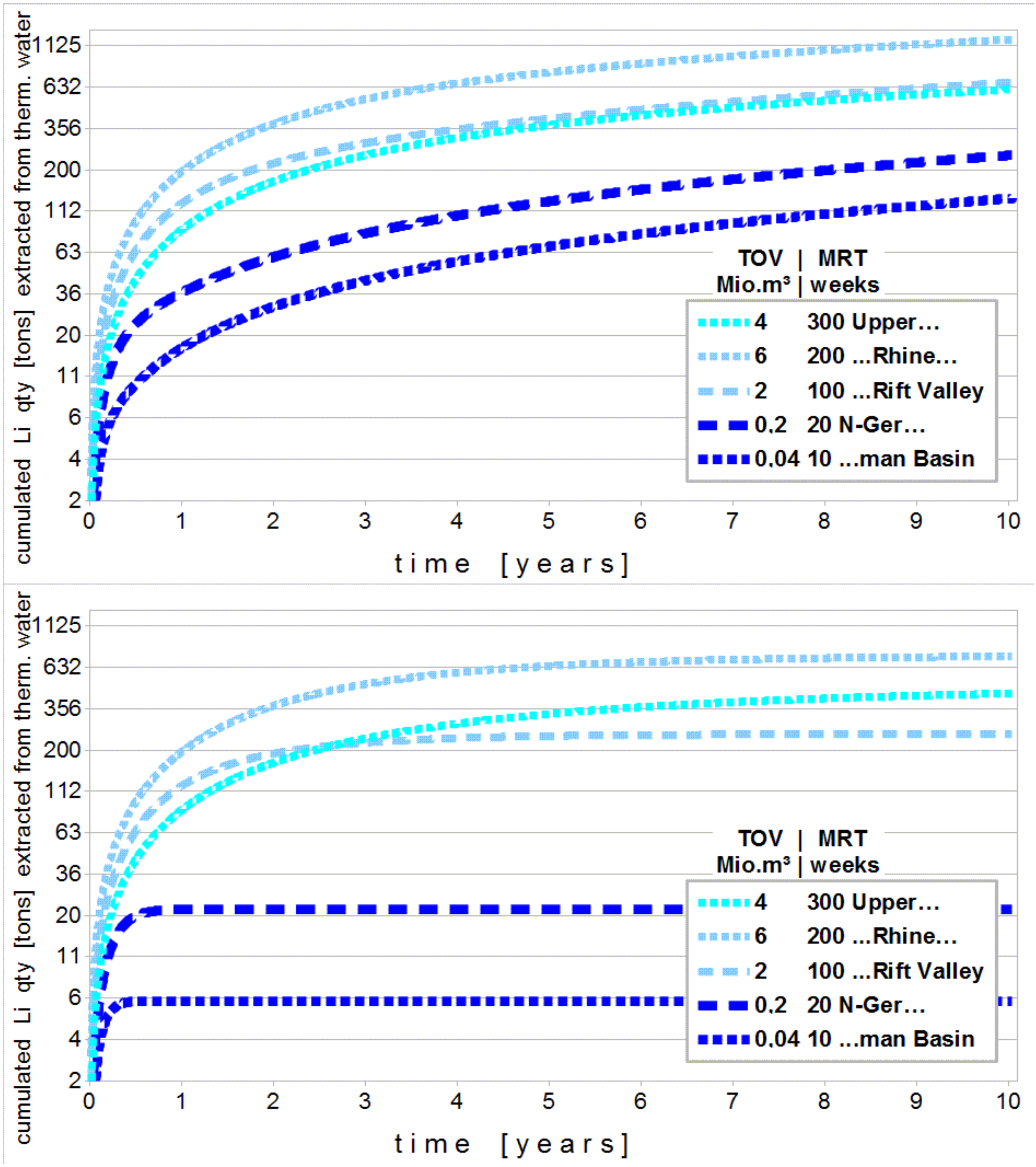


Figure 5: Mid- and long-term prospects for solute co-production from georeservoirs spanning the size range (bulk appraisal by fluid turnover volume TOV and mean residence time MRT) typical for the N-German Basin (plotted in strong blue) and the Upper-Rhine Rift Valley (plotted in light blue), assuming open loops with max. 60% tracer recovery (upper diagram) or closed loops with virtually full tracer recovery (lower diagram).

