# Dosimetric study of sediments at the beta dose rate scale: Characterization and modelization with the DosiVox software

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## HIGHLIGHTS

- The DosiVox software allows calculating beta dose rates at the grain size level.
- A micro-stratified sediment is characterized and a model created.
- Factors inducing significant micro-dosimetric effects are identified.
- The spatial distribution of grains and/or radioelements are critical factors.

## ARTICLE INFO

## ABSTRACT

The effects of sediment heterogeneity on beta dose rate have been investigated by simulation with the DosiVox software. Basic sediment cases, as well as a model of a micro-stratified sediment from the Mas d'Azil cave have been modeled at a few centimeters scale. The results of the simulations have highlighted different factors having a significant impact on the beta dose rate dispersion, among which the heterogeneity of the radioactive elements, the distribution of grains in the matrix and their proportion in the sample. These factors contribute to enlarge beta dose distributions and even create complex ones, and inevitably induce errors in the dating process. These effects are discussed, as well as the potential of the simulation to calculate beta dose rates in sediment samples and the necessity of using sampling protocols adapted to sediment complexity.

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## 1. Introduction

The dating of micro-stratified sediments presents a challenge in luminescence dating. With the development of the single-grain dating technique that allows getting information on the absorbed dose at the grain size level (Duller et al., 1999), spatial variations in the material properties as well as in the distribution of the radioactivity for distances smaller than a few millimeters have to be considered since they can induce grain to grain variations in beta dose rates (*e.g.*, Nathan et al., 2003; Kalchgruber et al., 2003; Mayya et al., 2006; Cunningham et al., 2012; Guérin et al., 2012). In this study, we use the DosiVox software to construct models of multi-layered sediments and calculate the beta dose distribution. All references to dose rate in this paper concern only the beta dose rate contribution. In a first part, simplified models of sediments are described, followed in a second part by the modeling of a sediment sampled from the Mas d'Azil cave (France).

## 2. Sample and methodology

The Mas d'Azil cave (France) has been repeatedly carved by the Arize river during the Pleistocene and it is nowadays partly filled with alluvial deposits (Pallier et al., submitted). The sample considered in this study was taken by inserting a metallic box of $10 \times 5 \times 5$ cm in these deposits. It was indurated directly in the box using a polymer resin and cut in the vertical plane in order to extract 1 cm thick slices. The macroscopic appearance of the sample and the different analyzes performed on the slices indicated a succession of micro-layers in the vertical axis (Fig. 1a). Considering the similarity between the different slices, it was assumed that this sample is relatively homogeneous in the horizontal plane. This structure also suggested interesting dosimetric effects, typically at the scale of single grains, and as such it seemed well suited for a modeling study with the DosiVox software to explore dispersion in beta dose rates.

DosiVox is a software dedicated to dosimetric simulations (Martin et al., submitted). It is based on the Geant4 toolkit (Agostinelli et al., 2003) and allows simulating particle–matter interactions following a Monte-Carlo approach. In DosiVox, the user defines a three dimensional grid and sets the material chemical composition, density and water content, as well as the different sources of radioactivity in each voxel. A graphical interface allows a user-friendly input of these data for a grid size of $20 \times 20 \times 20$ voxels. Dose recorders can be superimposed on this grid: in our study, quartz grains representing randomly packed spherical grains acted as detectors. This packing takes into account the composition and granulometry of the grains, and the compactness of each sediment layer defined by the user. The dose absorbed by each grain during the simulation is then recorded. However, performing such simulations necessitates getting information on the chemical composition of the sediment and on the distribution of the main natural radioactive isotopes (U series, Th series and $^{40}$K).

Three different techniques were applied for characterizing the composition of this sample: X-ray fluorescence (XRF) was carried out for mapping the major elements and identifying a region of interest (ROI) (Fig. 1a and b). This ROI (~33 mm long) is formed by thin alluvial deposits of clay and sand containing fine and coarse grains. Major elements were mapped using a scanning electron microscope (SEM) on the ROI to collect data about the micro-layers: these images allowed the identification of the grains and the granulometry of the different layers was obtained by image processing.

