We would like to thank the reviewer for his/her feedback and constructive comments. We reply to these comments individually below and will also address them where possible in the revised version. Reviewer comments are reproduced in plain text with underline, with our explanations in *italic*.

Response to reviewer #1

1) This paper describes a coupling between OGS software and iPHREEQC software, for reactive transport numerical modeling. OGS deals with flow, transport and heat transfer, whereas PHREEQC deals with geochemistry. The coupling is of type SNIA. Does it mean that the time discretization scheme is explicit?

Within OGS we can apply both implicit and explicit time discretization schemes for flow, transport and heat transfer. The time discretization for the coupling scheme itself is explicit. The SNIA approach is applied for operator splitting, so no iterations are made between mass transport and geochemical reaction. Consequently, adequate small time step sizes are needed in order to reduce the operator splitting errors. It is worth mentioning that the SNIA approach is sufficient here, as there are no strong feedbacks of reactions on flow and mass transport in the given simulation examples. For applications involving strong feedbacks, we can implement a sequential iterative approach accordingly.
Additional information:
We can also apply a linearized algebraic flux corrected transport (FCT) algorithm (Kuzmin, 2009) which has been implemented to reduce the effects of spurious numerical oscillations for hyperbolic terms (Kosakowski and Watanabe, 2013).

2) Is there any condition on the time step such as a CFL condition?

CFL condition is not embedded in the code itself. However, we have always taken it into account for the spatial and temporal discretization of reactive transport problems, in order to reduce the operator splitting errors. Additionally, if we solve the advection-dispersion-equation (ADE) explicitly, then we need to obey the CFL condition to ensure numerical stability.

3) The flow and transport models can be nonlinear. How is the nonlinearity handled in OGS?

We can use Picard and Newton-Raphson schemes in OGS for nonlinear problems such as Richards flow, density-dependent flow or multiphase flow.

4) Flow and transport and heat transfer can be coupled. How is the coupling handled in OGS?

Within OGS we handle such couplings by using either a monolithic or staggered approach (Wang et al. 2011). With the monolithic approach, all unknowns from different coupled partial differential equations (PDEs) are solved in one global equation system. With the staggered approach, each PDE in the coupling

is solved individually with update of the solutions of the other coupled PDEs in an iteration loop. The staggered approach was applied for the test examples shown in the manuscript.

5) The coupling between OGS and PHREEQC is done with files. This is probably very costly. What is the computational overhead? Would it be possible to develop a more efficient interface between the two software?

This is a very important issue raised by the reviewer. Indeed, the data exchange through files is relatively time consuming, and especially one of the crucial factors for simulating large problems. Additionally, for clusters, in which the file reading and writing is realized through the general parallel file system (GPFS), this process can become more time consuming, especially when the GPFS is highly loaded.
In the meantime, we have developed and implemented a character string-based data exchange for the coupling between OGS and IPhreeqc, and analyzed the performance improvement within different cluster environments. In the revised manuscript we can describe methods, results and their impacts on simulation runs in distributed memory systems.

6) Parallelism is defined with Domain Decomposition in OGS. What does DDC mean? As many cores as domains are used in OGS. How is decomposition done? Does it use METIS for example? How are communications between subdomains defined?

Thank you for these questions to give the reader better insights into parallelization techniques. Domain decomposition (abbreviated by DDC) means the splitting of an initial-boundary value problem (IBVP) into smaller IBVPs on sub-domains. In a more figurative sense, the finite element mesh is decomposed into smaller mesh domains. We will add a short and explaining definition of DDC to the manuscript to make it clearer to the readers.
Yes, we use METIS as a preprocessing tool for DDC in order to balance the node quantities and minimize the border nodes among subdomains efficiently. Then we extract and convert the information about element indices (global) and the internal border nodes and store them in a DDC file. Based on these subdomains and their mesh topology, the global equation system can be partitioned into local equation systems. For coupled processes different local matrices and vectors are assembled for each process and subdomain in individual CPU cores. Then we solve the local equation systems. The local solutions are obtained by a number of product calculations of system matrix and vectors.
Finally, communication is required among sub-domains for updating the iteration steps if the components of local matrices and vectors are associated with border nodes among different sub-domains. Furthermore, communication is also required, when we collect the norm of production from different sub-domains. More detailed information of decomposition procedures can be found in previous works by Kalbacher et al. (2008) and Wang et al. (2009).

7) Geochemistry is trivially parallel, since computations in spatial elements are independent. New cores can be added for these computations, but these cores remain idle when OGS computations are done. Is this efficient?

