SINGLE-WELL AND INTER-WELL DUAL-TRACER TEST DESIGN FOR QUANTIFYING PHASE VOLUMES AND INTERFACE AREAS

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ABSTRACT

Single-well tracer push-pull tests suffer from poor sensitivity w. r. to rapid-equilibrium exchange of tracer between solid and/or fluid phases. This renders parameters like phase volumes (saturation) and phase interface areas difficult to invert from single-well PULL signals of phase-partitioning or sorptive tracers. Two possible workouts are proposed: (A) using the time-dependent, in-situ release of a second tracer, from the originally injected tracer, with contrasting partitioning or sorption properties; (B) exploiting the temperature dependence of sorption in conjunction with the injection (PUSH) of a sufficient volume of cold water into a geothermal reservoir. The idea of (A) is inspired by TOMICH ET AL. (1973), who used the in-situ release of an alcohol by hydrolysis from a injected ester, in order to make residual-oil saturation determinable from a single-well push-pull test.

Key-words: tracer, fluid-rock, surface area, sorption, partitioning, reactive, thermosensitive, single-well, push-pull, in-situ release.

Abbreviations:

IW	inter-well flow-path [tracing test]
SW	single-well push-pull [tracer test], a. k. a. 'SWIW' [single-well injection-withdrawal, not to be confounded with IW as used here]
alc	'alcohol', symbolically standing for a non-reactive tracer with the (non-)partitioning behavior described by Tomich et al. (1973)
est	'ester', symbolically standing for a reactive tracer with the (well-balanced) partitioning properties described by Tomich et al. (1973)
ref	reference tracer: a non-reactive, liquid-phase tracer, indiscernible from the reservoir brine in terms of its transport behavior
BTC	breakthrough curve [of a tracer in a test]

INTRODUCTION

Geothermal reservoirs, and other technology-relevant georeservoirs in the realm of energy production (like CCS, gas-storage or spent-radionuclide repositories) contain mobile and immobile fluid regions, and often also different fluid and solid phases. The lifetime of a particular georeservoir (from a hydraulic, thermal, geomechanical and/or hydrogeochemical point of view) depends on the volumes and/or interface areas of some of these regions and/or phases (fig. 1). Mostly, these cannot be measured by geophysical and hydraulic methods. Since they essentially relate to fluid-based transport processes, attempting to measure them by tracer tests is a sensible endeavor.
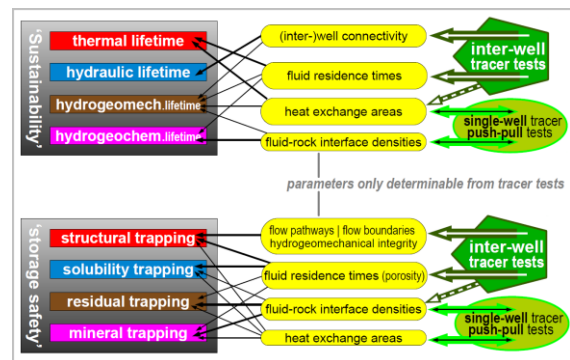

Figure 1: Overview of some lifetime-controlling parameters for geothermal or CCS reservoirs that essentially relate to fluid transport processes, and thus can only be measured from tracer tests (IW, or SW).

Both inter-well and single-well tracer tests appear as indispensable to georeservoir characterization and lifetime prediction (fig. 1). However, in designing and dimensioning IW or SW tracer tests, one should keep in mind that not every tracer test will be sensitive to every fluid transport parameter. A certain complementarity exists, in terms of parameter sensitivity,

- − between SW and IW methods,
- − between equilibrium and kinetic exchange processes,

– between volume and area parameters.

From experience and from theory, from beautiful studies and from nice examples (MAŁOSZEWSKI AND ZUBER 1985, ROBINSON 1985, KOCABAS AND HORNE 1987, ZEMEL 1995, CARRERA ET AL. 1998, SCHROTH ET AL. 2001, HAGGERTY ET AL. 2001, SHOOK 2001, KOCABAS 2005, NERETNIEKS 2007, PRUESS AND DOUGHTY 2010), we may learn that

- mobile-fluid volumes can be measured from conservative-tracer IW tests, whereas SW tests are generally insensitive w. r. to mobile-fluid volumes;
- immobile-fluid volumes, in single-phase systems, are rather difficult to measure, by either kind of test;
- different-phase volumes can be determined from IW tests using partitioning tracers at equilibrium exchange between phases; whereas SW tests are not very sensitive w. r. to tracer exchange processes at equilibrium;
- interface areas between im-/mobile fluid compartments, or between different fluid phases can be determined from SW tests relying on kinetic exchange processes between compartments or phases;
- SW tests are often believed to be more sensitive w. r. to such processes than w. r. to advection-dispersion processes, and than IW tests.

