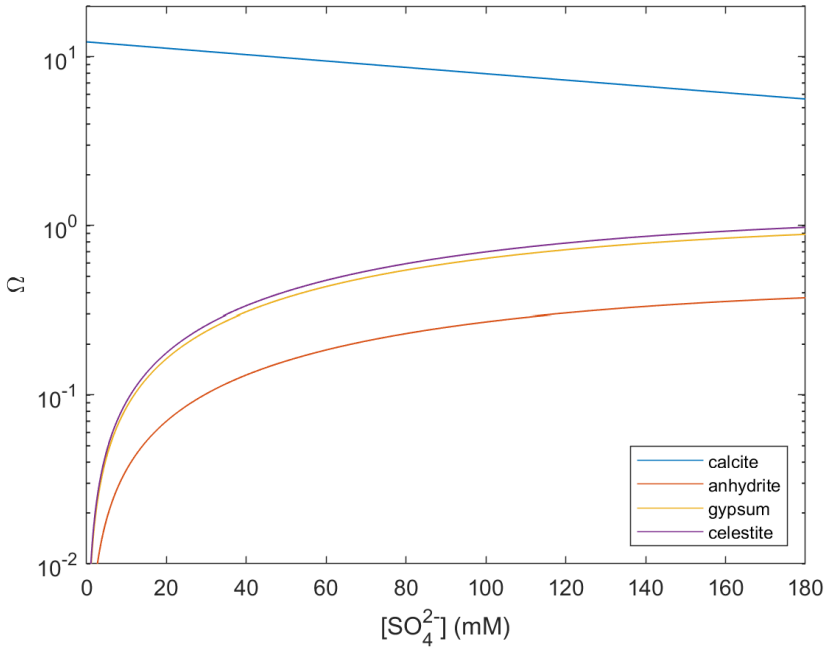
The manuscript 'Impact of seawater sulfate concentration on sulfur concentration and isotopic composition in calcite of two cultured benthic foraminifera' by Thaler *et al*. presents laboratory culture experiments of a strain of Rosalinid foraminifera grown under different seawater sulphate concentrations and sulphur isotopic compositions. The rationale is that foraminiferal carbonate associated sulphate (CAS), i.e., sulphate that is presumably incorporated as an impurity into the mineral lattice, may be an archive of past changes in the seawater sulphate concentration and isotopic composition. To this end, the authors have performed a set of carefully-conducted and well-described experiments covering a wide range in $[SO_4^{2-}]$ and $\delta^{34}S$. The results show that, up to a point, the foraminifer CAS concentration is proportional to that in seawater, and that $\delta^{34}S_{shell}$ is related to $\delta^{34}S_{seawater}$ with a slope of 1 but an offset of 1.4-1.6 ‰. Together, these results provide promising indication that foraminiferal CAS is likely to be a useful archive of past changes in the sulphur cycle, while highlighting the need for calibration datasets to identify species-specific vital effects.

I have a few main comments, related to the possible impact of seawater carbonate chemistry on some of the results and whether or not the data presented here really enable us to say something about the organisms' ability to regulate sulphate within the cell or at the biomineralisation site via active transport. While I suggest rephrasing parts of the discussion with this in mind, the manuscript is interesting and presents an important dataset, which I look forward to seeing published.

Main comments

1. As the authors acknowledge, previous work has identified the seawater SO_4^{2-}/CO_3^{2-} ratio as being a likely key control on S/Ca, as SO_4^{2-} probably competes for the anion position in calcite. Given this, experimental work aiming to understand S incorporation must have excellent carbonate chemistry control, which is largely the case here (pH and DIC were measured and were broadly held constant). It does not appear that changes/variations in seawater carbonate chemistry within and between experiments is a major issue for this study, but I would suggest that the authors calculate SO_4^{2-}/CO_3^{2-} for each experiment, and additionally (or alternatively) plot the results (Fig. 6) against this parameter. For example, the DIC in experiment ASW[60] was ~20% higher than in ASW[40]/ASW[50], which may explain the lower S/Ca ratio of this experiment.
2. In several places the manuscript contains inferences about biomineralisation which are very interesting but for which there is arguably little evidence based on the data presented here. For example, in Sec. 4.2, the possibility that foraminifera can regulate the biomineralisation site $[SO_4^{2-}]$ or actively maintain a constant SO_4^{2-}/CO_3^{2-} is mentioned. Both seem unlikely to me, although it is clearly stated that these are hypotheses, which is of course fine. However, these hypotheses appear in more certain terms elsewhere (e.g. the abstract 'highlighting the extent of control on the precipitation fluid chemistry…' (line 26) which should be removed or qualified, and lines 30, 44, 301 (even within the framework of hypothesising, I don't think 'probably indicates' is accurate), lines 370-375, 381). All of these sentences/sections should be rephrased more cautiously in my view.
3. Adaptation. The importance of acclimatisation and the benefit of working with benthic foraminifera is highlighted in a couple of places in the manuscript (e.g. line 68), but it was not quite clear to me whether only specimens that grew entirely under experimental conditions were selected for analysis, and to what extent reproduction can be considered adaptation. Was the original population/were empty shells following reproduction removed before geochemical characterisation? Please clarify. If not, then the point about acclimatisation to experimental conditions doesn't stand.
4. Complexation is mentioned in the discussion, and I agree that it would be helpful to calculate speciation in these experiments. Indeed, doing so (using phreeqc; see figure) highlights that i) the

