The work presented here by Nogueira et al concerns the evaluation of the reactivity of the hyporheic zone, and more globally of a small stretch of a river-aquifer interface that is in a losing condition. The paper is well written and proposes a new framework for assessing the potentiel reactivity of this interface by coupling a physically based model, that simulates stream water (SW) and groundwater (GW), with a Hydraulic Mixing Cell (HMC) module. However it suffers from various flaws that must be addressed before further publication.

We are grateful for the reviewer's constructive comments, which have helped to clarify unclear points in the paper and to generally improve the manuscript. We also appreciate the suggestions for additional points and references in the discussion of our work, which we believe have enriched not only this section, but also the overall quality of the manuscript. Below are the detailed responses to the reviewer's comments.

1. The system at hand must be detailed and a significant effort must be done for positioning it in a much broader picture (see for instance the review of Flipo et al. 2014). From the title and the abstract, it must be clear that the focus of the study is a 900m stretch of a river connected to a small portion of porous medium (the lateral extent of the model seems too narrow to speak about an aquifer). The river is in a losing condition, which is not the most probable configuration as far as SW-GW are concerned since rivers constitute the water outlet for GW at the catchment scale. Finally the connection of the small portion of river stretch with a broader regional aquifer system must be explained.

   We recognize that some points may not have been clear at the beginning of the abstract and in the introduction of the manuscript. We have revised those sections according to the suggestions of the reviewer. However, we would like to additionally emphasize the following points:

   We disagree with the notion of the reviewer that the narrow lateral extent of the water bearing alluvial sediments around the stream does not justify the use of the term aquifer. The term aquifer is primarily defined in terms of a water bearing subsurface layer's permeability and transmissivity for water (to distinguish it from an aquitard or aquiclude) and not in terms of its spatial extent. The hydraulic conductivities in the gravelly and sandy formation at the site with a thickness of up to 8 m are on the order of 10^{-4} to 10^{-3} m/s (see Nogueira et al. 2021, Trauth et al., 2018; Zhang et al., 2021), which is highly permeable and justifies to call the formation an aquifer. We refer to Lohman et al. 1972 for a definition (*Definitions of selected ground-water terms – revisions and conceptual refinements*, USGS Water Supply Paper, 1988). Borelog and geophysical data indicate low-permeability clayey and silty deposits (forming an aquitard) on top of a relatively shallow, Mesozoic, vertically tilted bedrock (inhibiting lateral groundwater flow) at the base of the coarser water bearing alluvial sediments, hydraulically disconnecting the alluvial aquifer from potential groundwater in the deeper fractured bedrock. We refer to the German geological survey for information related to the underlying bedrock, which is available online at: https://produktcenter.bgr.de/terraCatalog/OpenSearch.do?search=61ac4628-6b62-48c6-89b8-46270819f0d6&type=/Query/OpenSearch.do.

We made sure to emphasize this geological setup and the associated geological information in the revised manuscript, and have added the relevant references on the description of the study site and on the presentation of the numerical model. We also brought this point back in the discussion section as suggested by the reviewer (see answer to question 3 below). It now reads (section 2.1):

> *The alluvial aquifer consists of up to 8 m-thick alluvial sediments, with grain sizes ranging from medium sands to coarse gravels, underlain by less permeable clayey-silty deposits on top of the Mesozoic bedrock forming the bottom of alluvial aquifer. Borelog and geophysical data indicate that the thickness of the alluvial aquifer slightly decreases with distance from the stream (Lutz et al., 2020; Trauth et al., 2018).*

