In this work, the authors introduced, in the software MAGEMin (Riel et al., 2022), a more efficient formulation to compute the Gibbs free energy minimization based on the thermodynamic database of Holland et al., (2018). The previous formulation implemented in MAGEMin solved the minimization problem by imposing inequality constraints to bound the fraction of elements in a given crystallographic site between 0 and 1. This formulation is computationally expensive, because it has to be repeated for all the mixing elements in each crystallographic site of the solid solution, thus taking 75-90 % of the total computation time. Moreover, the minimization can occasionally fail due to violations of the inequality constraints, leading to a poor success rate of 57-97 %. The low performances of the inequality constraints formulation, hinders the coupling of phase equilibria calculation with geodynamic models (e.g., self-consistent reactive fluid transport models). The new formulation introduced in MAGEMin by the authors employs the nullspace approach ($Ax = 0$) to transform the non-linear inequality constraints into a set of linear equalities, which are subsequently turned into a bound-constrained optimization problem: min $f(x)$, with $x > 0$. This formulation can be solved using gradient-based algorithms, which are computationally efficient. While the nullspace approach is not novel, the authors have further compared the performances of four Gibbs free energy minimization algorithms: two with inequality constraints (CCSAQ and SLSQP), and two with bound-constrained optimization methods (CG and BFGS). Here the authors show that the bound-constrained formulation combined with the BFGS algorithm yields the best performance by significantly reducing the minimization time regardless of the dimensionality of the phase model (i.e. the number of oxides used to define the solid solution), while also maintaining a 100% success rate.
This work remarks the efficiency of the bound-constrained optimization method in computing Gibbs free energy minimization compared to inequality constraints formulations. The algorithm described in this paper represents a valuable computational tool, which can be implemented in other phase equilibria calculators. I believe that the bound-constrained formulation is relevant for the broad geodynamic community, as it will allow more efficient coupling between petrological and thermomechanical models. The quality of the manuscript is good, but some minor revisions are needed before its publication.

We thank the reviewer for the very careful read and helpful correction/suggestions that largely improved the quality of the manuscript.

Line 19: The acronym ‘BFGS’ in the abstract should be explained before its first use.

Corrected

Line 21: In the conclusions (line 330) you report that the new approach improves the computational time by a factor of ≥ 3; here in the abstract, instead, you report a factor of ≥ 5. Which one is the correct factor?

Thank you for spotting this discrepancy. We corrected the conclusion to ≥ 5.

Lines 28-30: there is an extra bracket between 'MELTS and Connolly'.
We added a bracket to close the list of code before the citations.

Line 37: the notation 10s 100s is misleading, as it can be read as 10 or 100 seconds. It is better to use the order of magnitude, i.e. 101, 102 milliseconds.

We corrected to "the order of 10 to 100 ms"

Line 39: same as before line 37; it is better to use the order of magnitude.

We corrected to Constrained Optimization by Quadratic Approximations (CCSAQ) "from thousands to hundreds of thousands"

Line 59: The acronym 'CCSAQ' should be explained before its first use.

We clarified the acronym: "Constrained Optimization by Quadratic Approximations (CCSAQ)"

Line 62: GeoPS starts with a capital G. Moreover, it is better to start the sentence introducing what GeoPS is, e.g., "The phase equilibria calculator GeoPS…".

Corrected accordingly

Line 76: does *xeos* mean 'compositional equation of state'? The definition of this term is not intuitive, and it should be explicitly defined in the text.

This was corrected as "we present a revised implementation of the compositional and order variables (xeos)" during previous revision. It seems the reviewer did not have accessed to the reviewed manuscript.

Line 83: here the symbol and the unit of Gibbs energy is missing, i.e. $G\lambda$ and [J].

We thank the reviewer for spotting this omission. We corrected it accordingly.

Line 85: does $pi(\lambda)$ represents the mole fraction [mol]? Otherwise, the unit of measurements of Gibbs energy [J] in equation (1) is not consistent.

We added the unit [mol]

Line 86: the unit of measurements of molar chemical potential $\mu i(\lambda)$ [J mol-1] is missing.

Corrected

Line 90: "the *molar* chemical potential of a phase is…" (here molar is missing).

Corrected

Line 94-95: the unit of measurements of the ideal activity $aid i(\lambda)$, the reference Gibbs energy $g0i(\lambda)$ and the excess energy $gex i(\lambda)$ are missing. It should explicitly mention that $aid i(\lambda)$ is dimensionless [/]. To satisfy the dimensional consistency of the equation (2), the two energies $g0i(\lambda)$ and $gex i(\lambda)$ should be [J mol-1]. If this is the case, they should be named: reference molar Gibbs energy and excess molar energy.

We agree and corrected all points accordingly

Equation 4, line 101: why is $N\lambda$ now referred to olivine (*Nol*)? This has not been established in the text. *Nol* is also presented in the summation in line 102. The parameters $\phi'n$ and ϕn are not described, whereas ϕi does not appear in the equation.

We corrected Nol to be N λ to be more generic. Moreover, we clarified equation 4 by correcting th $[\mathrm{J \cdot mol^{-1}}]$ e definition of the terms.

Line 102: can you explain what the asymmetry parameter *vi* is?
{m,n}
We added that the asymmetry parameter is the van Laar parameter.

Line 104: the unit of measurements of interaction energy *Wm,n* is missing. It should be [J mol-1].

