Referee #2

General comments:

1. Although the interest of the program is undeniable, the article is not sufficiently clear to explain the aims of the program. In addition, the comparison of the program with those are currently used is not developed and the interest of the development proposed by PyXRD v0.6.2 are not enough supported by the discussion.
   We believe the currently reworked manuscript addresses these issues by adding a more thorough comparison and by clarifying the tested hypothesis.
2. Moreover, the “test case simulations” exposed are not enough discussed in details to be considered relevant for a test of sensitivity.
   We never stated these test case simulations are a test of sensitivity. We do mention parameter sensitivity when trying to explain why the used refinement strategy did not converge for a few parameters.
3. First part of the introduction presents a short paragraph about clay minerals. This part is not enough detailed and too restricted to clay minerals by contrast to the title in which the used of the program is generalized to “layered minerals”. If PyXRD v0.6.2 can provide XRD calculation of 00l reflections from different types of layered minerals, the authors should enlarge this first part to other types of layered structures such as layered double hydroxides or layered carbon. If the program provides XRD calculation for only clay minerals, the authors should change the title.
   Although PyXRD can, in theory, produce X-ray diffraction patterns for any kind of layered material, it is geared towards using it for clay minerals. Therefore, we have changed the title to indicate this.
4. The second paragraph deals with the previous programs that were developed to performed XRD calculation in order to identify and quantify clay minerals. This part seems not enough detailed and the differences between XRD calculations from powders and from oriented preparations is not clear. These approaches are complementary but the authors do not explain their own interest. Development of XRD calculation for powders has been used in first for the identification of crystal structure and then has been applied to quantify the contribution of each mineral in a mixture or a sample. Nevertheless, even powerful, this approach was not able to refine structure for layered mineral when mixed layering structures are present. For this reason, XRD calculation of the 00l reflections of layered minerals has been developed in the aim to first determine the structural defect due to mixed layering and second, quantify the contribution of such MLMs in a sample. Thus, the development of profile fitting of 00l reflections from oriented sample is mainly due to the difficulty to identify and quantify MLMs from powder samples and remains the main interest of this type of XRD calculation. This is no mentioned in this paragraph and a background part could be dedicated to this topic in the paper in order to highlight the interest of the program proposed. The authors should split the two types of calculation approaches (powder and oriented sample) after their presentation and focus on the programs that were developed for the calculation of 00l reflections. There are several other programs than those given in the text (NEWMOD, MLM3C, Sybilla) and the authors should give a complete overview of them, they have been developed since the 70’s. Particular attention should be paid to their ability to calculate XRD patterns from complex MLMs with more than 2 types of constitutive layers. Note that the authors present some drawbacks about XRD calculation of oriented samples that are not improved in the manuscript because they cannot. This has a limited interest for the reader.
   We have clarified this paragraph by elaborating on the differences between powder and oriented samples, and by adding some more background as to why modelling of oriented samples is important for mixed-layer minerals. We disagree that we should add a complete overview of all the models that have been developed since the '70, since we are not writing a

review paper. We believe we have included the most commonly used, well known and recent models in our introduction and this should be sufficient. We agree on the reviewers point that the mentioned drawbacks and advantages are perhaps not very relevant and have removed them from this paragraph.