In order to obtain information on the U, Th and K distributions, transects in the vertical axis were carried out using a femto-second laser ablation ICP-MS. During this process, information concerning the major and minor elements were also collected in order to characterize the micro-layers, in addition to the SEM data. U, Th and K contents were determined all along the vertical profile of the ROI. To complete these data, a beta ray auto-radiography image was taken indicating potential heterogeneities in the beta dose rate (Ruffer and Preusser, 2009) (Fig. 1c).

## 3. Simulation results for simple cases

Before creating a model of the Mas d'Azil sample, it was important to make sure that the DosiVox software provides reliable results when simple cases are modeled and that it allows identifying the main factors of dose variation. To test this software, the following two cases were modeled: 1) a case where quartz grains are embedded in an almost uniform infinite matrix (grains and matrix proportions in the sediment are respectively 5% and 95% in volume)and 2), a case of sediment containing a single micro-layer of sand between two clay levels.

### 3.1. Sedimentary grains in an infinite matrix

The purpose of this model is to test the accuracy of the DosiVox software when simulating infinite matrix conditions, which is the dosimetric model commonly used in luminescence dating (Aitken, 1985). In this simulation, all the voxels of the $20 \times 20 \times 20$ grid are filled with siliceous material. In the central vertical column of the grid, non-radioactive quartz grains with diameters of 25, 50, 100 and 200 μm and density of 2.65 g cm$^{-3}$ are explicitly simulated and distributed according to Fig. 2. These grains represent 5% of each voxel volume in order to keep a small impact on the infinite matrix assumption. These grains were created from the silica presents in
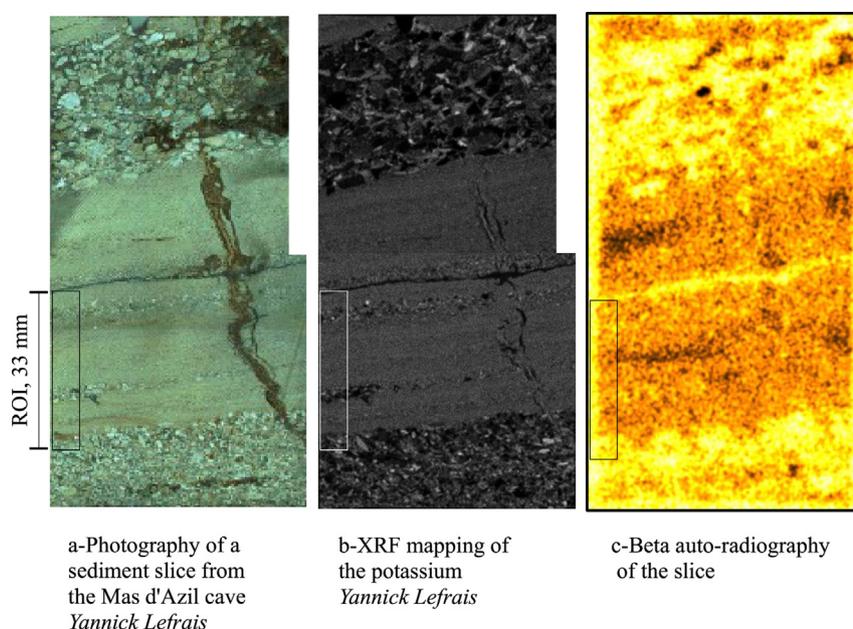


a-Photography of a
sediment slice from
the Mas d'Azil cave
*Yannick Lefrais*

b-XRF mapping of
the potassium
*Yannick Lefrais*

c-Beta auto-radiography
of the slice

**Fig. 1.** Qualitative characterization of the Mas d'Azil sediment heterogeneity on a thick slice.
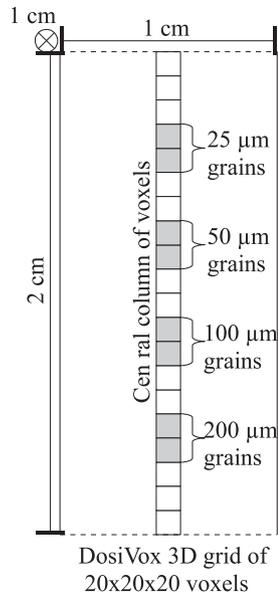
**Fig. 2.** Simulation model of quartz grains in homogeneous infinite quartz matrix. The grey voxels of the central column contain grains of the indicated size representing 5% of the voxel volume. The other voxels contain an homogeneous material formed by the mix of the matrix and the grains.