We also respectfully disagree with the statement that losing conditions are an unlikely condition in river catchments. While we agree that at regional-scale groundwater will eventually discharge to surface water bodies such as rivers, lakes or estuaries, river reaches with temporarily or even permanently losing conditions are by no means uncommon and are not only found in arid or semi-arid regions. Local losing conditions on streams may be caused by several natural (geology, topography and climate) and human-induced conditions (Brunner et al., 2009; Irvine et al., 2012; Jones et al., 2008; Liao et al., 2014; Munz et al., 2019; Poole et al., 2008; Schilling et al., 2017; Su et al., 2007; Treese et al., 2009; Vogt et al., 2010a, 2010b). Our field site is just downstream of the transition between the steeper, mountainous upper- and the flatter, alluvial lower catchment of the Selke river. This location is on the lee side of the Harz Mountains, which block parts of the westerly winds that deliver most precipitation in Central Europe. Therefore annual precipitation is relatively low (~500 mm) and in turn groundwater recharge rates are small (on average < 100 mm) facilitating the disconnection between the alluvial aquifer and the stream. Such disconnections are a rather common condition on the lee side of mountains in larger river catchments, even in temperate and more humid climate regions. Furthermore projections indicate that losing conditions are becoming more common due to global change and increasing groundwater withdraws within alluvial aquifers (Jasechko et al., 2021).

Alongside the HMC application, with our study we also aim to emphasize the importance of losing streams in the role of riparian biogeochemical processes, especially on mixing-dependent reactions. Previous studies have already demonstrated the role of losing streams in providing DOC and other solutes to trigger and boost riparian biogeochemical turnover processes (Hester et al., 2013, 2014, 2017, 2019; Lutz et al., 2020; Munz et al., 2019; Trauth et al., 2014, 2018; Trauth and Fleckenstein, 2017), which is especially important for the turnover of groundwater-borne solutes, such as the case of Nitrate in the studied area (Gassen et al., 2017; Lutz et al., 2020; Trauth and Fleckenstein, 2017). As suggested by the reviewer, we have further emphasized this point in the abstract and in the description of the study area.

We agree with the reviewer that the connections of our findings to broader scale processes could be outlined more explicitly in our manuscript. We now indicate the connections and the relevance of our study for larger scales at different points in the text (see for instance answer to question 3 below). However, at the same time we want to be careful not to make statements at scales that are clearly beyond the scale and data of this study.

2. The main conclusions highlighted in the abstract only make sense if it is clearly stated beforehand that the stretch of river is in a losing configuration otherwise readers could be misled at the reading of the abstract. On the one hand, the highlighted results of evaluated water mixing values should be moderated in the abstract considering the remark 3. On the other hand, it seems to me that an important result of the study is not sufficiently reported in the abstract, it is the fact that the potential hot spots of reactivity of such a system in terms of nitrate removal is located at the fringe of the HZ and not directly below the leaking river.

   We have emphasized the net losing conditions of the local reach in the abstract. We have also rewritten the main findings of our study in the abstract taking the suggestions into account (referring the computed percentages in terms of the model domain rather than the riparian aquifer). We agree with the reviewer that the original statements were not sufficiently highlighting the important key finding on the location of the potential hot-spots of reactivity in terms of distance from the stream channel. The respective text in the abstract now reads:

   > *Our results show that on average about 50% of the water in the alluvial aquifer consists of infiltrating SW. Within about 200m around the stream the aquifer is almost entirely made up of infiltrated SW with practically no significant amounts of other water sources mixed in. On average, about 9% of the model domain could be characterized as "mixing hot-spots" (locations with more balanced fractions of the different water sources), which were mainly located at the fringe of the geochemical hyporheic zone rather than below or in the immediate vicinity of the streambed.*

3. The GW model set up must be detailed. What is the extent, in the x, y and z directions ? what are the lateral boundary conditions and also at the bottom of the system, as well as for the upstream part of the simulated porous media. If no water flux conditions are used for the lateral and the bottom of the porous medium system, it has consequences on the presented result, entailing them with a large uncertainty related to the misconception of the connection of the system to the larger regional aquifer system. A discussion on the consequence of the model set-up should be added to the paper.