experimental design means that $\Omega_{calcite}$ decreased with increasing [SO_4^{2-}], and ii) that at the extreme high end of the [SO_4^{2-}] studied here, the seawater was possibly saturated with respect to gypsum/celestite. Again, I doubt this impacts any of the main conclusions, but it would be good to include a more thorough discussion of the topic rather than talking in very general terms (lines 311-314). For example, perhaps the decreasing $\Omega_{calcite}$ contributed to the lower growth/survival rates of foraminifera grown at higher [SO_4^{2-}], given that $\Omega_{calcite}$ approximately halves between the lowest and highest [SO_4^{2-}] experiments.
Finally, given that some of the experimental seawaters used here are close to being oversaturated with respect to gypsum and celestite, was any inorganic precipitation observed in the cultures? Gypsum precipitation seems unlikely, but e.g. if [Sr_{sw}] was a little higher than the assumed/target value, then this may have been an issue in the higher [SO_4^{2-}] experiments.



[Phreeqc calculations using the solution compositions given in Sec. 2.1.2, pH = 8.2, DIC = 4 mM, and the pitzer database. The Matlab code used to produce the figure is given on page 4.]

5. There isn't a lot of foraminifera S/Ca data out there, so it would be a nice addition to the manuscript (and not too much work) to compare directly to previous studies, especially that of van Dijk et al. [2017].

Minor comments

1. Line 23. Consider clarifying in the abstract why this range is different than on line 21 (the reason is given later, but to avoid confusion).
2. Line 36. Clarify that this is the case at higher (room) temperature.
3. Line 124. I didn't understand – it is stated that the second set was 'designed to extend our concentration range', but the range is narrower than in the first set.
4. Line 149. Table 3 alternatively states ±4%.
5. Section 2.2. Were foraminifera from set 1 and set 2 combined for analysis, where they were grown under the same conditions?

6. Section 2.3. I think it's fine to do so briefly, but please give some basic details of how the instruments were set up, exactly how the analyses were run, how the data were processed, what the blank data looked like etc. etc., rather than simply referring to a previous publication.
7. Line 222. I'm not sure that this range (= ~50% of the modern ocean [DIC]) could really be considered fairly stable. I was also missing an explanation of why DIC was much higher in these experiments than natural seawater.
8. Section 3.3/figures. It might be helpful to report molar S/Ca ratios, to maintain consistency with the vast majority of the geochemical literature and previous work on foraminifera S.
9. Line 248. Please state what the uncertainty represents.
10. Lines 274-275. On the other hand, over the range that $[SO_4^{2-}]$ is thought to have varied within the Phanerozoic (~5-30 mM) there is no relationship between reproduction (growth?) and $[SO_4^{2-}]$.
11. Lines 286-288. Were the foraminifera crushed prior to cleaning? If not (possibly even if so) then inter/intra-crystalline organics likely remained.
12. Lines 294-296, lines 326-327. I found the comparison a little simplistic as of course pH/DIC/$[Mg^{2+}]$/organics are also very important.
13. Line 297, lines 322-324. I would also add a possible kinetic effect to the list. If crystal growth rates are lower at higher $[SO_4^{2-}]$, as the inorganic work indicates, all else being equal, then a nonlinear seawater-shell relationship might be expected.
14. Line 315. Please rephrase. Kadan *et al*. studied coccolithophores, which have a completely different biomineralisation pathway (e.g. centred on transmembrane ion transport rather than seawater vacuolisation).
15. Line 381. Does it become lethal? Or does it simply prevent calcification?
16. Figure 6. Seawater SO_4^2/Ca^{2+} is unitless, and the $/CaCO_3$ is unnecessary on the y axis.