5. The last paragraph is about the ability of the programs to automatically refine parameters for XRD calculation. As for the second paragraph, this paragraph should develop with more details the differences between the automatic approaches used because the automatic procedure is the main input of the program proposed.
   We would again like to point out we are not writing a review paper, and do not feel it is appropriate to give a very detailed description of all the different methods for automatic parameter refinement. We have added some more detail to this paragraph which we believe should help the reader in understanding the significance of the choice without going too far off-topic.
6. About the multi-specimen approach, the authors should explain deeper this procedure because this procedure is traditional in the study of clay minerals for their qualitative identification, and was added latter as a constraint for their XRD calculation.
   Changes were made accordingly.
7. Materials and methods: The main problem of this part is the lack of explanation about the calculation of XRD patterns for layered structures. The authors present the different components of their program but there is no explanation and no calculation that could validated the part that concerns the calculation of the layered structures themselves. This is fundamental because before work on mixture of layered minerals, the authors must prove that their program well reproduced experimental XRD patterns of layered structures from which, the crystal structure as well as the chemistry is know. The author should develop a large part about the parameters computed and give some example that validate the XRD calculation for know layered structures, discrete and MLMs (this could be the first section of the results part). In addition, the authors should compare their resulting calculations with XRD patterns calculate and validate from other software that used a similar approach (Sybilla, MLM3C. . .). In such case, they should compare the direct XRD calculation from simple to complex layered structures, such as discrete to 3 components MLMs with complex stacking order (R2).
   The lack of explanation about the calculations is not relevant, since these are included in the manual of the program, as also mentioned in the manuscript. We have made the suggested comparison. We compared our model with Sybilla, since it was readily available to us and MLM3C is not. Also see the remarks below.
8. Test case simulations The cases tested do not appear relevant except for the automatic refinement. Indeed, the authors test solely their automatic refinement procedure by using theoretical mineral assemblage from their own program. The first test proposes in the comment about “Materials and methods” part seems to be a first step before the case proposed. There are several possibilities to evaluate the program and the choice is too restricted and do not allow to judge the validity and the limits of the automatic refinement procedure. I would suggest to the authors an example of gradual test for the result part:
   - First section: validity of the XRD calculation. Comparison with the results obtained from other programs with a gradual increase of the complexity of the layered structures (discrete clay without swelling layers in air dried state; discrete swelling layers under various treatments such as air dried and after glycolation; R0 two component MLMs without swelling layers; R0 two component with swelling layers; R0 three component MLMs without swelling layers; R0 two component with swelling layers; R1 and R2 three component MLMs with or without swelling layers to finally validate the calculation of parameters for the stacking order).
   - Second section: Validity of the quantitative results from mixture of layered structures: Based on the same idea (comparison with Sybilla results for example), the contribution

of discrete and complex MLMs could be compared on a set of examples which have been validated in the first section. A gradual complexity of the mixture that can reach the complexity of **natural** soil samples could be proposed (from mixture with two clay minerals to mixture with eight or ten clay minerals with 3 or 4 types of R0 three component MLMs). One example with mixture that mimics the clay paragenesis of diagenitic rocks could be also interesting for the geologist. Indeed, such types of samples were, and are, the most studied and such example could allow enlarging the audience. One or two samples from the Golf coast series seem to be good examples (with R0 and R2 MLMs, see the XRD calculations performed in Lanson, B. et al, 2009. Diagenetic smectite-to-illite transition in clay-rich sediments: a reappraisal of X-ray diffraction results using the multi-specimen method. Am. J. Sci. 309, 476–516.).

- Third section: assessment of the automatic refinement based on multi-specimen approach. Based on the same examples than those proposed in the second section, the automatic refinement could be assessed. The main interest of the automatic refinement is for complex mixtures because for less complex mixtures with two or three clay phases (even with R0 2 components MLMs like assemblage 1 and 4 presented in the article), a reliable structure can be find very fast (about one hour) with the manual trial and error approach. Thus, two complex structures, one that mimic <2 µm fraction of soil sample (high number of discrete and R0 3 components MLMs) and one that mimic diagenetic sample (lower number of clay minerals but with R2 MLMs) could be interesting. In such cases, the interest of the multi specimen approach should be more evident because the presence of 3 component MLMs with swelling layers (that can have heterogeneous hydration or swelling behaviours) need to be constraint by using various treatments. One additional interesting example could be the original XRD calculation from which the multi specimen approach was developed for XRD profile modelling (Sakharov, B.A. et al, 1999. Determination of illite–smectite structures using multispecimen X-ray diffraction profile fitting. Clay Clay Miner. 47, 555–566).
- Results and discussion: The “Test case simulations” part could be removed and replaced by a results and discussion part. The different sections proposed below could be discussed in order to demonstrate that the program proposed is relevant for i) XRD calculation of complex layered structures (first section), ii) relevant for the quantification of complex layered structures in mixture (second section), iii) the automatic refinement proposed based on the multi-specimen approach is relevant to rapidly obtain coherent structural models (third section).

We have tried to address most of the suggestions made by the reviewer. More specifically, we have added a direct comparison of PyXRD and Sybilla output for 13 discrete phases (first section) and for 5 mixtures (second section). We have kept the original data regarding the assessment of the automatic parameter refinement with and without the multi-specimen approach. We have not added the examples as suggested by the reviewer, due to time constraints. Nonetheless we believe the currently included comparison is sufficient to illustrate the validity of PyXRD's output.