   We have added respective information on model setup and its extent. It now reads:

   > *The simulated domain (900 × 770 × 10 m) was divided into four main hydrogeological units according to geophysical and borelog data, which further indicates the thinning of the alluvial aquifer with distance from the stream (Lutz et al., 2020; Trauth et al., 2018). Thus, the simulated domain covers most of the mapped alluvial aquifer present in the area. The bottom of the numerical model was set as a no-flow boundary in line with the less permeable clayey-silty deposits and the low-permeability bedrock at the base of the coarser alluvial sediments. The boundary conditions (BCs) on the model surface domain were defined as (i) groundwater recharge (as a fraction of daily precipitation) at the model top, (ii) specified water flux at the model stream inlet according to discharge values measured at a gauge station about 3000m upstream of the study site, and (iii) a critical depth BC at the model stream outlet (Fig.3a). The BCs on the subsurface model domain were defined as (iv) specified water flux representing ambient groundwater flow at the upstream side of the model, and (v) prescribed time-varying hydraulic heads at the downstream side of the model (Fig.3a). The other lateral subsurface boundaries of the model domain were set as no-flow boundaries based on field observations indicating that GW flowlines are somewhat parallel to the stream with distance.*

   For more details on the model setup we refer the readers to Nogueira et al. (2021).

We have also added a discussion on the consequence of the model setup to the larger-regional aquifer system. We agree with the reviewer that this point was not discussed in the text. We highly appreciate this suggestion from the reviewer. It now reads (section 4.5):

> *Even though the numerical model matched field observations well, it represents a simplification of reality (a characteristic inherent to all models), which in turn results in some limitations and uncertainties. For instance, based on available geophysical data we have assumed the clayey-silty formation on top of the vertically tillted low-permability bedrock as the bottom of the alluvial aquifer and impermeable in the model. We assumed that the alluvial aquifer has a limited lateral extent (Lutz et al., 2020; Trauth et al., 2018), which was backed by geophysical data and the presence of bedrock outcrops along parts of the lateral model boundaries. These assumptions and the chosen model geometry, however, may not fully account for larger-scale hydrological fluxes, which are inherent to nested SW-GW systems. For instance, as showed by Flipo et al. (2014) and by other studies (Boulton et al., 1998; Magliozi et al., 2018; Toth 1963), SW-GW system are connected interfaces, which are linked to each other through different spatio-temporal processes. For instance, longer and deeper flowpaths that might have been not represented in our numerical model could lead to the development of additional mixing spots at greater depths or distances from the stream (Lessels et al., 2016). This could further emphasize and explain how alluvial aquifers and riparian zones act as buffer zones connecting low-frequency processes occurring at regional scale and high-frequency processes occurring in the stream network (Ebeling et al., 2021; Flipo et al., 2014; Rivett et al., 2008; Sun et al. 2017). Equally, lateral influx of groundwater through the lateral boundaries of the model domain could also effect the dynamics and main directions of GW flow paths and therefore SW-GW mixing spots development. However, head data at the site did not show any indications of such effects. Furthermore the specific geology of the site with shallow, low-permeability mesozoic bedrock strata, which inhibit lateral groundwater movement as they are vertically tilted, rules out the presence of a laterally extensive, continuous regional aquifer. Exchange fluxes between the shallow alluvial aquifer with deeper groundwater were therefore considered to be negligible.*

4. One way to clarify the paper is to add a summary of the other Nogueira et al papers

   We appreciate this suggestion. We have added additional information from our earlier paper throughout the text taking into account the comments from the reviewers. For more details, the readers are directly referred to Nogueira et al 2020 and 2021.

5. The added value of using HMC rather than a fully coupled transport model is not clear and is in the current state of the paper an affirmation, not a scientific statement. As it is stated that Nogueira et al in press used the transport module of HydroGeoSphere, a comparative assessment of computational duration should be provided. This quantification is essential because from line 609-618 it seems more efficient to directly use a transport model than a HMC for the quantitative assessment of the stream-aquifer interface in terms of nitrates removal.