IW tests are not physically insensitive w. r. to kinetic exchange processes, but they may be strongly affected by ambiguity between dispersion and non-advective non-equilibrium processes (actually, this ambiguity also impedes upon SW tests, as seen, for instance, in figure 2 of BEHRENS ET AL. 2009).

MOTIVATION

The DOE REPORT (2008) on the state of EGS technology emphasizes the need for measuring rock-fluid interface areas in geothermal systems, stating that "*reliable tracers that can measure and/or monitor the surface area responsible for rock-fluid heat and mass exchange do not exist, limiting the ability to quantify and predict heat extraction efficiencies*" (p. 22); remarkably, its Glossary lists two separate tracer definitions: a mere "*tracer*" being used to determine flow paths and velocities, and a "*smart tracer*" being needed for determining "*the surface area contacted by the tracer*" (...).

The sorption of solutes from the flowing fluid to the reservoir rock being a process that directly involves the fluid-rock interface, it seems that sorptive tracers can provide the answer to the cited DOE challenge. ROSE ET AL. (2011) investigated how the use of "*quantum dot tracers with controllable surface sorption characteristics*", and with "*low matrix diffusivity*" within "*single-well tracer testing*

methodologies (...) *should result in significant advances in the interrogation of surface area in enhanced geothermal reservoirs*" (p. 6). Indeed, unlike matrix diffusion (cf. CARRERA ET AL. 1998, HAGGERTY ET AL. 2001), tracer sorption appears as a robust, easily-quantifiable process, whose modeling is much less intricate than that of matrix diffusion, and also much less dependent on various theoretical assumptions regarding void-space structure and geometry details, and on their parametrization.

Around the time the DOE Report was disseminated, the geothermal community also witnessed a revival of popularity for SW methods, rooted in

- the expectation SW tests were more sensitive w. r. to non-advective processes (like tracer sorption or partitioning between fluid phases), than IW tests, and than w. r. to advective processes within the SW tests themselves, owing to field-flow reversal (the PULL stage would "revert" the effects of advective processes that occurred during the PUSH stage, a suggestion that is commonly derived from HAGGERTY ET AL. 2001);
- the obvious practical interest in conducting SW tests (maybe even 'instead of' IW tests), since SW tests require lower tracer quantities, lower fluid volume turnover, and less time to perform than IW tests, while promising to yield higher tracer recoveries, and higher (i. e., more easily detectable) tracer signals, than can be expected from a reservoir-scale testing.

For all of these reasons, the sorptive-tracer SW methodology appears to deserve a closer look.

SW TEST INSENSITIVITY W. R. TO RAPID-EQUILIBRIUM EXCHANGE, AND TWO POSSIBLE WORKOUTS

A major limitation confronting the above-listed ideas, however, occurs from SW's poor sensitivity w. r. to tracer exchange processes (between fluid and rock, for sorptive tracers, or between fluid phases, for partitioning tracers in multiple-phase systems), when these exchange processes are at equilibrium – which they very often are, in subsurface flow systems, given the 'slow' groundwater velocities, against the 'fast' sorption-desorption rates at fluid-rock interfaces, or dissolution-diffusion rates at liquid-gas interfaces.

As can be seen from the examples provided by SCHROTH ET AL. (2001), BEHRENS ET AL. (2009), it is only by virtue of never absent flow-field dispersion(!) that differently-sorbing tracers can produce different BTCs during the PULL stage of a SW test. It is the dispersion of the very flow field that 'pulls' the sorptive-tracer BTCs apart, during the PULL stage, rather than being itself "annihilated" by virtue of flow-field reversal. Not the effects of dispersion, but those of (rapid-)equilibrium sorption

get "reverted" during the PULL stage. As a matter of fact, what made dispersion look like "revertible" in the SW tests described by HAGGERTY ET AL. (2001) was not the flow-field reversal, but the choice of a very large time scale for BTC observation: at this scale, advection-dispersion influences on a tracer's BTC tailing become negligible, against matrix-diffusion fluxes – not just for SW tests, but even in IW tests.