the voxel. This ensures that all the voxels of the grid keep containing on average exactly the same material composition. Moreover, a single uranium content (with the U-series at secular equilibrium) was considered in all the voxels, which thus represents a uniform radioactive matrix. Fig. 3 presents the results of the beta dose distributions obtained in different cases: for dry materials made of $SiO_2$ with a density of 2.65 g cm$^{-3}$ (1) or a density of 2.0 g cm$^{-3}$ (2), or for materials made of clay (3) (composition in mass: $SiO_2$ 60%, $Al_2O_3$ 30%, $Fe_2O_3$ 10%). Moreover, results for a matrix made of $SiO_2$ material containing 20% of water (in mass) are also shown (4). The grain size attenuation factors from Guérin et al. (2012) are represented on the figure for comparison, as well as when the water content attenuation factor of the beta dose given by Nathan and Mauz (2008) for 20% in mass of water is applied to them. The error bars on the graph represent twice the standard deviation of the successive simulations.
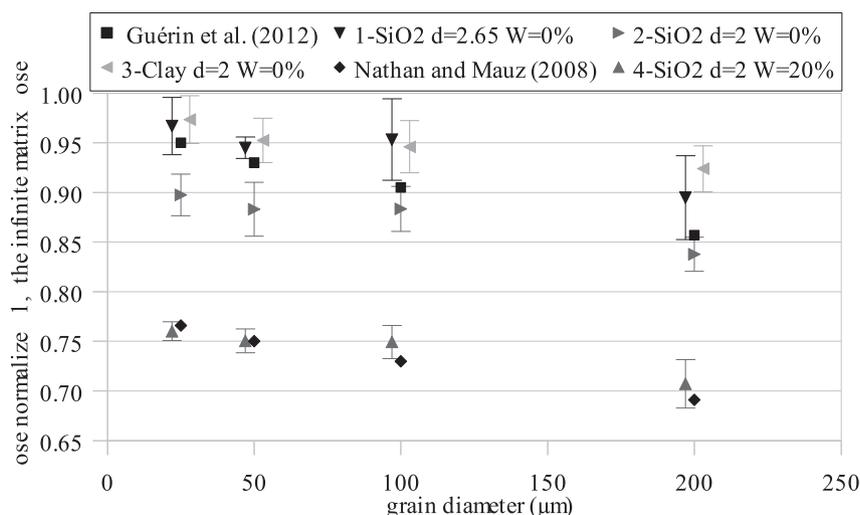
Our simulation results are close to the tabulated data considering the uncertainties, especially for the wet sediment (4). The systematic increase in the simulation values observed for larger diameters may be due to geometrical effects since grains of these size start to represent an non-negligible heterogeneity in regard of the infinite matrix assumption, even in small proportion (Guérin et al., 2012). Meanwhile, the results obtained for the dry matrix of $SiO_2$ with a density of 2.65 g cm$^{-3}$ (case 1) are in agreement with the previous data within uncertainties, as well as are the results we obtained for the wet material (case 4). These setups represent the standard case used for calculating attenuation factors, so it seems that DosiVox is able to provide results similar to the previously published data. The dose rates obtained in the other cases differ only by a few percents from the values given by Guérin et al. (2012), but it is likely that these small differences result from the differences in the beta stopping power coefficients or in the particle scattering in the different matrix materials, as it was observed for carbonate-rich sediment by Nathan and Mauz (2008). The importance of considering the chemical composition and density of the sediment in the dose rate calculation is then highlighted here.

The extent of the error bars, in particular for the largest diameters, results from the low number of grains created (5% of the total volume) compared to the matrix occupying the remaining volume. This limits the probability of a beta particle to reach a grain, and consequently affects the statistical representativeness of the simulation results. Meanwhile, this low proportion is necessary for keeping the conditions close to the infinite matrix model. Additional and/or longer simulations would allow reducing the results dispersion.

