Données complémentaires sur l'evolution de la conductivité des effluents de colonnes de sol non remanié durant le drainage

Ces données sont rattachées au chapitre IV et permettent de constater l'effet de l'infiltration de l'eau de pluie - de faible conductivité ionique (environ 17 µS cm^{-1}) - sur la conductivité des effluents récupérés durant le drainage (colonnes 1, 2 et 3). Un effet de dilution est constaté durant toute la durée de la pluie, puis la conductivité remonte rapidement après son arrêt pour retrouver des valeurs proches de la conductivité initiale.

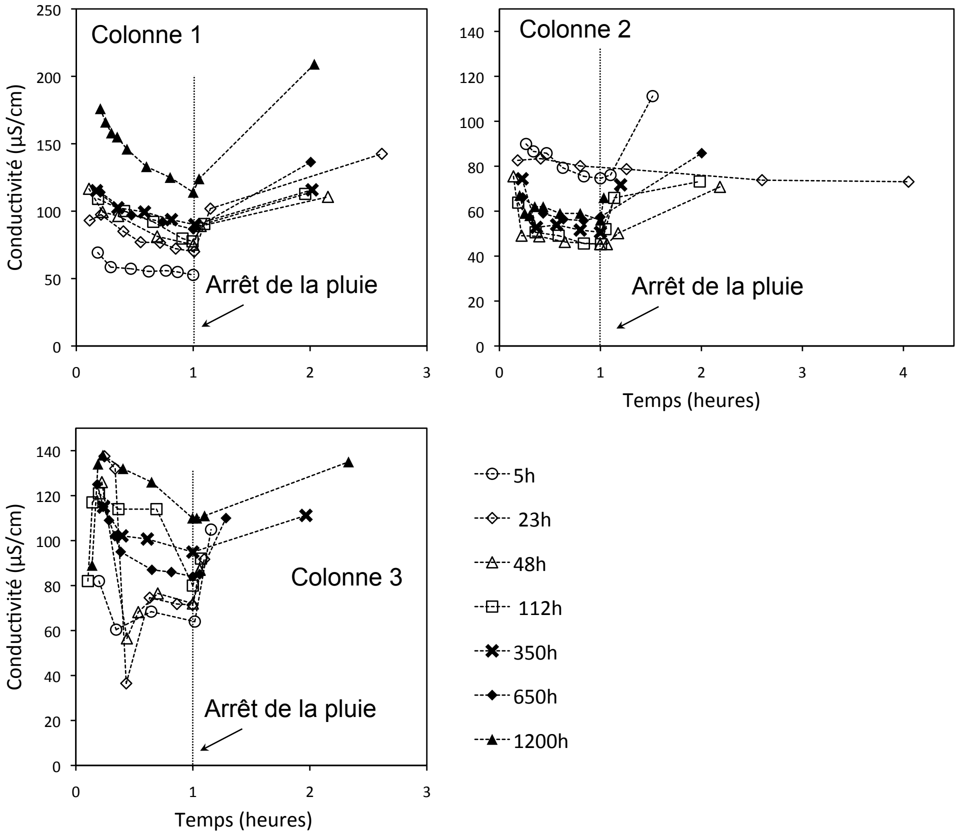

Figure XI -1 Evolution de la conductivité des effluents en fonction du temps durant les pluies. Les effluents sont récupérés en sortie de colonne de luvisol non remanié. Les propriétés de ce sol sont décrites Tableau II-1 et dans le chapitre IV traitant de la rétention de microsphères.

XI.2. Annexe 2 : Localization of particle deposition in soils: process identification towards modelling

Cette annexe présente une activité complémentaire de recherche, menée durant cette thèse, dont les résultats, complexes à interpréter, n'ont pas pu être inclus dans le présent manuscrit et durant le temps imparti. Il s'est agit de localiser par micro-diffraction des rayons X sur source synchrotron des revêtements supposés s'être formés durant des expérimentations de pluies sur colonnes de sol non remanié (Quénard, 2011) et supposés enrichis en smectite.

