Authors' response

The authors would again like to thank both referees for their time and effort in reviewing our manuscript. Although minor, both referees made good and helpful points. Below is an overview of the comments and remarks made by the referees followed by our response and actions in italic.

Comments from referees

Referee #1 (Report #2)

Minor remarks:

1 l. 52: “Programs” rather than “Models”. Double check throughout the Ms as this was said (in the rebuttal letter) to be changed.
Changes made accordingly and double-checked to make sure model and program is used unambiguously.

2 l. 237-251: It could be said somewhere how the user (or the program) selects one algorithm or the other and what could be the criteria for this choice.
A short explanation has been added. More information can also be found in the manual, in the section on the guided example.

3 l. 291-293: “planes” rather than “layers” to comply with AIPEA nomenclature recommendations. Check throughout the Ms.
Changes made accordingly.

Referee #3 (Report #1)

General comments:

1. The authors have created “calculated” mixtures with PyXRD and modelled them using Sybilla. To validate the fitting obtained with PyXRD, the authors give the results of QPA analyses of the mixtures and differences in proportions are below 2%. However, the authors do not explain clearly i) if such differences in QPA are significant and acceptable, and ii) to which mineral it is attributed. It should be useful to provide a figure with the ‘input’ pattern, the different contribution of individual species and the calculated patterns and better evidence the differences between the two softwares. For detail of the calculation procedure please refer to the reports of the other reviewers.
   The referee raises a good point. We have added an additional table with Rp and Rwp factors for the (more ore less) separated first order reflections of phases in mixtures 1-3 to illustrate that the observed misfits can actually not be attributed to a single phase, or, with other words, each phase contributes more or less equally towards the total misfit. I would also like to stress that these are minor misfits (when considering the patterns). Because of this, we have not provided an additional figure with comparisons of the contribution of each separate phase, as it will not help in clarifying the difference in obtained wt%. We think these differences are mostly due to differences in unit cell size. Sadly the unit cell dimensions in the a and b direction used in Sybilla are not known. We have adjusted the manuscript to clarify this.
2. The use of the multispecimen approach is somehow confusing in some part of the Ms. Multispecimen approach was developed to constrain the calculation for mixed-layers containing swelling to find accurate proportion of each layer types in the structure (structure, stacking order, probabilities) and to avoid misidentification based on one single treatment. Structural parameters and proportion of each species should be identical between the different treatments (or with acceptable variations). Multispecimen is not used to obtain the parameters that are input data for the calculation.
   We believe the relevant sections of the manuscript have been adequately re-written in the latest revision to clarify this. We have double-checked and made some minor modifications.
3. For the ‘calculated’ mixtures used the response to the different treatments (in fact calculation in this case) is easy to obtain as the structures (even complex) are ideal and made of the stacking of identical layers with the junction probabilities chosen. However in the ‘real-life’ the multispecimen is more complex (heterogeneity of the sample at different scales) and some parameters are obtained by the time consuming but necessary trial and error modelling procedure. This step is at least necessary in a first step if a series of identical sample is treated.
   We are fully aware of this fact, and are also using the trial-and-error approach for other research topics. However, our idea for this paper was to get an objective or at least unambiguous comparison possible, for which calculated mixtures are the better choice.
4. In the discussion and the conclusion, the authors argue that a good identification is a pre-requisite to obtain a good QPA. Of course this is true and MUST be verified, and multi-specimen is used for this. Softwares such as Sybilla or PyXRD offer the opportunity to do that and QPA is obtained in another step. It is somehow dangerous to think that PyXRD, or Sybilla, or other softwares could avoid any accurate identification before quantification. The authors that are aware of this should write it more clearly in the Ms.
   Changes made accordingly.
5. The authors indicate that the modelling gave acceptable results for the ‘calculated’ model but should be more difficult for natural geologic samples. One of the advantages of PyXRD should be to use the possibility to share species parameters across different particle fraction

size to model the bulk < 2 µm fraction sample from the models obtained for the different (infra-micrometric) particle-size fractions and their mass % as done by Hubert et al. 2012 and the authors in Geoderma 2014.
It is indeed possible to share parameters in this way. We have also been playing with the idea to include a 'loosened' linking of parameters, where one parameter's 'valid' range is defined using the value of another parameter. However this is just an idea for the moment.

Minor remarks:

→ We noticed this referee's line number references did not match with revision #3, but did match with revision #2, some of his minor remarks have been removed since they were no longer relevant.

1. All the Ms.: Glycol should be replaced by ethylene glycol.
   Changes made accordingly.
2. Pg 5, line 63: The objective is not currently to have automated QPA but to use modeling to have better accuracy in identification and secondly to semi-quantify. Please remove automated or rewrite.
   Changes made accordingly.
3. Materials and methods: The authors use PyXRD on HPC clusters. Is it possible to use the software on PC or the calculation time will be too long.
   Clarified in the manuscript – the refinements don't take that long at all, but running them on an HPC allows to run refinements several times without hindering the performance of the user's PC. Also we ran 50 iterations of each set-up to eliminate the stochastic nature of the chosen refinement strategy.
4. Page, 16, lines 326-327. Hydroxy-interlayered smectite or hydroxy-interlayered vermiculite are not always poorly crystallized. They may be present in coarse clay fractions (> 0.2 µm). I agree that they have to be differentiated from primary (trioctahedral) chlorite.
   We don't exclude the occurrence of well-crystallized HIS or HIV, in this case we just included what could pass as a poorly-crystalline one.
5. References: The authors should cite the works of:Sakharov, B. A. and Drits, V. A. (1973) Mixed-layer kaolinte-montmorillonite: a comparison observed and calculated diffraction patterns. Clays and Clay Minerals, 21, 15-17. One of the first works on modeling X-ray diffraction pattern of clay minerals. Viennet, J.C. Hubert, F., Ferrage, E., Tertre, E., Legout, A. and Turpault, M.P. 2015 Investigation of clay mineralogy in a temperate acidic soil of a forest using X-ray diffraction profile modeling: Beyond the HIS and HIV description. Geoderma 241–242, 75–86. One of the most recent works on the subject.
   We have included these references where deemed appropriate.