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PyXRD v0.6.5: a free and open source program to quantify disordered, phyllosilicates using multi-specimen X-ray diffraction profile fitting.
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General:
The manuscript presents new software, PyXRD, dedicated to the identification and semi-quantification of clay minerals assemblage using multi-specimen profile fitting. The Ms is written in good English. The authors have focused their work on the comparison of the fittings obtained using Sybilla © Chevron, one of the most robust and used program, to calculate 00l peak series of clay minerals, and the fittings obtained using PyXRD. In general, PyXRD provides acceptable fitting compared to Sybilla ones with however some discrepancies.
The authors have created "calculated" mixtures with PyXRD and modeled 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 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. 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.

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.

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.

In summary, PyXRD appear to be a good program to the accurately identify (structure determination) the clay minerals encountered in complex mixtures, such as geologic samples, and provide semi-quantification. This open source program is a new step forward that could be used and improved by clay scientists.
For these reasons, I strongly recommend the authors to revise their manuscript to be in order for publication with moderate revisions.

The following lists some specific remarks:

Title
It should be useful to specify that PyXRD is a program to **identify and semi-quantify** disordered phyllosilicates.
Remove the comma after disordered
All the Ms.
Glycol should be replaced by ethylene glycol.
Introduction
Page 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.
Page 6, lines 104 and 109: The multispecimen approach does not require to record multiple specimen but to record patterns of the same specimen after different treatments (e.g. saturation with different interlayer cations, heating, ethylene glycol solvation, …). It is used to identify mixed-layer structures containing expandable layers. The authors should clarify this paragraph.
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.
Results
Page 14, line 292: replace glycol by ethylene glycol and insert a space between glycol and molecules.
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.
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. 2015Investigation 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.