XI.2.1. Réponse à un appel à projet au synchrotron SOLEIL

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XI.2.1.1. *Background*

Lessivage is among the most widespread processes in soils and it has been described in many soils types (WRB, 2006). This process is defined as a substantial vertical transfer of fine particles (with size ranging from less than 2 µm to less than 10 µm according to different authors) from a horizon, called eluviated horizon to another horizon referred to illuviated horizon (Legros, 2007). Physical and chemico-physical mechanisms involved in this process are not yet fully understood and lessivage fluxes are still not quantified due to obvious technical difficulties.

The Agriped project started end of 2010 has been funded by the ANR to identify the mechanisms responsible for lessivage, quantify and model it. Within the frame of this project a complex experimental setup was build up in order to simulate lessivage in lab (i.e. simulation on a sequence of rainfall events) and to identify the processes and factors responsible for it.

The lab experiment setup consists in a sequence of 30 rainfalls on undisturbed and unsaturated soil columns of decimeter size. In order to simulate separately eluviation and illuviation two experiments were run. As smectite were described as especially sensitive to eluviation (Mercier et al., 2000), the first experiment designed to simulate eluviation consisted in columns made of an undisturbed soil monolith containing smectite. The second experiment, designed to simulate illuviation, consisted in columns made of two overlaid soil monoliths, the upper one being that used for the eluviation experiment and lower-one containing no smectite.

These experiments demonstrated that particles migrate from the upper horizon and were partially (25 to 90 %) fixed in the lower horizon. About 50 g m^2 particles were fixated within the 15 cm depth of the lower soil monoliths. This fixation was shown to be due to physico-chemical processes during the first 17 rains and to column clogging on the rest of the experiment (Cornu et al., 2014). However the origin and the location within the lower soil monolith of the clogging could not be identified. Clogging could be either due to the collapse of the soil structure within the column or to the closure of pores by illuviation (particle fixation). To answer this question the lower soil monolith have to be open and analysed. Therefore thin sections were made on these monoliths for different amounts of rain and different rainfall intensities. On these sections, clay coatings were identified along pores and their geometrical relationship to the porosity described. However, the coatings may have pre-existed the experiment. Differentiating old from new coating by optical microscopy is feasible but only in very well developed clay coatings, which is probably not our case (Akamigbo and Dalrymple, 1985). Mapping the mineralogical composition of the clay coating would allow distinguishing with certainty the new coatings thanks to the presence of smectite. The clay coatings in soils are generally of 60 to 200 µm thick (Dalrymple and Theocharopoulos, 1987; Thompson et al., 1990), but they are developed on longer time period. Therefore the coatings build up over the experiment are more probably 20 to 30 µm thick. For this reason, analysing them with a lab-DRX is not possible as the beam obtained on this type of apparatus is about 150 µm in size.

XI.2.1.2. *Objectives*

The aim of this project is thus to localize with respect to the soil porosity clay coatings containing smectite and thus considered as having been formed over the duration of the lab-experiment. The mechanisms responsible for this clay fixation with respect to the soil porosity will thus be interpreted thanks the mapping of the mineralogical composition of the coatings and even quantified. This mapping is only feasible with a synchrotron light as the expected smectite concentration in the coatings are low and as the beam size of lab-apparatus is too large to allow the mapping of the coatings.

XI.2.1.3. *Experimental method*

Two rain intensities were used over the experiment for two different sets of columns (20 mm h^{-1} and 6 mm h^{-1}). For these two rainfall intensities, columns were stopped after 20, 25 and 30 rains. Twenty-four columns were thus run over the illuviation experiment. Among these columns, 6 of them were selected in order to have a kinetic gradient (20, 25 and 30 rains) and to compare the impact of the 2 rainfall intensities after 30 rains.