In the current study, the proposed scheme provides the possibility for a flexible allocation of computational resources for calculation of geochemical and "non-geochemical processes" (e.g. flow, transport). Based on the specific features of the reactive transport problem and the computational

resources that are available on the hardware platform, an allocation of the compute cores can be chosen to obtain the best parallel performance.
It is true that the adding of cores solely for geochemical reaction will lead to the degradation of parallel efficiency, if we can also get further speedup for the "non-geochemical processes" by using these cores. For these problems, we can use the same number of compute cores for geochemical and "non-geochemical processes". All cores will take part in both OGS and IPhreeqc computation. In this case, there are no "idle" cores.
However, adding of compute cores solely for geochemical reactions can be overall efficient for certain problems, in which the degradation of parallel performance for "non-geochemical processes" occurs. In this case, we can adjust the number of cores for flow and mass transport at optimum while increase that for geochemical reactions.

8) Communications seem to be done with files. Is this efficient?

Please see the reply to question 5.

9) Also, it seems that global communications occur quite often. Is this scalable?

In the current coupling scheme, the global communication occurs only when the concentration values of the local buffers are transferred into a global concentration vector, after the calculation of geochemical reactions is done (see page 2379, line 5 - 6). The MPI routine MPI_Allreduce (…) is applied for this purpose ("MPI_Allgather" in page 2379, line 6 is a typo and hence will be corrected in the revised manuscript). We think this operation should provide good scalability.

10) Numerical examples are simple. In fact, 2D and 3D examples remain 1D, since the results do not depend on the (y, z) coordinates. Could real 2D or 3D examples be presented?

It is true that the 2D and 3D example presented in the manuscript are geometrically simple. We selected these examples, since the present manuscript is focusing on the development and analyzing the novel parallelization scheme. However, OGS has already been applied to simulate complex coupled processes (Kolditz et al., 2012). In the current scheme, geochemical reactions are solved locally on each finite element node, which means that it is independent of the complexity of the model geometry.
Nevertheless, we can present more complex 2D or 3D reactive transport problems in the revised version. One example is shown in Fig. 1, in which the sequential degradation of PCE in a hillslope is simulated (modified from Šimůnek et al. 2012).

11) In the examples, geochemistry represents most of the CPU time, so that parallelism is globally efficient. What happens if time of OGS computations becomes higher?

In the 2D example (see Sect. 4.1), geochemistry do represent most of the simulation time. In the 3D example (see Sect. 4.2), however, the time consumption of OGS computation is actually in the same order as that of IPhreeqc computation (comparing Fig. 1a and Fig. 1b). In this case, it is important to allocate enough compute cores for both OGS and IPhreeqc computation to achieve better speedups.

As already mentioned in 7), it is possible that the increase of compute cores can lead to a degradation of parallel performance for OGS computation. In this case, we may still get better speedups by adjusting the compute cores for OGS computation at its optimum while adding more cores for IPhreeqc computation. One of the major advantages of the current parallel scheme is the capability to flexibly allocate computational resources for geochemical and "non-geochemical processes".

Literature

Kalbacher, T., Wang, W., Watanabe, N., Park, C.-H., Taniguchi, T. And Kolditz, O.: Parallelization concepts and applications for the coupled finite element problems, Journal of Environmental Science for Substainable Society, 2, 35-46, 2008.

Kolditz, O., Bauer, S., Bilke, L., Boettcher, N., Delfs, J. O., Fischer, T., Goerke, U. J., Kalbacher, T., Kosakowski, G., McDermott, C. I., Park, C. H., Radu, F., Rink, K., Shao, H., Shao, H. B., Sun, F., Sun, Y. Y., Singh, A. K., Taron, J., Walther, M., Wang, W., Watanabe, N., Wu, Y., Xie, M., Xu, W., and Zehner, B.: OpenGeoSys: an open-source initiative for numerical simulation of thermo-hydro-mechanical/chemical (THM/C) processes in porous media, Environ Earth Sci, 67, 589-599, 2012.

Kosakowski, G. and Watanabe, N.: OpenGeoSys-Gem: A numerical tool for calculating geochemical and porosity changes in saturated and partially saturated media, Phys. Chem. Earth, 70-71, 138-149, 2014.

Kuzmin, D.: Explicit and implicit FEM-FCT algorithms with flux linearization, J. Comput. Phys. 228, 2517–2534, 2009.

Šimůnek, J., Jacques, D., Šejna, M., van Genuchten, M. T.: The HP2 program for HYDRUS (2D/3D), A coupled code for simulating two-dimensional variably saturated water flow, heat transport, solute transport and biogeochemistry in porous media (HYDRUS+PHREEQC+2D), available at: http://www.pc-progress.com/Documents/HYDRUS3D_HP2_Manual.pdf, last access: 9 April 2015.