3. DO TRACER TESTS ENABLE MODEL-INDEPENDENT PREDICTIONS OF GEORESERVOIR OUTPUT?

One might feel tempted to think such a tracer-based approach to solute output forecast should also apply to heat output from systems with a rather fast thermal drawdown – which unfortunately are not quite exceptional (cf. Behrens et al. 2020/2021, to which fig. 6 here provides an update, or Ghergut et al. 2013/2016 for both 'aquifer'-like and petrothermal systems). For geothermal reservoirs operated by production/re-injection wells, thermal lifetime is usually defined in terms of a temperature drop threshold, and estimated (cf. Horne 1985, Pruess and Bodvarsson 1984, Grant and Bixley 2011, Ghergut et al. 2016 for various equivalent formulations) as a function of fluid turnover time and heat exchange surface-area-per-volume,

$$T_{\mathrm{heat}} = R_{\mathrm{heat}} \cdot T_{\mathrm{fluid}} + D \cdot \sigma^2 \cdot T_{\mathrm{fluid}}{}^2 \tag{3}$$

with T_{fluid} supposed to be measurable by means of a tracer test; whereas σ is rather difficult to infer from tracer signals alone.

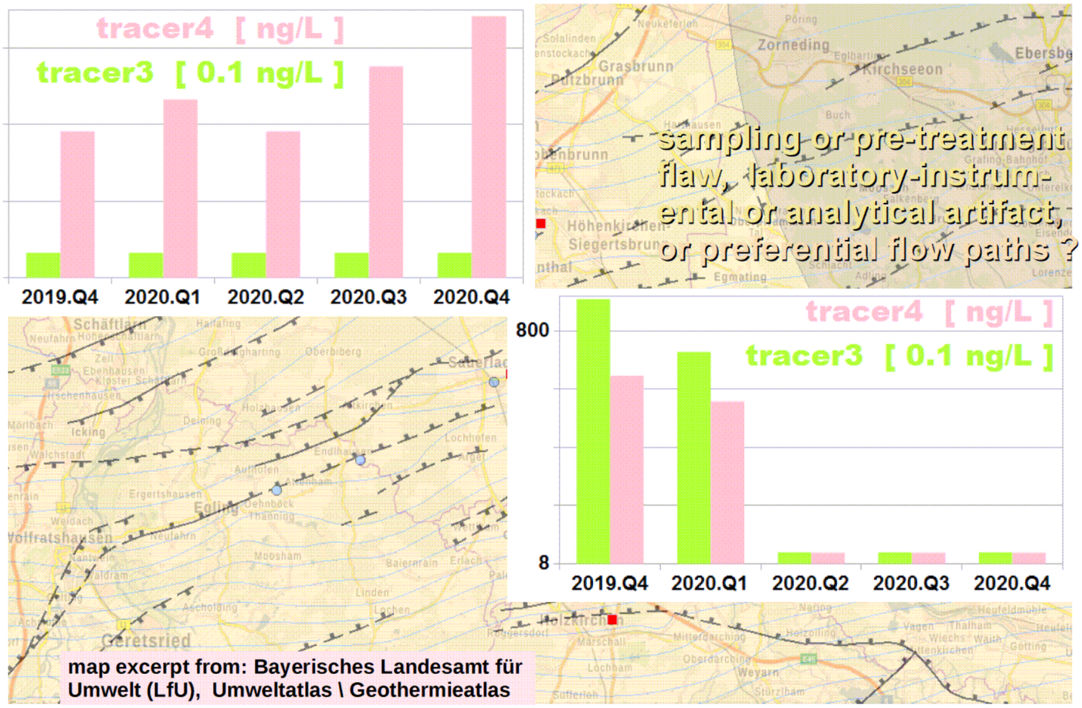
For 'aquifer'-like reservoirs, the linear term prevails:

$$R_{\mathrm{heat}} >(>>) 1 \ , \quad D \cdot \sigma^2 \cdot T_{\mathrm{fluid}} << 1 \tag{4}$$

For fracture-dominated ('petrothermal') reservoirs, the quadratic term prevails:

$$R_{\mathrm{heat}} \approx 1 \ , \qquad D \cdot \sigma^2 \cdot T_{\mathrm{fluid}} >> 1 \tag{5}$$

Deriving T_{fluid} from artificial-tracer signals looks 'model-independent', but is subject to large-time extrapolation uncertainty (which restores model-dependence).



map excerpt from: Bayerisches Landesamt für Umwelt (LfU), Umweltatlas \ Geothermieatlas

Figure 6: Sample contamination by 'alien' tracers, or solute transport connection between local and regional flow systems? Tracer tests in a 'small' well-doublet loop may reveal solute exchange ('replenishment'?) and flow contributions from fluid turnover at larger scales – from which solute co-production could also benefit.