### 3.2. Sediment with a single micro-layer

The second model represents a 4 mm micro-layer of sand (layer 2) between two layers of clay (Fig. 4): a "standard" clay (layer 1) and a clay rich in organic matter (layer 3). The standard clay (density = 2 g/cm$^3$; water content = 10%) contains 100 μm quartz grains with a compacity of 5%, whereas the "organic" clay contains the same grains but has a lower density (1.8 g/cm$^3$) and a water content of 15%. The sand layer (layer 2) is made of quartz grains of 50, 100 and 200 μm which represent respectively 15, 45 and 40% of the total volume of the grains. The compactness of these grains is 60% and this layer has a water content of 18% and a density of 1.9 g/



**Fig. 3.** Attenuation factors for quartz grains close to the infinite matrix conditions. Results from DosiVox simulation compared to tabulated data. "d" is for density in g cm$^{-3}$ and "W" is for water content in mass percent.
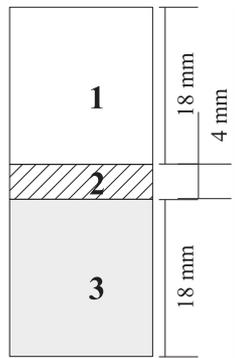
**Fig. 4.** Simulation model of quartz grains in a sediment with a sand micro-layer 1-clay, density = 2 g cm$^{-3}$, WF = 10% 100 μm grains with a compacity of 5% 2- sand, density = 1.9 g cm$^{-3}$, WF = 18% 40% volumic of 200 μm grains 45% volumic of 100 μm grains 15% volumic of 50 μm grains with a total compacity of 60% 3- clay with organic matter, density = 1.8 g cm$^{-3}$, WF = 15% 100 μm grains with a compacity of 5%.

cm$^3$. This model is homogeneous in the horizontal plane and allows evaluating the beta dose heterogeneities in the different grain sizes, and to identify the main parameters involved in the dose variations. The grains were only explicitly simulated in the central column of the grid and defined as radioactive free. Simulations were carried out for the beta rays from the Th-series (at secular equilibrium). The dose distributions in the 100 μm grains are displayed on Fig. 5 for the case where layers 1, 2 and 3 have contents of 2, 1 and 3 ppm respectively, and for the case where the Th content in sediment dry mass is the same in all the layers. The different tints represent the

layers of origin of the grains. The two voxels at the top of the grid and the two at the bottom were excluded because of edge effects (Martin et al., submitted).

