

General comments.


Berg et al. present an interesting and mostly original study on sulfur transformations and sulfur isotope partitioning into different inorganic and organic sulfur fractions in a previously well studied Swiss lake. The isotope geochemical study includes microbiological informations, which is nice, but of secondary importance, since it represents the present situation and not necessarily for the 'paleo'-ones that are responsible for the formation of benthic geochemical signatures. Lago Cadagno is well known for its unusual element cycling, including the development of euxinic conditions due to a groundwater derived carbonate-buffered substantial sulfate supply derived from gypsum and dolomite dissolution in the catchment area.

I suggest to change the title and remove 'active', since no rates measurements of the current sulfur transition have been carried out and are only deduced from sediment solid phase parameters. What is missing is a competent discussion of the oxidative part of the sulfur cycle and relevant associated sulfur cycling.

It is true that rates have not been measured, though steep gradients of redox-active compounds along with clear isotopic signatures in these pools are telling evidence of microbial activity in surface sediments and at the sediment-aquifer transition. We have removed 'active' from the title. We have also added a discussion of sulfur oxidation, especially as a contribution to light sulfate isotopes in deep, glacial sediments to the extent possible in the absence of *soxB* gene identities. We have also cited the 16S dataset of Berg et al (2022) in which some sulfide oxidizers were recovered.

The presentation of data, for instance in the supplementary data, file has to be substantially improved. The labels at X axes of TIS, TOC etc. are missing, the label at the $\delta^{34}\text{S}$ profiles from 1991 are not of publishable format/quality. In general, the basic geochemical data should be presented in the main text.

The supplementary figures have now been imported into the main manuscript (Figure 2). We have ensured that all the axes are correctly labeled and are logically presented in terms of dissolved versus solid phase compounds. We apologize for the poor quality of the 1991 data, which was stored on an original floppy disk. The data has now been retrieved and overlain on the 2019 dataset revealing a high level of consistencies. The surface and deep sediments are now presented in separate figures for better visualization (axes scales).

 Detailed comments:

- I suggest to add the fundamental PhD thesis of Losher (1989) on S cycling in Lago di Cadagno into the introduction and reference list.

The PhD thesis of Losher (1989) has now been cited in the introduction.

-L33: ...catchment groundwater... instead of 'aquifer groundwater'

This was changed from 'aquifer water' to 'groundwater'. Analyses were made on water collected directly from the subsurface springs.

-L33: Is the $\delta^{34}\text{S}$ of lake water column sulfate really constant?

According to Canfield et al. (2010) water column sulfate signatures change the most at the redoxcline, but are relatively constant within the deep, anoxic water which is in contact with the sediment. We have now specified 'compared to sulfate in the lake monimolimnion (+24 ‰)'.

- L34: d34S values for dissolved sulfate in the deduced steep gradient are not measured, but only deduced. Therefore, this statement on apparent isotope enrichment factors is a substantial oversimplification.

The d34S values for dissolved sulfate were in fact measured, but from pooled samples. This simply means that there is a lower depth resolution. The deduction to be made is that the measured d34S of groundwater sulfate seeping into the lake is the same as the sulfate seeping into the deep sediment, but that is not what is referred to here.

-L35: Due to the lack of pore water data, it stays questionable if the term 'closed system' really describes the boundary conditions

In nature it is rather improbable that any system is 100% open or closed and we could only estimate where Lake Cadagno lies on a spectrum. Due to the lack of available porewater data for the deep sediments, we have cautiously rephrased this statement and text in the discussion. The line has been rephrased as follows: 'The isotopic offsets between pools of humic acid-sulfur, acid-volatile sulfur (AVS) and chromium-reducible sulfur (CRS) in both surface and deep sediments suggest differential timing of formation, with sulfide oxidation to sulfur/polysulfides playing an integral role in organic matter sulfurization.' Now lines (31-34).

-L53: I am sure that there are really more basic studies from the 90s of the last century that should be cited here: e.g., Jorgensen, 1990, Thamdrup et al. etc.

These citations are from marine sediments which have historically been greatly studied compared to freshwater sediments. We specifically want to distinguish studies of sulfur cycling in freshwater environments. We have added the study of Hansel et al 2015.

-L57/58 ...cultures and marine sediments... References should include: Kaplan & Rittenberg (1964), Wortmann et al. (Geology, 2001); Rudniki et al., (GCA, 2001)

These three citations have been added.

-L66: add reference: Jorgensen et al. (GCA, 2004)

We thank the reviewer for pointing out this interesting reference which has been added to this line.

-L68: This is not a new result of Bryant et al., but has already been found out in the 80s of the last century and the 90s. You should refer to the original studies.