Unlike thermal forecasting, tracer-based prognosis of solute co-production (more precisely, of its lower-bound level, assuming conservative transport by fluid turnover only, non-'replenished' from adjacent rocks) is not impeded by large-time extrapolation uncertainty, nor by reservoir model and/or parameter ambiguity, since mass output prediction as a function of time, based on rel. (2) as derived in section 2, requires just knowledge of conservative-tracer fluxes within the forecasting time horizon. Once a tracer test was conducted in accordance with the rules of the art (usually including observance of flux-type boundary conditions for tracer input and fluid sampling, as discussed by Kreft and Zuber 1978), the reservoir can be treated like a 'black box' with 'response function' (Green's kernel surrogate) g . This approach is adequate for (conservative) solute co-production, but not for heat transport (fig. 7 versus fig. 8). Attempting to predict production temperatures by the same approach as illustrated in fig. 5 (based on conservative mixing, rel. (1)) would systematically discard the diffusive contribution (heat conduction, quadratic term in rel. (3), which prevails for petrothermal reservoirs (5)), while overestimating the speed of heat advection (as if the heat retardation factor R_{heat} of rel. (3) were 1, which matters primarily for aquifer-like reservoirs (4)). By the same token, far-field contributions (in open loops) would become over-weighted:

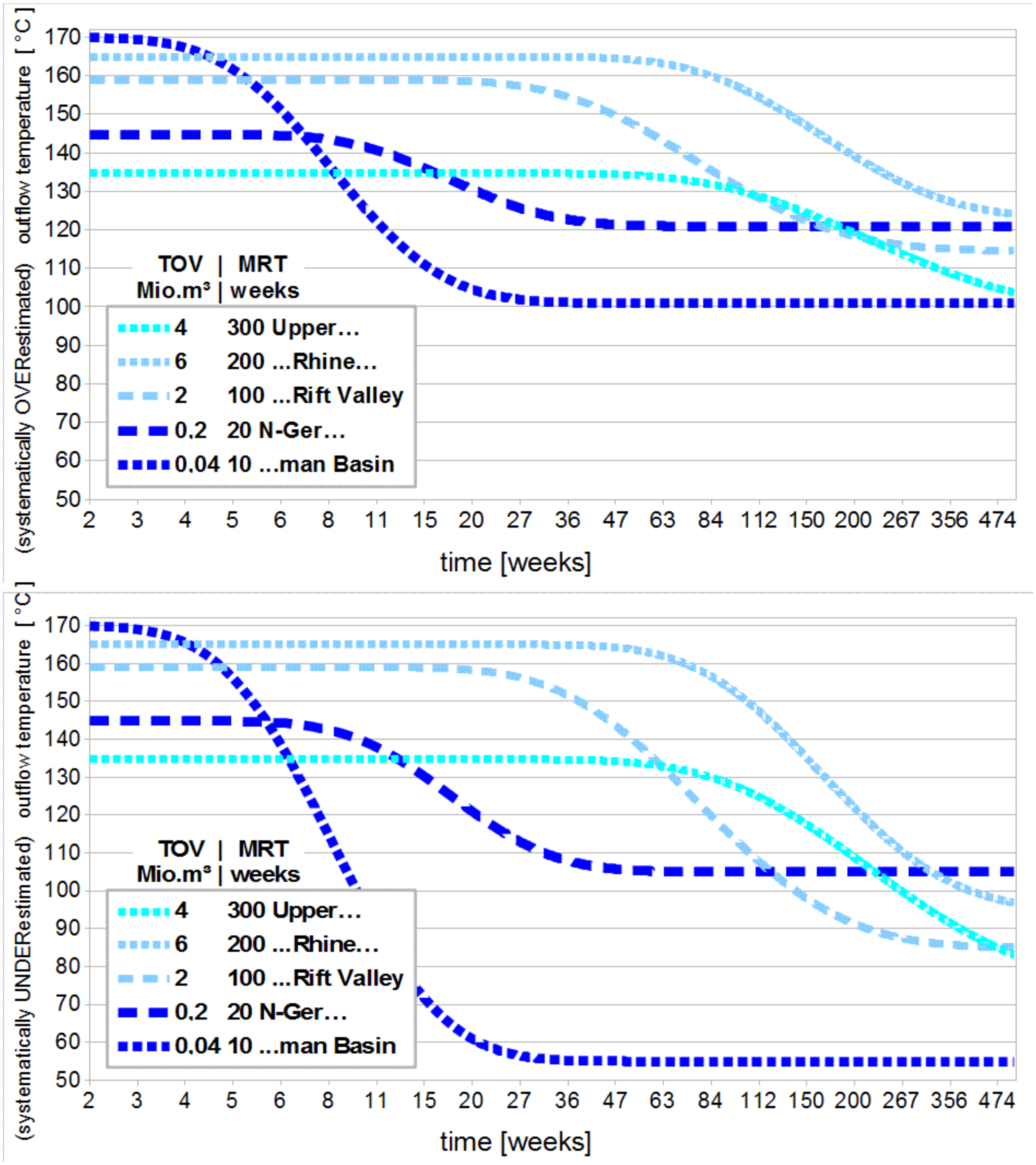


Figure 7: Attempting thermal forecast the same 'tracer-based' way as for solute co-production would systematically misestimate outflow temperatures: shown here for similar reservoir sizes and scenarios as in fig. 5 (the upper diagram for open loops with max. 60% tracer recovery, the lower diagram for closed loops with virtually full tracer recovery).