Wang, W. Q., Kosakowski, G., and Kolditz, O.: A parallel finite element scheme for thermo-hydro-mechanical (THM) coupled problems in porous media, Comput. Geosci., 35, 1631-1641, 2009.

Wang, W., Schnicke, T. and Kolditz, O.: Parallel finite element method and time stepping control for non-isothermal poro-elastic problems, CMC-COMPUTERS MATERIALS & CONTINUA, 21, 217-235, 2011.

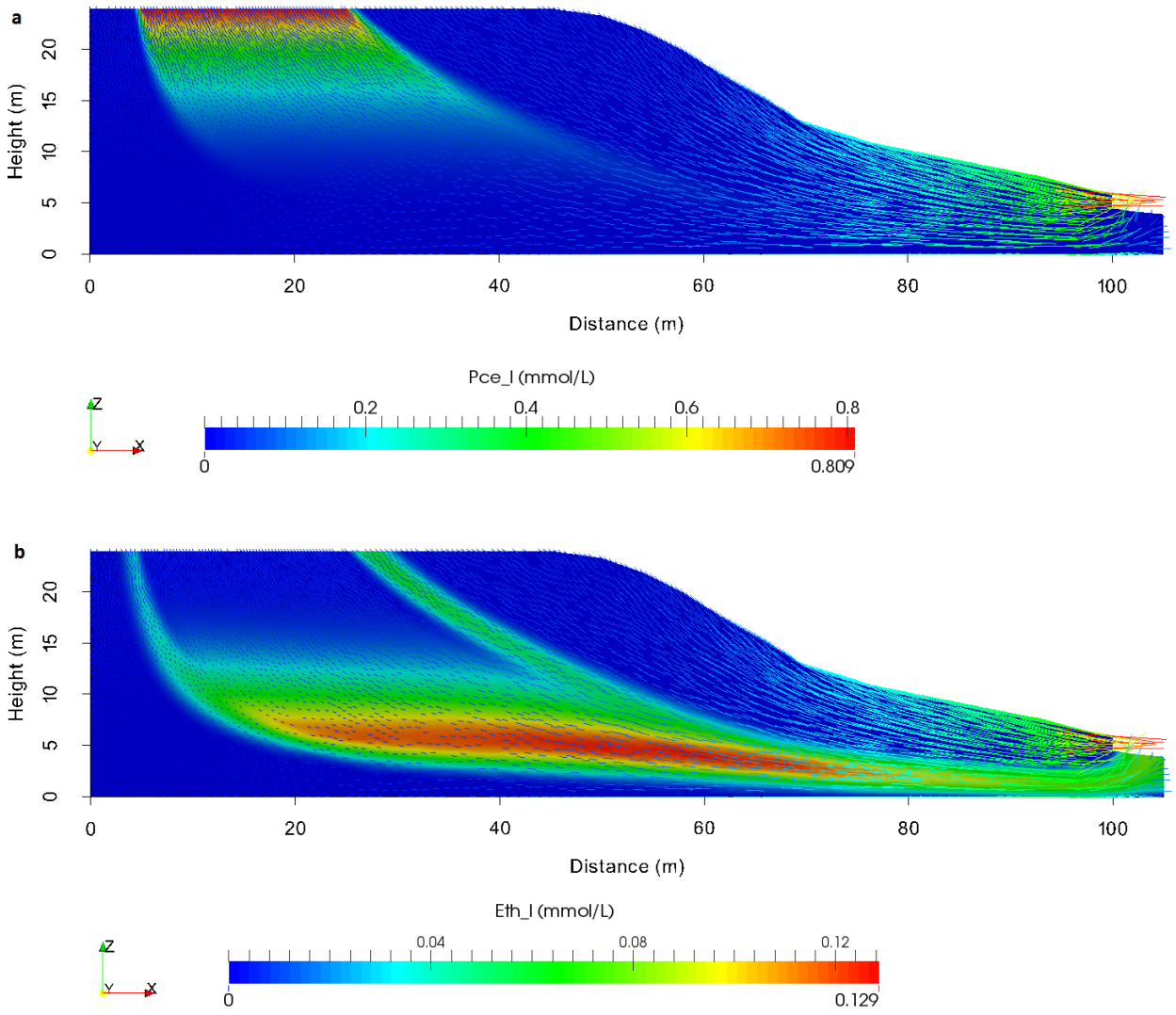

Fig. 1 Concentration profiles of light isotopologue of PCE (a) and ETH (b) together with streamlines in a hillslope. The concentration of PCE is fixed at the top of the model domain.