On the lower soil monolith of these 6 columns, undisturbed centimeter blocs were sampled at the top and at the base of the monolith for thin sections. The blocs were air-dried for 2 months, then oven-dried at 40 °C for 2 weeks, and impregnated under a vacuum of -5 kPa with a polyester resin diluted to 30 % by volume with a styrene monomer (Bruand et al., 1996). Undisturbed soil thin sections, 50 x 50 mm in size and 25 µm thick, were prepared on glass support. Twelve thin sections were observed by using an optical microscope and six representative thin sections were selected. This selection is based on morphological and optical characteristics of the clay deposit contrasting with the soil matrix (Sauzet, 2012). Then for each of the 6 thin sections, 4 zones of 500 µm^2, representative of clay coatings, were selected for µXRD mapping.

These zones will be mapped by µXRD on the DIFFABS beam line. Both transmission and reflection will be used in order to optimise both the beam size (10 or 70 µm depending on the chosen mode) and the beam attenuation and thus sensitivity. To identify smectite from the other minerals present in the coatings (illite, kaolinite, chlorite or quartz), we will map an angular domain ranging from 1,7 to

17 °2theta at 17 keV. This angular domain will allow to detect the main peaks of these minerals. For that, we will use a 2D detector such as XPAD or MAR CCD and the µbeam setup of the line.

XI.2.1.4. *Results expected*

Mineralogical cartography will allow identifying and quantifying the new deposits containing smectites. We suspect that the particle deposition occurs preferentially in the active part of the porosity during the rain events. We will thus identify i) the proportion of porosity that has been active for the deposition over the experiment as a function of the rain intensity, ii) the location of particle deposits either directly along the macropore channels or in the surrounding microporosity and iii) the kinetics of the process.

XI.2.1.5. *Beam time requested justification*

The present project requires approximately 15 shifts: 5 shifts for setup adjustments and 10 for the mapping of the 6 thin sections.

XI.2.1.6. *References*

Cornu, S., Quénard, L., Cousin, I., Samouëlian, A., 2014, Experimental approach of lessivage: quantification and mechanisms. Geoderma.

Quénard L., 2011, Analyse d'un processus pédogénétique par une approche expérimentale : le lessivage des argiles dans le sol, Thèse de doctorat.

Sauzet O., 2012. Quantification by a micromorphological approach of argilluviation and bioturbation rates in soils in response to land use and agricultural practices changes at a pluri-decennial time scale. Master degree report.

Akamigbo, F.O.R and Dalrymple, J.B., 1985. Experimental simulation of the results of clay translocation in the B horizons of soils; the formation of intrapedal cutans. Journal of Soil Sciences, 36, 401-409

Bruand A, Cousin I, Nicoullaud B, Duval O, Bégon JC. Backscattered electron scanning images of soil porosity for analysing soil compaction around roots. Soil Sci

Soc Am J 1996;60:895–901.

Dalrymple J. and Theocharopoulos S., 1987. Intrapedal Cuntans – Lateral Differences in Their Properties and Their Spatial Clustering. Geoderma, 41, 149-180.

Legros, J.P., 2007. Les grands sols du monde. Presses Polytechniques et Universitaires Romandes, Lausanne. 574 pp.

Mercier, P., Denaix, L., Robert, M., de Marsily, G., 2000. Caractérisation des matières colloïdales évacuées au cours du drainage agricole: incidence sur l'évolution pédogénétique des sols. Comptes Rendus de l'Académie des sciences 331, 195–202.

Thompson M., Fédoroff N., Fournier B., 1990. Morphological Features Related to Agriculture and Faunal Activity in the Three Loess-derived Soils in France. Geoderma, 46, 329-349.