   We agree that the HMC framework presented here does not allow for a direct quantification of nitrate removal rates. It is rather a complementary tool that indicates locations where the potential for removal can be high due to SW-GW mixing. For a direct quantification, reactive-transport models or additional field data in combination with data-driven analyses would be needed. However, the parameterization of such a transport model requires significant amounts of spatially distributed data (e.g. local concentrations) to constrain parameter ranges (e.g. reaction rate coefficients). Instead we decided to use the field data from Gassen et al. (2017) for a more qualitative evaluation of our simulated patterns of mixing potentials obtained from the robust HMC model, assuming that reactions facilitated by the mixing of the different water sources can explain the sharp concentration fronts observed by Gassen et al. (an assumption also implicitly made by the authors of that study). In that regard results from the HMC method, which can be well constrained with the existing hydraulic data and does not require extensive data on transport and reaction parameters, can serve as a proxy and complementary tool to interpret observed concentration patterns. We have emphasized these points in the discussion section 4.5.

We would like to further clarify that Nogueira et al. (2021) did not use a transport routine within HGS. Instead they developed a sequentially coupled reactive-transport model based on the flow simulations from the HGS and other field data since HGS does not allow for temperature-dependent reaction rates to be implemented at the moment. Therefore a comparison of computation-times between this model and the model in this study would not be meaningful. To clarify this we have rewritten the text in the respective section of the manuscript. It now reads:

> *We intentionally did not conduct explicit simulations of reactive transport in this study since our main goal here was to explore the HMC method (coupled to a flow model) to assess the development of mixing spots on the riparian zone and their relation to hydrological variations. Spatial patterns of mixing hot-spots can provide a meaningful proxy for the interpretation of reactivity patterns in the absence of extensive data for the parameterization of an explicit reactive transport model. Along those lines we could illustrate the importance of such macroscopic mixing spots for groundwater-borne* NO_3^- *turnover by comparing the quantitative mixing results of the HMC method with previous biogeochemical assessments carried out in the study area. For a direct quantification of nitrate removal rates, however, the use of reactive-transport models or additional field data combined with data-drive analyses would be needed. Such simulations would have allowed a comparison of observed and simulated concentration values and their dynamics for a more rigorous evaluation of model performance (Nogueira et al., 2021b).*

6. Errors in mathematical formulas are unacceptable and must be corrected :
    1. Eq 1. f_(w)^ t-1 not defined, as well as vbc_k^t

       Thank you for pointing this out. Vbc_k^t should be Vbc_w^t in the equation as it is in the text (we have replaced k for w in the equation). On the other hand, we believe that the terms $f_{i(w)}^{(t-1)}$ and $f_{j(w)}^{(t-1)}$ have been defined in the text just after the equation is presented.

    2. Eq 2 not homogeneous in terms of units between left hand side and right hand side

       Thank you for bringing this to our attention. There was a missing term on the equation and in the explanatory text. We have corrected the problem. It now reads:

       $$V_w = \frac{\sum_{p=1}^{P} (V_p f_{w,p})}{V_{tot}} \times 100\%$$

       In line with the suggestion from the reviewer to clarify the *Integration* function (see answer to other remarks 5), we have also added the following sentence before the Eq.2:

The function integrates the numerical cells within the simulated domain taking into account only the fraction of interest that comprises each cell volume. The calculation sums the resulting quantities over the domain to produce the integrated result, which is then normalized by the total volume of the simulated domain (V_{tot}). Thus, the resulting volume represents a percentage of the total simulated domain

3. Eq 4 the denominator seems wrong, please check and either add the original reference or detail the math. L 250 the value of the denominator of eq 4 in case only two pools of water are concerned is 1, root square(2)/2 as stated by the authors.

   Based on comments from the other reviewer, we have now added more explanation on the development of the equation and on how the mixing degree can be calculated based on an analytical geometry approach. We have added a figure to the supplementary material (see now Fig.S2 – also attached at the end of this document) to illustrate the concept.