We added the unit

Equation 5, line 108: there is too much distance between the introduction of the variable *xcv* (line 114) and its usage in equation 5 (line 108). What does the subscript _cv stand for? composition (c) and order (v)? This parameter should be introduced before equation 5, e.g.: "Given this formulation, the set of equations 1 to 4 can be directly transformed into the following Gibbs free energy minimization problem as a function of the compositional (c) and order (v) variables *xcv*".

We corrected the introduction of the variable x_cv as suggested by the reviewer:
"… the following Gibbs free energy minimization problem as function of the compositional and order variable x_cv"

Line 109: here you should explicitly introduce the first inequality constrain, i.e. the fraction of the element X ≥ 0.

We modified to: “subject to the site fraction of the element”

Line 111: here you should explicitly introduce the second inequality constrain, i.e. the compositional and order variables must be within an upper (ubcv) and a lower (lbcv) limit.
We corrected this part of the text accordingly

Paragraph 118-126: Here the authors should make clear to the reader that they are implementing this new formulation in the code MAGEMin based on the thermodynamic database of Holland et al., (2018). Moreover, the authors should report some literature to remark that the introduction of the nullspace approach is not a novel and it has already been used to computed the Gibbs free energy minimization in other phase equilibria calculators (e.g., HeFESTo Stixrude and Lithgow-Bertelloni, 2011 - https://doi.org/10.1111/j.1365-246X.2010.04890.x).

We agree and added the reference to HeFESTo.

Line 120: It is better to avoid having nested brackets e.g., "... crystallographic sites i.e., Bragg–Williams-type formulation (Myhill and Connolly, 2021)".

We removed the nested bracket.

Lines 122-123: too many consecutive adverbs (numerically, significantly, costly). Try with: "which has a significantly higher numerical cost compared to the bound-constrained minimization algorithms".

We thank the reviewer for the suggestion and changed the text accordingly

Line 131: the sentence in the bracket can be moved after the equations e.g., "Note that we have dropped the ion charges in the notation of the equations".

Corrected

Equation 17, line 155: It would be clearer to the reader to define x = Xes already here.

We clarify this part by now stating that x and c are the compositional and order variable of olivine as defined in Holland et al., (2018).

Equation 24, line 184: Is the matrix *Nz* related to the number of endmembers *Nλ*? If yes, could you state it explicitly in the text? If not, wouldn’t be better to use a different letter?

Nz is the matrix that span the nullspace of A and its size is function of the number of site fractions and number of equality constraints

Line 222: the term '*unconstrained*' should be substituted with '*bound-constrained*'.

Corrected

Line 227: as stated above by the authors (line 220), the case *dk* = -*gk* has not been explored in this study. Perhaps it is better to remarks this to the reader also in line 227 to avoid confusion. E.g., "… if the iteration increment k = 0 (not investigated here), or..".

The conjugate gradient method is initialized for the first iteration as a steepest gradient method and then updated in subsequent iterations of the algorithm. We clarify the sentences to avoid misunderstandings.

Line 243: the term '*unconstrained*' should be substituted with '*bound-constrained*'.

Corrected

Line 252: the equation number should in within brackets, e.g., equation (42).

Corrected

Line 262: the compositions (NCKFMASHTO, KNCFMASTOCr, and FMATOCr) should be explained here, since they refer to the oxides components and it might not be intuitive: N = Na2O; C = CaO; etc. Moreover, there should be an explicit reference to Table 1.

We added the reference to table 1 and now present the chemical system with oxide list.

Line 271: the term '*unconstrained*' should be substituted with '*bound-constrained*'.

Corrected

Line 273: The acronyms 'SLSQP' and 'CCSAQ' should be explained before their first use.

The acronyms are now explained.

Table 1: Temperature should be expressed in [K] not [°C] to be consistent with eq. 2. However, if the authors prefer to keep the units of measurements typically used in metamorphic petrology (kbar and °C) they should state it in the caption of Table 1.

We corrected the temperature unit to K

Figure 2: It would be useful to report the dimensionality below the mineral e.g., clino-amphibole (dimensionality 10) or (10 oxides composition).

Corrected

Line 281: the term '*unconstrained*' should be substituted with '*bound-constrained*'.

Corrected

Figure 3: It would be useful to report the dimensionality also in this Figure.

Corrected

Lines 292, 293, 300: the terms '*unconstrained*' should be substituted with '*bound-constrained*'.

Corrected

Figure 4. How do you compute the local minimum of the solvus (yellow dots)? Have you obtained them with THERMOCALC using the Holland et al. (2018) database? The reference of these data points must be added.

The spinel solvus has been computed using the Gibbs hyperplane provided in table 1 which was computed using MAGEMin. We added this clarification to the figure 4 caption.

Line 304: when referring to figures and tables you should be consistent throughout the manuscript: either Fig. or figure; Figure or figure; Table or table, etc...

Corrected to "Figure"

Line 313: Figure 5A to 5C.

We corrected the figure call to Figure 5A-C

Line 315: 5A, 5B, 5C.

We corrected the figure call to Figure 5A-C

Line 316: 5D, 5E, 5F.

We corrected the figure call to Figure 5D-F

Figure 5: It would be useful to report the dimensionality also in this Figure.

Added

Lines 319, 325, 328: the terms '*unconstrained*' should be substituted with '*bound-constrained*'

Corrected

Line 330: could you elaborate the ≥ 3 factor improvement? Why is it different from the one reported in the abstract?
The first ≥ 3 factor improvement estimate was achieved without considering the minimization failure rate of the CCSAQ and SLSQP methods. When considering it, we find that a ≥ 5 factor improvement to be a better estimate. We corrected this in the text.

Competing Interests: This statement should be more explicit e.g., 'BK (co-author) is a member of the editorial board of GMD'.

Corrected