Typos

1. Line 19. Calcifiers.
2. Line 321. van Dijk.
3. Line 578. de Nooijer.

```
clear variables

% requires a phreeqc installation
iphreeqc = actxserver('IPhreeqcCOM.Object');    % create PHREEQC COM object
% load desired database (pitzer for seawater)
dirP = uigetdir('C:\Program Files\USGS','Select directory containing PHREEQC databases');
iphreeqc.LoadDatabase([dirP '\pitzer.dat']);

% SO4, Na, and Cl co-vary in these experiments
SO4in = 0:0.1:180;
NaIn = 479:-(479-402)/(size(SO4in,2)-1):402;
ClIn = 612:-(612-175)/(size(SO4in,2)-1):175;

outputData = NaN(size(SO4in,2),6);
for i = 1:size(SO4in,2)
    IPCstringCell= {'SOLUTION 1',  ...
        ['-temp ', num2str(22)],  ...
        '-units mmol/L',  ...
        '-density 1.025',  ...
        ['-pH ', num2str(8.2)], ...
        ['Ca ', num2str(10.3)],  ...
        ['Mg ', num2str(53)],  ...
        ['B ', num2str(0.4)],  ...
        ['K ', num2str(10)],  ...
        ['Br ', num2str(0.8)],  ...
        ['S(6) ', num2str(SO4in(i))],  ...
        ['Na ', num2str(NaIn(i))], ...
        ['Cl ', num2str(ClIn(i))],  ...
        ['Si ', num2str(0)],  ...
        ['Sr ', num2str(0.1)],  ...
        ['P ', num2str(0)],  ...
        ['F ', num2str(0.1)],  ...
        ['C(4) ', num2str(4)],  ...
    'SELECTED_OUTPUT',  ...
        '-molalities  CO3-2 HCO3- CO2 MgHCO3+ NaHCO3 CaHCO3+ MgCO3 NaCO3- CaCO3', ...
        '-activities  CO3-2 HCO3- Ca+2 NaHCO3 NaCO3- SO4-2', ...
        '-SI calcite anhydrite gypsum celestite', ...
        'soln false',  ...
        'pH true',  ...
        'sim false',  ...
        'state false',  ...
        'time false',  ...
        'step false',  ...
        'pe false',  ...
        'distance false'};
    IPCstring = sprintf('%s\n', IPCstringCell{:});

    iphreeqc.RunString( IPCstring );
    OUTphreeqSTRING = iphreeqc.GetSelectedOutputArray;

    % retrieve the data
    loc = find(strcmp(OUTphreeqSTRING,'m_CO3-2(mol/kgw)'));
    outputData(i,1) = OUTphreeqSTRING{2,(loc+1)/2};
    loc = find(strcmp(OUTphreeqSTRING,'si_calcite'));
    outputData(i,2) = OUTphreeqSTRING{2,(loc+1)/2};
    loc = find(strcmp(OUTphreeqSTRING,'si_anhydrite'));
    outputData(i,3) = OUTphreeqSTRING{2,(loc+1)/2};
    loc = find(strcmp(OUTphreeqSTRING,'si_gypsum'));
    outputData(i,4) = OUTphreeqSTRING{2,(loc+1)/2};
    loc = find(strcmp(OUTphreeqSTRING,'la_SO4-2'));
    outputData(i,5) = OUTphreeqSTRING{2,(loc+1)/2};
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    loc = find(strcmp(OUTphreeqSTRING,'si_celestite'));
    outputData(i,6) = OUTphreeqSTRING{2,(loc+1)/2};

end

% plot Omega calcite, anhydrite, gypsum, celestite
close(figure(1))
figure(1)
plot(SO4in,10.^(outputData(:,2)))
hold on
plot(SO4in,10.^(outputData(:,3)))
plot(SO4in,10.^(outputData(:,4)))
plot(SO4in,10.^(outputData(:,6)))
set(gcf,'color','w')
xlabel('[SO_4^{2-}] (mM)')
ylabel('\Omega')
legend('calcite','anhydrite','gypsum','celestite',...
    'location','southeast','fontsize',8)
set(gca,'yscale','log')
ylim([1e-2 20])
```