   In brief, for a three end-member mixing case, any combination of three different source water fractions can be represented as a point *d* in a 3D coordinate space, in which the maximum Euclidian distance between point d and the point of equal mixing (equal fractions of all mixing members) within the mixing space is the radius of a circle (centred at [1/3, 1/3, 1/3]) escribed on an equilateral triangle (side length of $\sqrt{2}$). For a two end-member mixing case, the maximum segment is the diameter of a circle (centred at [0.5, 0.5]) with side length of $\sqrt{2}/2$. For a four (or more) end-member mixing case a spatial representation is not possible but equation (4) would equally apply. Therefore we would like to keep it in the text.

4. Same problem in eqs 5 and 6

   Equally to Eq.2, there were missing terms on the equations and in the explanatory text. They now read:

$$V_d = \frac{\sum_{p=1}^{P} (V_p\, d)}{V_{tot}} \times 100\%$$

$$V_{d\_HZ} = \frac{\sum_{p=1}^{P} (V_p\, d)}{V_{HZ}} \times 100\%$$

7. The discussion about the reactivity of the interface should be enriched with other important references such as Newcomer et al. 2018, especially providing arguments on the added value of a 3D approach.

   We have rewritten this discussion following the suggestion from the reviewer on the added value of our 3D approach. We recognize that this has improved this section. It now reads (section 4.3):

Previous work on hyporheic reactivity has often been carried using 1D or 2D model setups focusing on biogeochemical processes in direct vicinity of the streambed (Hester et al., 2014, 2019; Newcomer et al., 2018). This study, using a larger-scale 3D model also considers lateral SW-GW exchange fluxes over longer distances into the riparian aquifer the associated longer-term mixing processes further away from the stream channel. In line with results from Nogueira et al. (2021b) and Trauth et al. (2018), results from our 3D model coupled with the HMC method reinforce that such larger-scale and long-term processes are important around losing streams for the creation of mixing hot-spots at larger distance from the stream. These mixing hot spots can facilitate mixing-dependent biogeochemical reactions, which may significantly contribute to the net turnover of groundwater-borne solutes at the stream corridor scale. These processes may have been overseen in small-scale studies, which have focused on the immediate interface between the stream channel and the alluvial groundwater only.

Nevertheless, we would like to emphasize that Newcomer et al., 2018 did not consider groundwater-borne solutes such as Nitrate ("*we simplify our model to the scenario where groundwater NO_3^- contamination is not present*"), and thus could not evaluate the links between SW-GW mixing and mixing-triggered reactions, which was the focus of our study. In turn, they have only considered stream-borne solutes (e.g., DOC, NO_3^-) and their turnover in the hyporheic zone below the stream bed. We also believe that the Newcomer et al. study, although it enriches our discussion, is less in line with our study than others studies, which have explicitly included groundwater-borne Nitrate in their simulations, for instance the studies by Hester et al. (2017, 2019) and Trauth et al. (2014).

8. Sec 2.4.1 the authors mention that the origin of water from the flood plain can be neglected, then developing eq 4 in that specific case. It is confusing since they use 3 origins in the remaining of the paper. Section 2.4.1 must be reworked l235-271

We recognize this section was confusing and we have rewritten it in order to clarify the idea behind the reduction from a three to a two end-member mixing model. We still think, however, that it is important to keep the three end-members represented throughout the manuscript since there are time periods when the three components are all present in the saturated portion of the domain in high fractions. These episodes are an important characteristic of the temporal exchange dynamics of the coupled GW-SW system. Therefore we would like to keep the three end-member case in the manuscript. We hope that with the additional explanation this idea is now clear in this section. Following Eq. 4, it now reads:

where f_1, f_2, and f_w represent HMC fractions. Based on preliminary results, we have observed that actual volumes of f_{FW} were very low in comparison to f_{GW} and f_{SW} in the fully saturated portion of the domain as it will be demonstrated in section 3.2. This occurs because recharge from precipitation is very low at the site (Nogueira et al., 2021b), and the percolation of water from the top of the model domain is limited to occasional episodes. Therefore, we have employed a simplified version of the Eq.4 considering a two end-member mixing only. To do so, we combined the two end-members f_{GW} and f_{FW} to a single one (e.g., [f_{GW}+ f_{FW}], Fig.S2, supplementary material), which reduces the mixing model to a two 2D case. This streamlined two end-member mixing is the preferred one used throughout the manuscript because otherwise resulting d values would be consistently very low in the simulations, which would impair their further analyses.