We have reworded this sentence as follows (L. 80-84): While the $\delta^{34}\text{S}$ -composition of pyrite has been widely used to interpret global changes in the Earth's sulfur cycle or microbial metabolic pathways, an earlier body of work (e.g., Schwarcz and Burnie, 1973; Goldhaber and Kaplan, 1975, 1980; Maynard, 1980) showed that $\delta^{34}\text{S}$ -isotopic variations in marine sediments are largely controlled by local physical factors, such as sedimentation rate, along with the supply of Fe and OM.' A reference to Bryant et al. was moved to the later in the text.

-L71: cite for closed system behaviour: Hartmann & Nielsen (1969 Geol. Rundschau, or 2012, Isot. Env. Health Stud.)

We have added the citation of (Hartmann and Nielsen, 2012) because this version is more accessible to anglophone readers.

-L73: This statement is questionable. Whereas AVS may reflect system behaviour, pyrite often reflects dominantly the conditions provided by essentially open system conditions

It is true that pyrite often reflects the compositions reflecting open conditions, especially in Fe-limited systems, but the large spread in $\delta^{34}\text{S}$ of sedimentary pyrites and gradients measured within pyrite grains (e.g. (Och and Shields-Zhou, 2012)) clearly shows that pyrite can also record sulfate reduction under closed system conditions.

-L78: Add Putschew et al. (OGC, 1996) and something from the 90s of the NIOZ group and more recently Amrani et al

This finding has been published in many papers and it is difficult to cite exhaustively but we have added two of the earlier articles of Putschew et al (1996) and Damste et al. (1998).

-L80: 'substantially' instead of 'significantly'?

Changed as suggested.

-L361: add Jorgensen et al. (2004, GCA)

This citation was not included because in this paper, unique conditions of extremely rapid sulfate reduction rates in an open system with iron trapping lead to the formation of very heavy CRS, which is the conditions in Cadagno where CRS becomes rather light compared to sulfate.

-L364: ...disproportionation of sulfur intermediates... (Canfield & Thamdrup, 1994; Cypionka et al., 1998 FEMS)...

Question: Is a repeated disproportionation cycling of sulfur intermediates really a plausible process in sediments? Canfield & Thamdrup (1994) proposed the model for an euxinic water column, the substantial role for benthic anaerobic sulfur cycling in competition to reductive processes is still to be shown

The discussion of disproportionation has been removed as it is quite unlikely, as mentioned in the original manuscript, requiring the removal of heavy sulfate from the system. We still mention the detection of bacteria belonging to clades of known disproportionators in line 351 and 389 out of general interest to the reader.

-L370: Which evidence of S^0 accumulation? What would be the role of S^0 reduction for isotope discrimination?

We have updated the figures to better highlight peaks in elemental sulfur at the surface and deep sediments. It is also mentioned that by reaction with elemental sulfur to form polysulfide (and rapid isotope exchange), the isotopic signature of sulfate reduction can be transferred to polysulfide reaction products (Amrani and Aizenshtat, 2004).

-L453: Is Fichtel et al. a suitable reference? The study belongs to a completely different type of system.

In the Conclusions we aim to demonstrate that inverse geochemical gradients and S-isotope overprinting can occur across a wide variety of environments, as Lake Cadagno is quite a unique lake. We have therefore changed the sentence to encompass a full range of terrestrial to deep subsurface environments as follows: In other environments, the reduction of sulfate diffusing from terrestrial aquifers (Porowski et al., 2019), submarine groundwater discharge sites (McAllister et al., 2015) and even deep crustal aquifers (Engelen et al., 2008), could essentially result in overprinting of new $\delta^{34}\text{S}$ signatures on buried sediments.

-L455. Why referring to the ocean crust as aquifer, instead of much shallower submarine or limnic groundwater discharge sites(?)

We have added references for sulfur cycling influenced by terrestrial and submarine aquifers (L. 765-766).

-L474: I do not understand this statement. Why should membership in an editorial board lead for an academic trained person lead to a competition of interests?

This statement was included upon specific request by the journal editor.

-L479: Didn't the co-authors also contributed to revision and editing of the ms? Who provided funding?

We have added that All co-authors contributed to revision and editing of this manuscript.

-Fig.2 Presentation should be improved: No combination of dissolved and solid phase species in one each plot; spread of the X axis for S isotope results

Plots in Figure 2 have been separated into surface sediments and deep sediments (as Figure 2 and Figure 3) for better scaling. Solid and dissolved species have also been separated. Supplementary data has now been incorporated into Figures 2 and 3. Ratios are presented as a separate Figure 4.