In the case of the heterogeneous distribution of the radio-elements (Fig. 5a), a large scatter of the dose is observed. This is mainly due to the difference in Th concentrations in the three layers since three peaks corresponding to the three strata are observed. One can however notice that the sand layer peak is positively skewed because of the influence of the more radioactive clay levels surrounding it. But what is certainly the most interesting observation is the fact that if one calculates the beta dose rate received by the 100 μm grains in considering the attenuation factors of Nathan and Mauz (2008) for moisture and Guérin et al. (2012) for grain sizes, and in taking the averaged Th and water contents measured on the bulk sample, a difference of about 30% would be found between this average dose rate and the average dose value really received by the 100 μm grains. This difference can be explained by the over-representation of these grains in the less radioactive sand layer compared to the more radioactive clay levels. Without a knowledge of the spatial heterogeneity of the Th contents, this large distribution of doses could then be interpreted as the results of the depositional chronology, or as the presence of partially bleached grains (what would seem in that case in accordance with the positive skewness of the sand peak in Fig. 2) while only dosi-metric phenomena are involved. It is then clear that a lack of information about the radioactive heterogeneities at a small scale can induce important errors in dose rate calculations, particularly when none of the peaks are centered onto the average dose received by the selected grain fraction.
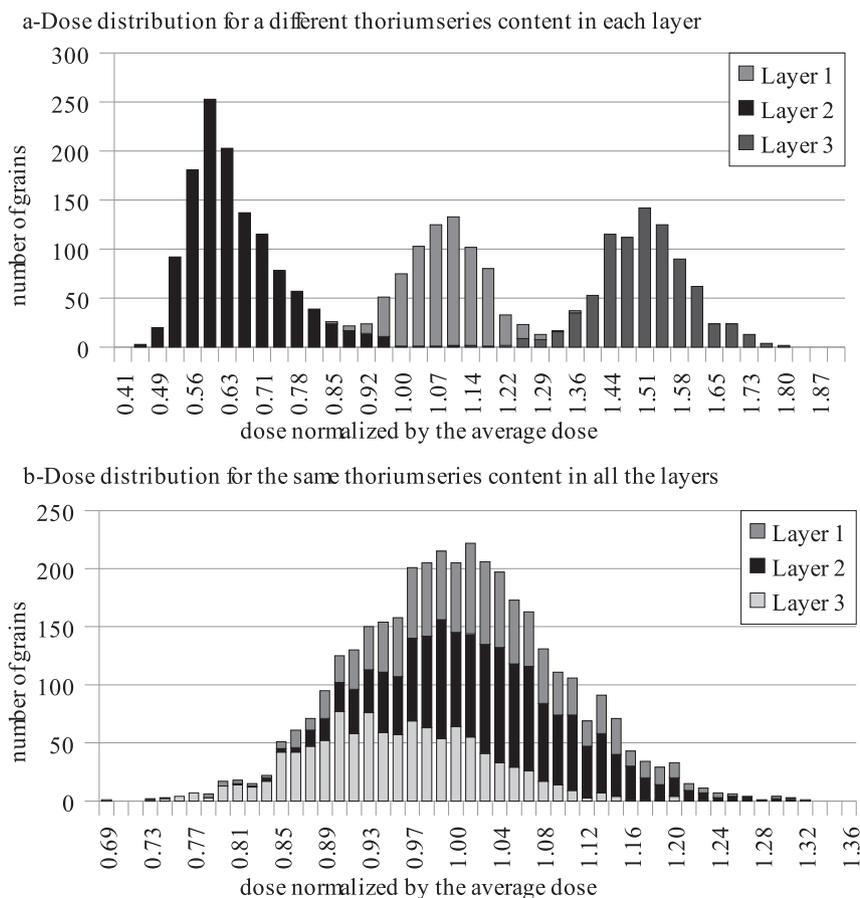


**Fig. 5.** Dose distribution in the 100 μm grains. The tints indicate the stratum of origin of the grains.

If a homogeneous distribution (in dry mass) of radioactive emitters is now considered (Fig. 5b), the dose dispersion is considerably lower but small dosimetric variations are still observable. The differences in the doses received by the grains belonging to the different levels cannot be explained only by the water content differences: if one considers the sand peak (layer 2) and the clay level 3, the average dose in the sand is higher than in the clay whereas the sand layer already contains more water. One can hypothesize here that the differences in the chemical composition and compactness of the two layers are the main factors controlling the differences in the simulated doses. The effect of compactness was already noticed by Guérin et al. (2012).

If one now considers a standard calculation of the dose rate as it was done in the previous case (*i.e.* in determining the Th series elements and mean water contents on the bulk sample), the calculated dose rate is about 7% higher than the dose received on average by the grains. This difference can be explained by both the important quantity of 100 μm grains in layer 2 that balances the high water content of this stratum, and by the differences in the beta rays stopping powers of the different simulated materials, compared to the stopping power of the material for which the usual beta attenuation factors had been calculated (Nathan and Mauz, 2008; Guérin et al. 2012).

To sum up, these two simple modeling show that, in addition to the spatial heterogeneity in the radioelements distribution, it is important to consider the distribution and compactness in the sample of the dated grains, in each layer if applicable. The beta stopping power differences between two different media seems to have a non-negligible influence on the dose rate too. Similar effects have been highlighted by Nathan (2010). The simulations done with the DosiVox software could help considering these issues in taking into account these different factors, at the condition of an extended sampling process requiring to consider the sediment layout and characteristics at small scales. In order to illustrate the information that can be brought by the dosimetric simulations taking in account data from beta scale analysis, one considers now the proposed modeling of a real sample from the Mas d'Azil cave.