XI.2.2. Compte rendu d'expérimentation

<table>
<tr><td></td><td>Experiment title :
Localisation of particle deposition in soils: process identification towards modelling</td><td>Experiment number 20130377</td></tr>
<tr><td>Beamline : Diffabs</td><td>Date of experiment :
From 12/11/2013 to 18/11/2013</td><td rowspan="2">Date of report 01/03/2014</td></tr>
<tr><td>Shifts : 15</td><td>Local contact : Cristian Mocuta</td></tr>
<tr><td colspan="3">Names and affiliations of participants:
Romain Van den Bogaert, INRA, UR 1119 Géochimie des Sols et des Eaux, F-13100 Aix en Provence, France
Sophie Cornu, INRA, UR 1119 Géochimie des Sols et des Eaux, F-13100 Aix en Provence, France
Daniel Borschneck, CEREGE, CNRS, IRD, AMU, F-13100 Aix en Provence, France</td></tr>
</table>

XI.2.2.1. *Objectives of the experiments*

Lessivage is a substantial vertical transfer of fine particles, from a surface horizon to deeper horizon, that occurs in many soil types but is still poorly understood. A complex experimental setup was build up in order to simulate lessivage in lab. The lab experiment setup consists in a sequence of 30 rainfalls on undisturbed and unsaturated soil columns of decimetre size. In order to simulate separately eluviation and illuviation two experiments were run. As smectite were described as especially sensitive to eluviation (Mercier et al., 2000), the first experiment designed to simulate eluviation consisted in columns made of an undisturbed soil monolith containing smectite. The second experiment, designed to simulate illuviation, consisted in columns made of two overlaid soil monoliths, the upper one being that used for the eluviation experiment and lower-one containing no smectite. About 50 g m^2 particles were fixated within the 15 cm depth of the lower soil monoliths. Thin sections were made on the lower monoliths for different amounts of rain and different rainfall intensities. On these sections, clay coatings were identified along pores and their geometrical relationship to the porosity described. The aim of this project was to localise with respect to the soil porosity clay coatings containing smectite and thus considered as having been formed over the duration of the lab-experiment. The mechanisms responsible for this clay fixation with respect to the soil porosity will thus be interpreted thanks to the mapping of the mineralogical composition of the coatings and even quantified. This mapping is only feasible with a synchrotron light as the expected smectite concentration in the coatings are low and as the beam size of lab-apparatus is too large to allow the mapping of the coatings.

XI.2.2.2. *Data acquired during the experiment*

Two rain intensities were used over the experiment for two different sets of columns (20 mm h^{-1} and 6 mm h^{-1}). For these two rainfall intensities, columns were stopped after 20, 25 and 30 rains. Six thin sections coming from different soil columns were selected in order to have a kinetic gradient (20, 25 and 30 rains) and to compare the impact of the 2 rainfall intensities after 30 rains. On these different thin sections different types of coating were observed: old pore filling (cutanes), light brown coatings and blackish ones. These three types of coatings were mapped by µXRD on the DIFFABS beam line in the second half of November 2013. Transects of several tens of micron in size were selected on the different thin sections. Since the studied object were of only a few tens of microns in size and the interesting minerals were clay mineral that diffract in small angles, the experiments were performed in transmission mode at 17.2 keV, with a 2D XPAD camera and a µbeam of 10 µm per 6 µm. The analysed angular domain, ranging from 1.7 to 47 °2theta, was divided in four subdomains: 0.18-17; 9.3-24.7; 19.3-33.4; 24.3-38. Initial analysis was performed on pellets of the fractions less than 2 µm of the two soils and on a pellet the smectite extracted from the soil. This first analysis was designed to identify the diffraction peaks of interest.

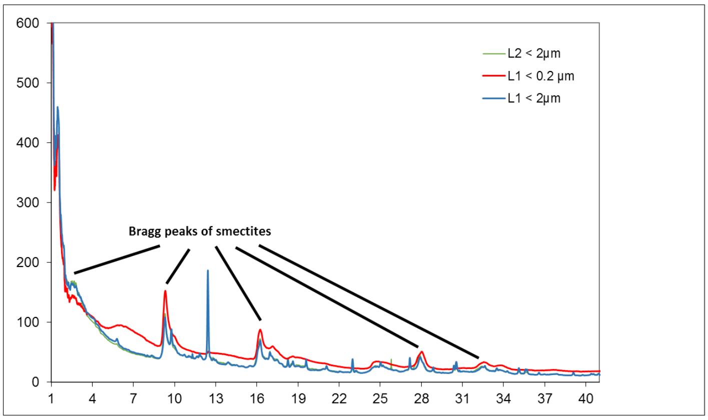

Fig.1: X-ray diffraction at 17.2keV of the 3 pellets used to locate the most characteristic peaks of smectite.