To overcome the insensitivity of SW tracer test signals w. r. to rapid-equilibrium exchange processes like sorption or partitioning between fluid phases, we can consider two ways:

–   (A) release of a second tracer, in-situ, from the originally-injected one, in a time-dependent manner; with 'source' and 'daughter' tracers having different (the more contrasting, the better) partitioning or sorption properties

–   (B) exploiting the temperature dependence of (covalent-type) sorption processes in conjunction with the injection (PUSH) of a sufficient volume of cold water into a geothermal reservoir.

With (A), we re-enact a very inspiring idea from TOMICH ET AL. (1973), who proposed to use the in-situ production of an alcohol by hydrolysis from a injected ester, with the purpose of measuring residual-oil saturations by means of SW tests. Ester hydrolysis being a rather 'slow' reaction (in contrast to the 'rapid' partitioning of ester between brine and oil phases), the SW tests of TOMICH ET AL. were designed such that most of hydrolysis (i. e., production of second tracer) occurred during a sufficiently long SHUT-IN stage, between PUSH and PULL. The need for rather long test durations raised concerns about tracer-plume drift-away by virtue of natural groundwater flow, and the analysis by TOMICH ET AL. was mainly devoted to correcting the measured tracer signals for background flow, before inverting for residual-oil saturation.

With (B), we are thinking of mainly covalent-type sorption, whose temperature dependence largely follows some 'Arrhenius'-like law, implying (LICHA 2009) a dramatic decrease of its effects beyond certain temperatures (in contrast to ion exchange processes, whose decrease with temperature is much weaker). According to the temperature-dependent reaction rates estimated by LICHA (2009), covalent-type sorption can be assumed as negligible, for most tracers of interest, at typical geothermal temperatures. From this perspective, covalent-type sorption of tracers within geothermal reservoirs were

–   neither a reason for fearing large tracer losses during IW tests,

–   nor a suitable instrument for quantifying fluid-rock interface areas in IW or SW tests.

However, as a unexpected side effect, it is precisely this temperature dependence that renders covalent-type sorption interesting for use in geothermal SW tests. If (prior to, and) during the PUSH stage of a SW test a sufficient volume of cold water is injected alongside with some sorptive tracers, then a sufficient amount of tracer will sorb onto rock surfaces, as long as surrounding temperatures remain low enough. During subsequent SHUT-IN and PULL stages, surrounding temperatures will steadily increase, mobilizing more and more of the sorbed tracer from rock surfaces back into solution. Owing to their prior retardation induced by PUSH-stage sorption, the radial distributions of more-sorptive tracers will be centered closer to the well, while those of less-sorptive tracers will be centered farther away from the well. Therefore, during PULL, the most-sorptive tracers will appear first, and the non-sorptive tracers will appear last. Thus, a difference in arrival time will be seen between the more- and the less-sorptive tracers, and also a difference in BTC peak height, with the more-sorptive showing higher signals ('in-situ enrichment' of tracer!). In other words, owing to temperature–modulated sorption, the sorptive tracers experience retardation of their transport only during the PUSH but not during the PULL stage of a SW test; thus, their retardation can no longer be "reverted" by virtue of flow-field reversal. PUSH-stage retardation remains visible during the PULL stage, the flow-field reversal only reverting its order of appearance: the more-retarded tracers will be seen earlier, the less-retarded will be seen later.

Essential prerequisite for the applicability of (B) is that sorption distribution coefficients at injection (PUSH) water temperatures not exceed a certain threshold, and tracer solubility into brine at final PULL temperatures be high enough, such that the whole tracer amount that was sorbed during the PUSH stage can be de-sorbed at higher temperatures during the PULL stage, without reaching a concentration plateau (maximum solubility) in the brine. Therefore, ideal candidates for geothermal SW application, in the sense of (B), are tracers with excellent solubility in reservoir brine, and moderate covalent-type sorption at reservoir rock surfaces.

MODEL SIMULATIONS

In order to illustrate the advantages of (A) and (B), we comparatively simulate SW and IW tests using partitioning and sorptive tracers (alongside with a reference tracer), with the properties and flow-field configurations listed in Table 1. The model system contains

–   a mobile-fluid phase, flowing through...

–   ...a homogeneous porous medium, at whose rock surfaces tracers (B) can sorb, or alternately hosting a immobile-fluid phase into which tracers (A) can partition.