Other remarks

1. L. 127 Please write the explanation of Fig. 2 in a paragraph at the beginning of section 2 Method. It is not currently detailed, only the Figure is in the document.

   We have added a sentence to briefly explain the figure in the text. It now reads:

   > *In brief, following field data collection, a 3D numerical flow model was developed and calibrated against the collected field data (Nogueira et al., 2021b). The HMC method is then coupled to the numerical model, whereas results are additionally evaluated according to additional hydrochemical data (i.e., water samples) for further mapping of water sources and analysis of mixing degrees within the riparian zone. In the subsequent sections we detail each step and the methods used.*

2. L. 141 AT each time step

   We have corrected it.

3. Fig 3a. Scales are not readable, especially in the Z direction. Overall the readability of the whole figure must be improved. The reader should be able to read the piezometer names

   We apologize for that. We made sure to increase the legend size, as well as the names of the piezometers to guarantee their good readability. We also increased the indication of the elevation isolines and changed their colors slightly to improve their readability.

4. L 180 grammar issue

   We have corrected it.

5. L 196 what is the integration function of Tecplot, please explain the math instead

   We have added the following sentence in order to clarify the utilized function just before the Eq.2:

   > *The function integrates the numerical cells within the simulated domain taking into account only the fraction of interest that comprises each cell volume. The calculation sums the resulting quantities over the domain to produce the integrated result, which is then normalized by the total volume of the simulated domain (V_{tot}). Thus, the resulting volume represents a percentage of the total simulated domain*

6. L 204 50% OF stream water

   We have corrected it.

7. L210 WHILE most

   We have corrected it.

8. Fig 6 and 7 are too small and therefore not very informative. The authors must select more dedicated illustrations that correspond more closely to their message in the text

   In our opinion, the figures convey key information on the spatial distribution and variations (in terms of expected minimum, maximum and average distribution) of the different water fractions in the simulated domain, as well as of the mixing degrees. In order to clarify this point and their relevance, we have added an extra sentence before the figure. It now reads:

The plots indicate the minimum and maximum possible distributions of each water fraction in the domain, as well as their typical distribution throughout the simulation period.

We have done the same for Figure 7. Moreover, we have enlarged the legends on the figures to improve their readability. We hope this has solved the issue with the figures. As we think they provide key information linked to the mapping of different water fractions and mixing degrees in the simulated domain, we would like to keep them in the manuscript.