On figure 1, a peak around 3° (2θ) is identified as specific of the smectite phase. Other peaks may also be considered but difficulties may arise from superposition or proximity with peaks from other clays (kaolinite, illite). Then 28 zones of coatings located using an optical microscope, including 16 light brown coatings, 10 dark coatings and 2 cutanes, were mapped with 10µm step. Micro X-ray fluorescence data were simultaneously measured in order to differentiate XRD-patterns of the coatings (enriched in Fe), from those of the resin of the pores or those of the matrix (Figure 2). Thus each XRD-pattern may be assigned to one of these three classes for each studied zone.

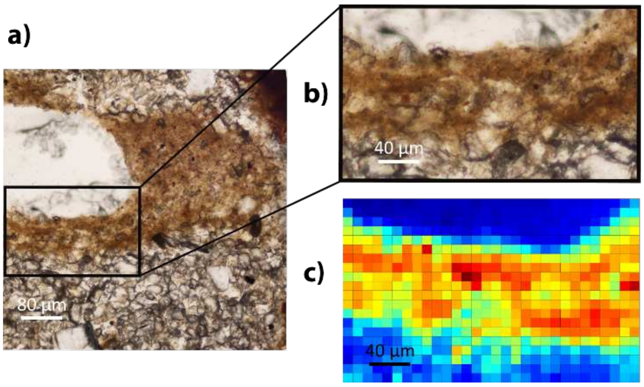

Fig. 2: Analyse of a dark coating: a and b- optic microscopy pictures, c- map of Fe by synchrotron X-ray fluorescence

This represents 12898 XRD-patterns and a total mapped surface of 2.5 mm^2. All the data were pre-processed thanks to a succession of python and ImageJ macros implemented by Cristian Mocuta: extraction of the raw diffraction image and background, background subtraction, Bragg rings straightening and integration to get a 2θ, I scan. As the macros had to be adapted to our samples, we could not see the results as we went along, so we could not adapt our experimental strategy. The following difficulties were encountered:

1. as working in transmission, the direct beam prevent using informations for angles lower than 3° in 2 theta. This is the range were the most characteristic peak of the smectite (001) appears at 17keV (15 Å, 2.75°(2θ)). Other peaks have to be used to detect this mineral but they are less specific of smectite (Figure 3).
2. The analyse volume is small (10*10*30 µm) when compared to the size of the studied crystals (generally larger than 2 µm), then few Bragg rings were observed rather spots as for monocrystal diffraction;
3. As the particles for identical mineralogical composition area were not always orientated in the same direction, the diffraction spots were not recorded for the same angle of diffraction;
4. These two last characteristics resulted in that when summing up different XRD-pattern of a studied zone, peaks disappeared. More sophisticated methods of data processing had thus to be found as the number of XRD pattern prevent manual interpretation.

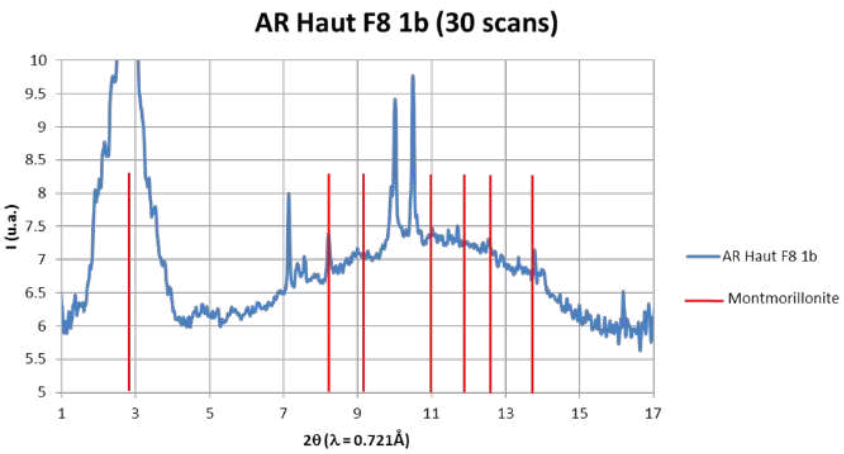

Fig. 3: Sum of 30 X-ray patterns of the dark coating mapped on figure 2.