Table 1:   Tracer species, tracer properties, and tracer-test configurations used in model simulations.

tracer label, and properties		mobile-phase reactions	retardation factor from equilibrium-exchange processes	injected where	injected when
1	'ester'	$1 \rightarrow 2$ with reaction rate coeff. κ_1	$R_1 > 1$, calculated from immobile-fluid phase saturation s, and distrib. coeff. K_d	push-pull well	beginning of PUSH phase
			Parameters κ_1 und K_d are assumed as known independently from the field SW test (they can be measured in laboratory experiments). Parameter s is to be determined from a SW test (model simulations are to show whether the SW method provides sufficient sensitivity w. r. to this parameter). Parameters κ_1, s, K_d are assumed as homogeneous in space, and constant in time; their possible temperature dependencies are ignored for the time being.		
2	'alcohol'	$1 \rightarrow 2$	$R_2 = 1$		
3	'reference' [SW]	no chemical reactions in the mobile-fluid phase	$R_3 = 1$		
4	'partitioning' or 'sorptive' [SW]		$R_4 = R_1 > 1$		
5	'reference' [IW]		$R_5 = 1$	in a finite distance x from the push-pull well	beginning of PULL phase
6	'partitioning' or 'sorptive' [IW]		$R_6 = R_1 > 1$	(similar to a monopole-convergent IW test)	

The symbolic names 'est' and 'alc' were kept for (A)-type tracers, not only for ease of comparison to the results of TOMICH ET AL. (1973), but also because this general reaction pattern (ester hydrolysis) has proven useful for a variety of oilfield, geothermal or CCS reservoir testing purposes (ROBINSON 1985, CHRYSIKOPOULOS 1993, LICHA 2009, NOTTEBOHM ET AL. 2010, 2011, SCHAFFER ET AL. 2011). In the context of this paper, they generically stand for two tracers with different (contrasting) partitioning or sorption behavior, of which one is produced from the other by a chemical reaction occurring (slowly) in-situ. It is not a necessary requirement that the product tracer always be less partitioning than the source tracer (the alcohol produced in TOMICH ET AL. was soluble only into the brine phase). It is, however, a prerequisite that the 'est' → 'alc' transformation take a finite time, from a practical point of view:

– the reaction should not be instantaneous, as it has to be slow, compared to the PUSH

duration of the intended SW test (which cannot be 'accelerated' without limit);

– the reaction should also not be too slow, because a long SHUT-IN duration (to allow for reaction) increases the risk of tracer-plume drift-away (this issue was analyzed in detail by TOMICH ET AL.).

Tracer tests no. 5 and 6 were included only for the sake of appreciating the parameter sensitivities attainable in SW tests against those attainable in IW tests along similar travel distances. The scenarios listed in Table 1 can be simulated using, for instance, FEFLOW 5.4x (DHI-WASY 2008), with a multiple-species, axially-symmetric flow and transport model.

RESULTS

Figures 2 and 3 demonstrate the enhancement of SW test sensitivity w. r. to target parameters by virtue of (A)-type or (B)-type tracer behavior.
Figure 2 compares the signals of (A)-type tracers and of reference tracers during SW and IW tests (as

defined in Table 1); it further shows how 'daughter'/'source' signal ratios evolve during PULL. Tracer BTCs are shown for three values of the target parameter s, which stands for

- phase saturation $s = \{0, 4\%, 8\%\}$ in the case of tracers partitioning between fluid phases, or for

- fluid-rock interface density $s =$ ibid. divided by the equivalent aquifer thickness, multiplied by porosity/(1–porosity), in the case of sorptive tracers (partitioning now being between fluid and rock).

(Actually, the BTCs in fig. 2, labeled like for two-phase fluid partitioning, were simulated, using FEFLOW, like a sorption process in a single-phase flow in a mono-porous continuum; partitioning into a residual, immobile-fluid phase being described by equations formally identical to those of sorption; conversion between residual-phase volume fraction (dimensionless s) and interface area density (s of dimension $length^{-1}$) resembles the definition found by CARRERA ET AL. (1998) for their 'area parameter' σ_m in the context of matrix diffusion.)

Remarkably, (A)-type tracer BTCs will have similar sensitivity w. r. to target parameter s, irrespective of whether s stands for a '*volume*' or for an '*area*'. Thus, the ideas of TOMICH ET AL. (1973) can be extended to reactive tracers whose daughter products have different sorptivity than themselves. Indeed, such sorption can be seen for some water-soluble esters at sandstone material over a broad pH range, with sorption rates at room temperature much higher than hydrolysis rates, and whose hydrolysis products show no sorption under the same conditions.