In other words, it would discard the dependence of heat transport on reservoir parameters further than T_{fluid}, as outlined in fig. 8 for aquifer-like (l.-h. s.) and petrothermal (r.-h. s.) reservoirs. While the factors R_{heat} and D in relationship (3) may, as a second-best, be regarded like 'material parameters' and roughly estimated from prior exploration data, the effective heat exchange surface-area-per-volume σ remains essentially unknown until a large (i. e., TOV-range) volume of fluid has been circulated and turnover data evaluated (by recipes that remain essentially model-dependent). Tracer test results from a particular Upper-Jurassic (Malm) carbonate aquifer near Munich (cf. Behrens et al. 2020, 2021) illustrate the issue with T_{heat} as an incomplete 'function' of T_{fluid}. Tracer signals available to date yield T_{fluid} in the range of months (still subject to extrapolation uncertainty), and are compatible with both fracture-dominated and 'aquifer'-like representations of reservoir structure; 'compatible' values for σ span four(!) magnitude orders.

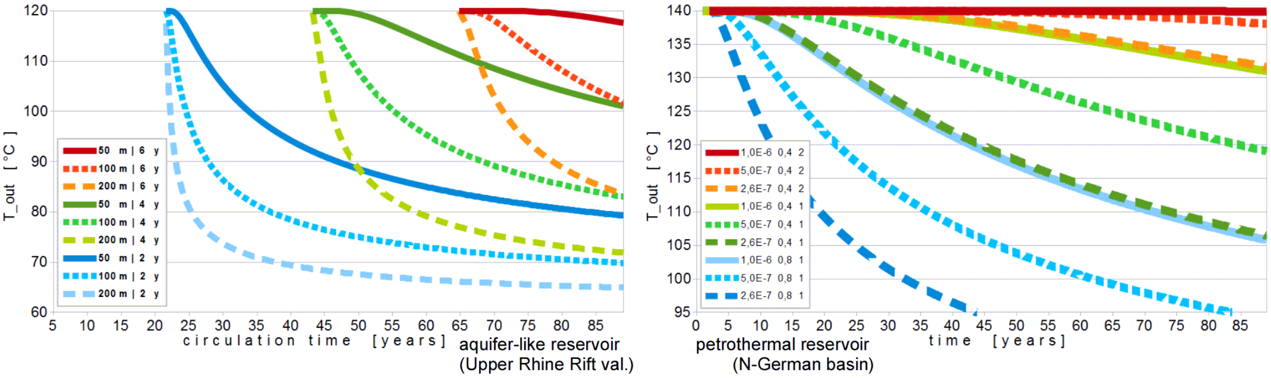


Figure 8: Heat transport in the reservoir, and thus production temperatures depend on parameters further than T_{fluid}, e.g., effective 'aquifer' or fracture thickness (whose reciprocal value yields $\sigma/2$) as well as heat diffusivity (proportional to parameter D, cf. relationship (3)).

By contrast, tracer signals from single-well or inter-well circulation tests conducted under representative flow conditions could be used to predict solute (in particular, 'geothermal lithium') output, and its gradual depletion in reservoir fluid turnover, irrespective of the availability and parametrizing of a reservoir model. If transport can be assumed as conservative, the forecasting scheme described in section 2 would still be applicable even when tracer tests yield 'unexpected' results, as illustrated in fig. 6 for the geothermal reservoir that was explored by Behrens et al. (2020, 2021, where tracer signals had been primarily supposed to 'explain' thermal drawdown). Moreover, the detection of 'cross-well' tracer signals in neighboring fluid loops would facilitate the estimation of at least an upper bound on a tracer's mass recovery ratio, and thus reduce the uncertainty associated with large-time extrapolation of this tracer's signals (both for the original loop and for the 'extraneous', cross-well flow path).

Last not least, solute co-production forecast also faces a non-trivial challenge: foresee and quantify overall water-rock interaction effects of 'spent fluid' re-injection, the latter being depleted of its particular micro-constituent (albeit at trace levels only), but likely acidized and 'unstably' buffered at major-ion levels. Water-rock interactions cannot be told from conservative-tracer signals; hydrogeochemical modeling (Kölbel et al. 2020, Maier et al. 2021) becomes indispensable.

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