First steps were (1) elimination of the noisy zones at both ends of the different angle ranges considered; (2) correction of the angular discrepancy among the different XRD-pattern thanks to the peaks of quartz; (3) identification of the diffraction peaks. Python macros were implemented for these treatments, unfortunately the method implemented for peak identification (simple method using a ratio of signal/noise larger than a given threshold for peak definition) yields a too large number of peaks.

XI.2.2.3. *Conclusion and perspective*

So far, many technical difficulties were encountered in the processing of the acquired data, we are thus still unable to conclude on our ability to detect the smectite in the studied coatings with the synchrotron XRD. We are building up a collaboration with statisticians in order to process the data. One trail would be to use an aggregation method to determine the peaks before a PCA analysis. Such a method was

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Analyse des mécanismes du lessivage des argiles dans les sols par des approches expérimentales

Résumé

Parmi les processus pédogénétiques, le lessivage des argiles dans les sols est un processus très répandu qui consiste en un transfert de particules, d'un horizon supérieur éluvié vers un horizon illuvié. Néanmoins, un certain nombre de lacunes persiste dans la compréhension de ce processus. Nous avons donc étudié : les processus d'écoulement de l'eau dans la porosité active, l'effet des cycles pluie - interpluie sur la rétention des particules, et les conséquences des perturbations physico-chimiques de la solution du sol sur la stabilité en suspension des particules de sols. Pour aborder ces différents points, nous avons fait le choix d'une approche réductionniste de laboratoire couplée à des méthodes analytiques modernes, souvent peu usitées en sciences du sol. Comme matériaux modèle, des horizons E de luvisol non remanié et des particules argileuses naturelles extraites de ces mêmes horizons ont été utilisés. Pour la première fois nous avons pu visualiser que les écoulements macroporeux préférentiels s'effectuent sous forme de ruisselets. Notre travail permet en outre, une première avancée vers l'estimation de la surface de contact eau – sol. Nous montrons que la rétention des particules s'effectue dans une zone de matrice à proximité macropores actifs, zone plus ou moins importante selon la durée de l'interpluie. Nous avons enfin montré que les variations de la concentration en calcium et du pH de la solution, consécutives à l'infiltration de l'eau de pluie dans la porosité, provoquent une évolution de l'arrangement des particules en suspension sur une échelle de temps correspondant à la durée d'un évènement pluvieux.

Analysis of clay lessivage mechanisms in soils by experimental approaches

Abstract

Clay translocation is a widespread process of particle transfer from a surface eluviated horizon to a deeper illuviated horizon. Nevertheless, a number of gaps persist in understanding this process. We therefore investigated: the water flow processes in the active porosity, the effect of rain-interrain cycles on the retention of particles, and the consequences of physical and chemical disturbances of the soil solution on the stability of soil particles in suspension. To address these issues, we have made the choice of a laboratory reductionist approach coupled with modern analytical methods, poorly used in soil science. As a model materials, we used undisturbed luvisol E horizons and natural clay particles extracted from these horizons. We show experimentaly for the first time that macroporous preferential flow consist in rivulets. Our work also provides a first step towards estimating the water-soil specific surface area. We show that particle retention occurs in the matrix close to active macropores, the thickness of the concern matrix being a function of the interrain duration. Finally we have shown that the variations of the calcium concentration and pH of the soil solution, subsequent to the infiltration of rain water into the pores, cause a change in the arrangement of the particles in the soil suspension on a timescale corresponding to the duration of a rainfall event.