From figure 2, one can realize that

- non-reactive tracers with different sorptivity or partitioning yield almost identical signals during PULL; they are insensitive w. r. to the target parameter (s) of the SW test;

- the time-dependent release of 'alc' from 'est' allows to regain sensitivity w. r. to the target parameter s;

- changes in parameter s produce opposite responses of 'alc' and 'est' signals; this enhances their joint sensitivity w. r. to s;

- in the low-s range, a SW test using (A)-type tracers is more sensitive w. r. to s, than a IW test using non-reactive tracers with the same partitioning (sorption) distrib. coeffs.; in this sense, SW tests indeed perform a 'sensitivity enhancement', compared to IW tests;

- if one of the species {'est', 'alc'} happens to be difficult to measure, then it would also suffice to measure only one of them, alongside with a reference tracer; best sensitivity,

however, is obtained from the 'alc'/'est' ratio (which requires measuring both).

Further, the comparison between two different porosity values in figure 2 illustrates the issue of SW ambiguity w. r. to advection. Remarkably, SW behavior w. r. to porosity is equivalent to SW behavior w. r. to dispersion, but this issue will not be discussed in depth here. Summarizing, one can tell that SW test signals are *sensitive w. r. to dispersion*, and *sensitive w. r. to porosity*; but they suffer from *ambiguity between porosity and dispersion* (cf. BEHRENS ET AL. 2009, Ghergut et al. 2011a).

Figure 3 compares between the PULL signals of sorptive tracers in a SW test at constant temperature or with temperature-independent sorption, and the signals of (B)-type, covalently-sorbing tracers during hot-water PULL, following a cold-water PUSH.

From figure 3, one can notice 3 main effects of temperature-modulated sorption upon PULL signals during a SW test conducted with cold-water PUSH:

- *accelerating effect* : 'peak arrival' times during PULL range between ZERO and T_{PUSH}, instead of being all equal to T_{PUSH}; the most-sorptive tracer shows the earliest arrival;

- *'in-situ enrichment' effect* : sorbing tracers produce higher peak signals than a non-sorbing tracer, oppositely to the temperature-independent case;

- *arrival-time sensitivity* (*'chromatographic' effect* : the arrival time of a tracer correlates unambiguously with its sorptivity (expressed by a nominal 'retardation factor' R), and thus with target-parameter s. Remarkably, high 'retardation' turns into *early* arrival!