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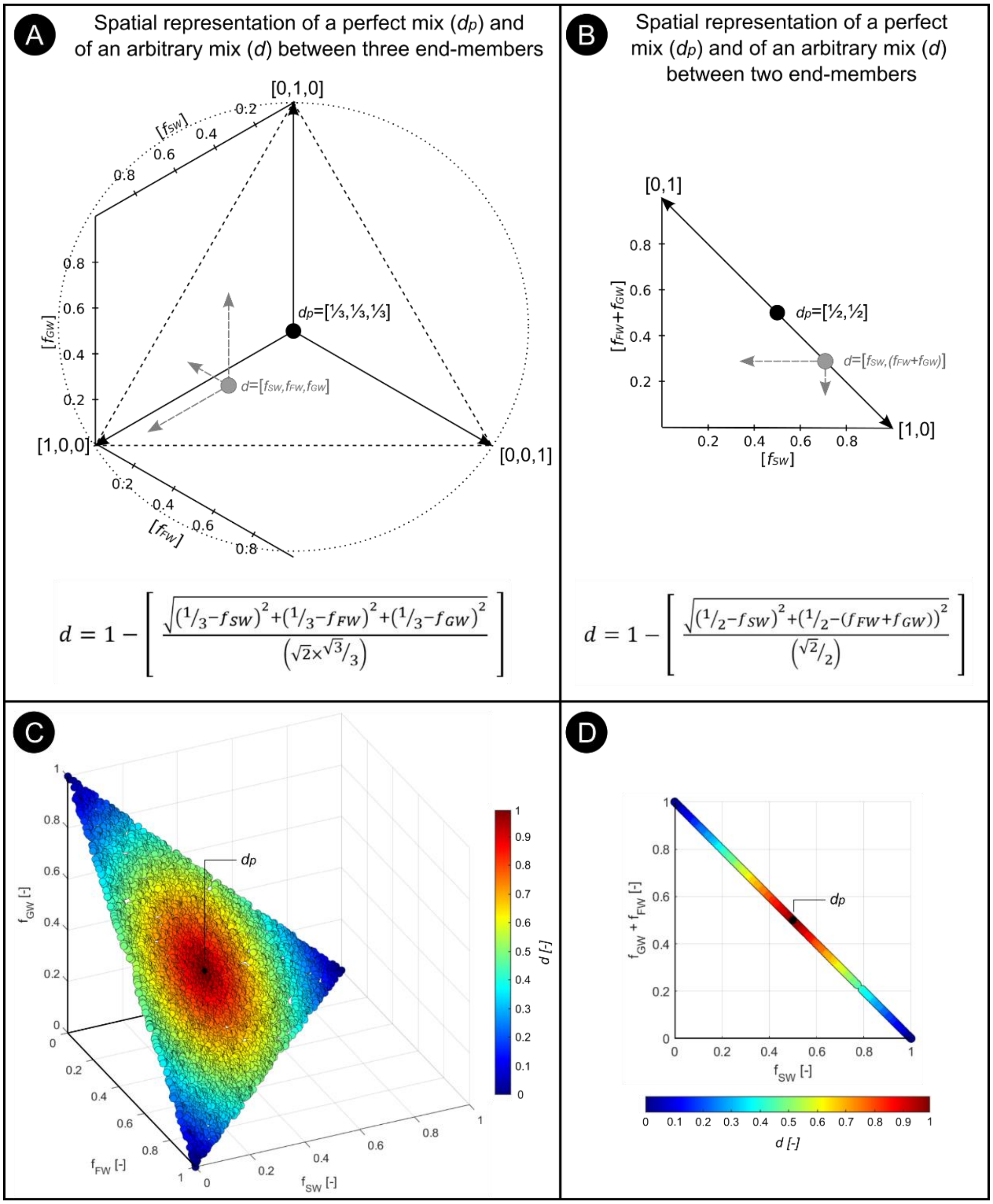

Fig.S2: Spatial representation of a perfect mixing (d_p) and of and arbitrary mixing (d) for the cases of three **(a)** and two **(b)** end-members mixing. The final mixing d can be calculated as the Euclidean distance between points d_p and d. For a three end-members mixing (3D case), any combination of fractions can be represented as a point d in a 3D coordinate space, in which the maximum distance is a radius of a circle (centred at [1/3, 1/3, 1/3]) escribed on an equilateral triangle (side length of $\sqrt{2}$). Thus, the maximum distance between d_p and d is $(\sqrt{2} \times \sqrt{3}/3)$ For a two end-members mixing (2D case), the maximum segment is the diameter of a circle (centred at [0.5, 0.5]), whereas the maximum distance between dp and d is $(\sqrt{2}/2)$. The long-dashed lines in (a) delimit the solution space for any possible mixing d where fractions sum up to 1. In (b) final mixing d values would fall over the solid line passing through d_p. Example of theoretical mixings between three **(c)** and two **(d)** end-members coloured according to computed d values (warmer colours indicate a more homogenous mixing); d_p is indicated as a black circle. The theoretical mixings were generated with 10000 random combinations of HMC fractions that sum up to 1. For a four (or more) end-members mixing a spatial representation is not possible but the general Eq.4 would equally work.