For shallow-subsurface systems (when fluid-rock interface quantification is of interest there), the temperature dependence of (B)-type tracers could be used the other way round, by first heating the well-screen interval before and during PUSH, then waiting for temperature decrease before PULL. The sensitivity of PULL signals w. r. to sorption parameters will be even higher than was seen in fig. 3 (but without the 'enrichment effect'), approaching the sensitivity of a monopole-convergent IW test along the same (R_{PUSH}) distance. In this case, however, thermal effects on flow during PUSH may require more careful consideration, than in the opposite case of a geothermal reservoir into which a small-sized cold-water slug was injected. On the other hand, if screen-depth temperatures are measurable during the SW test, the additional 'heat push-pull' information (cf. PRUESS AND DOUGHTY 2010, as well as figures 1–2 of BEHRENS ET AL. 2009) can be used to better constrain the inversion of tracer PULL signals.
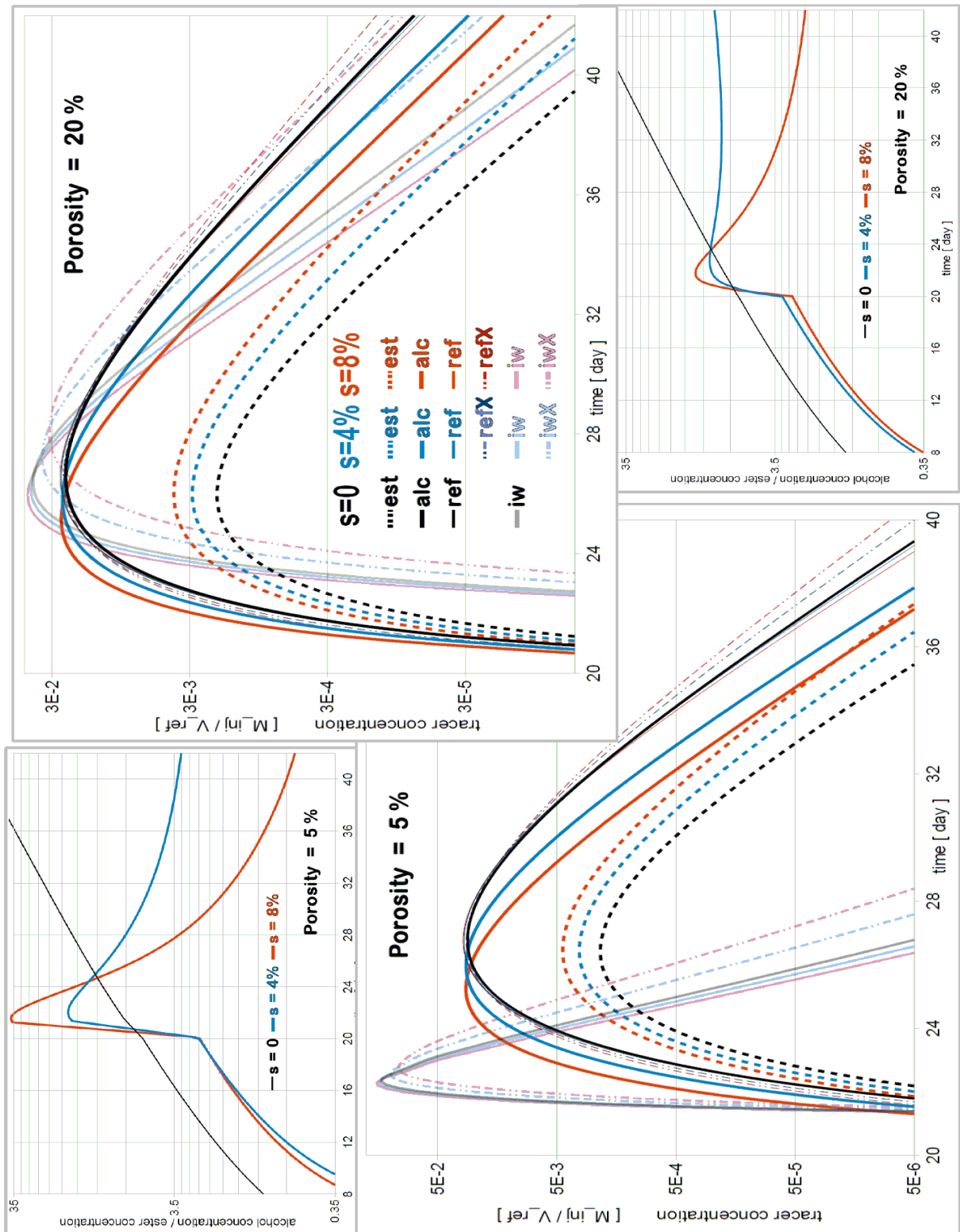
Figure 2: Signals of (A)-type partitioning or sorptive tracers, and of reference tracers during SW and IW tests that were defined in Table 1. Inserts: evolution of (A)-type tracers' signal ratio during SW tests.
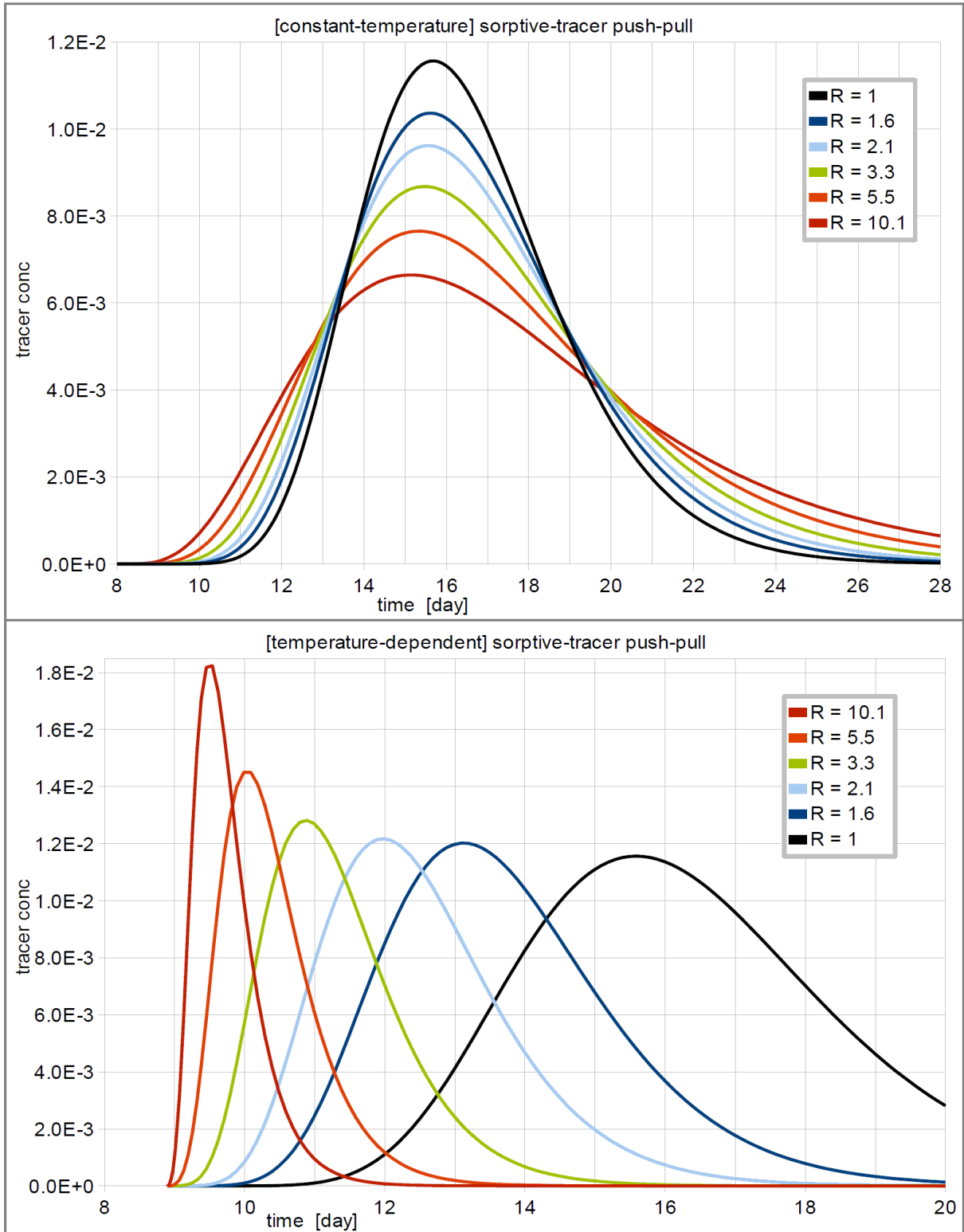
Figure 3: PULL signals of sorptive (R > 1) tracers in a SW test at constant temperature or with temperature-independent sorption, versus signals of (B)-type, covalently-sorbing tracers during hot-water PULL, following a cold-water PUSH. With the former, all tracers are 'pulled' simultaneously, irrespective of R. With the latter, high R values turn into early arrival times!
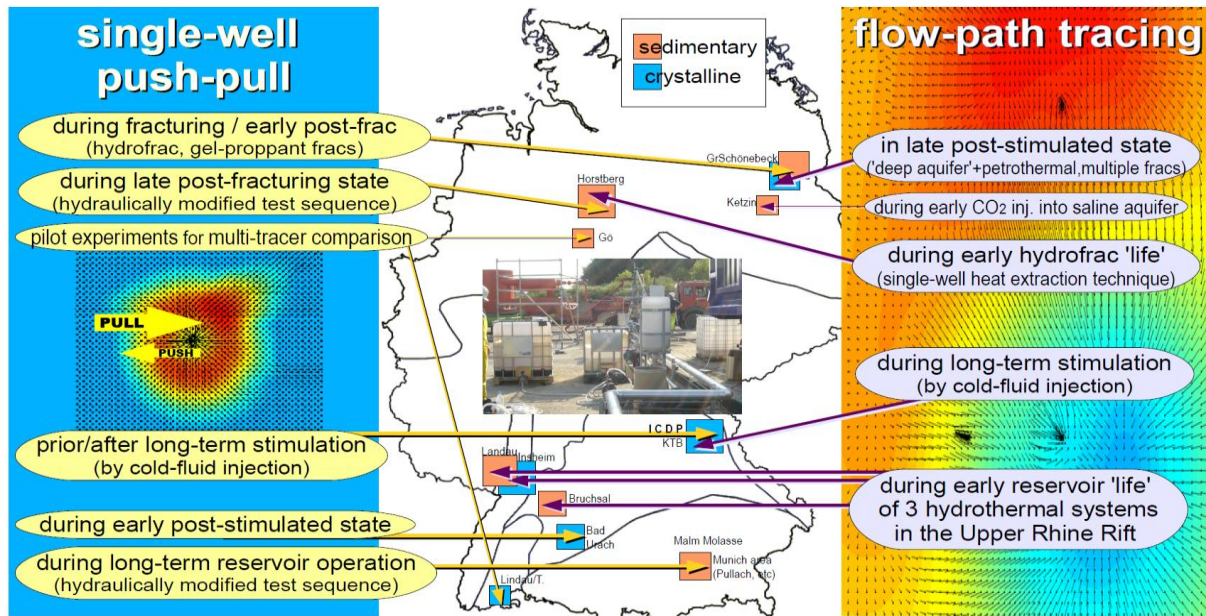
Figure 4: Overview of IW and SW tracer tests conducted Germany, aimed at quantifying flow paths, brine displacement, fluid-rock interfaces, heat exchange areas, and stimulation outcome in deep sedimentary and crystalline reservoirs.

Let us tentatively summarize these findings as follows:

– *owing to in-situ tracer creation in a time-dependent manner* (from another initially-injected tracer with different partitioning or sorption properties),

– *or to temperature-modulated sorption* (covalent-type sorption prevailing),

a satisfactory compromise can be achieved between the IW-typical, advective- or equilibrium-dominated parameter sensitivity regimes, and the SW-typical, advection- or equilibrium-insensitive regimes. Sensitivities thus attained w. r. to target parameter *s* (phase saturation, or interface area density) are intermediate between the poor SW-typical sensitivities and the good IW-typical sensitivities w. r. to (nominal) retardation factors *R*.

REMARKS

First, a caveat: despite a seemingly strong sensitivity w. r. to porosity (cf. l.-h.s. vs. r.-h.s. in fig. 2), attempting to measure effective porosity values from such SW tests remains a tricky endeavor (an issue also discussed by PRUESS AND DOUGHTY 2010 in the context of thermal SW tests, and further by Ghergut et al. 2011).

Further, before becoming too enthusiastic about the advantages of using (A) or (B) for georeservoir characterization, complex laboratory research is yet necessary, regarding

– temperature influences upon (A),

– temperature influences upon (A),

– undesired, or poorly quantifiable hydrogeo-chemical (pH, salinity etc.) influences upon both (A) and (B).

Once having defined a nominally-expected reaction as well as partitioning or sorption behavior for selected tracers based upon laboratory experiments, small-scale field SW tests could be used to check how the tracers perform under georeservoir conditions. As emphasized by LICHA (2009), in multiple-phase systems, species reaction rates and partitioning coefficients may be difficult to measure independently of each other, thus requiring joint inversion from coupled laboratory process models.

The 'quantum dot' tracers with tailorable surface-ligand sorptivity proposed by ROSE ET AL. (2011) appear extremely promising. From the point of view of (B)-type tracers described here, it would be interesting to find ligands with these properties:

– moderate sorption at typical reservoir rocks, at ~10–25 °C,

– covalent sorption prevailing against ion exchange, and showing a significant decrease with temperature in the reservoir-temperature range,

– high solubility in reservoir brine.

Last not least, maybe a suitable compromise can be found between ligand protection (against undesired hydrogeochemical influences) and ligand exposure (to allow for desired sorption).

SOME PROJECTS ADDRESSING GEORESERVOIR (PHASE) VOLUMES AND INTERFACE AREAS

Laboratory experiments for identifying suitable organic tracers and quantifying their reaction and partitioning behavior were conducted by Prof. Licha and Ph. D. students (F. Maier, M. Nottebohm, K. Nödler, M. Schaffer, a. o.) within the projects MUSTANG (on brine-CO_2 partitioning tracers) and LOGRO (on thermosensitive tracers). Modeling of IW and SW tests was conducted within the project GEBO (task unit G6).

A brief overview of field experiments conducted within, or providing tracer-test data to these projects is given in figure 4.

Working-group leaders (Profs. Sauter, Licha, Ptak) certify that no cross-financing occurred for any of the individual tasks between the above-named projects, despite scientific interaction between WG members.

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