## 4. Reconstruction of the Mas d'Azil sample and results of the simulation

### 4.1. Data processing

The first step for reproducing the sediment in the region of interest (ROI) was to identify the different observable strata. Only a qualitative identification was required because of the low definition of the chosen DosiVox grid (20 voxels in each axis) and because the purpose of the present study was to evaluate the possible micro-dosimetric effects. Note that creating more accurate simulations would require analysis with a higher spatial resolution and would be considerably more time consuming.

Fourteen layers were identified in the ROI based on both the microscopic layout seen on the SEM images (Fig. 6) by considering in particular the grain sizes and their relative proportions, and the profiles obtained with the LA-ICP-MS measurements giving the chemical composition of the major constituents. Approximations were made about the thickness of each stratum to fit an integer number of voxels; each one has then a thickness of 1.67 mm (corresponding to the height of the ROI -33 mm-divided by 20 voxels on the Z axis). We also assumed that this basic division of the sample allows making representative simulations in order to evaluate the heterogeneities effects on the beta dose.

The chemical composition of each represented stratum was calculated by averaging the contents of the major components ($SiO_2$, $Al_2O_3$, $Fe_2O_3$ and $K_2O$) in the corresponding level (Fig. 7a).
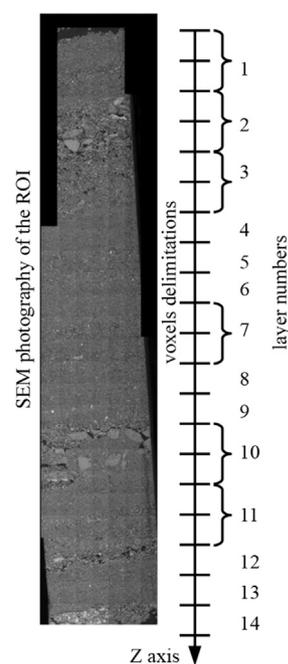


Fig. 6. Identified layers in the region of interest.

These contents were normalized by the sum of the mass percentage of all major components in order to compensate for any variation in the quantity of matter ablated by the laser. We assumed that all the iron atoms were in the third oxidation degree and that the mass percentage of the minor elements was negligible, but further analysis and simulations would be required for confirming these hypotheses or measuring their impact on the simulated dose rate.

The uranium and thorium contents of each layer were also measured by LA-ICP-MS and normalized with the sum of contents
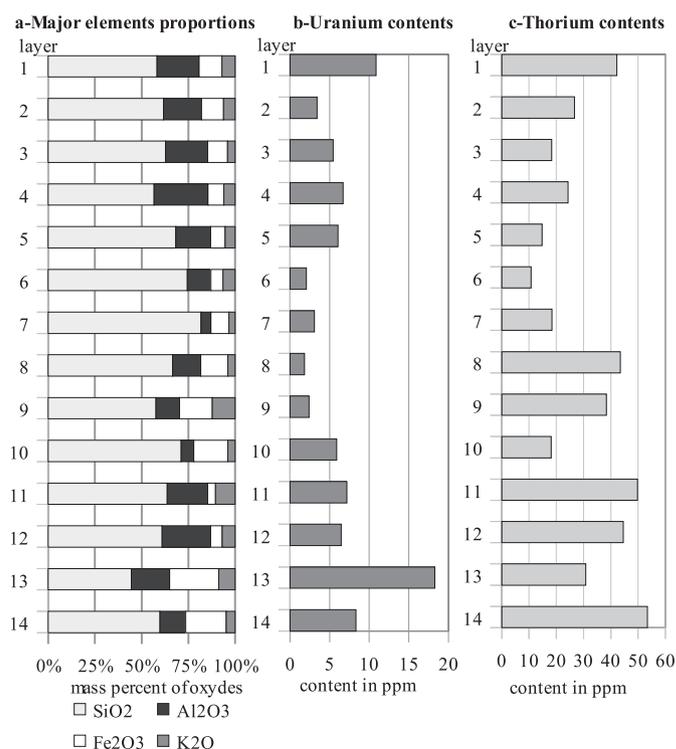


Fig. 7. Elements contents in the different layers.

of the major elements (Fig. 7b). Before averaging the contents in each layer, the out of range values detected by the ICP-MS were suppressed since they were out of the detector calibration, and are then suspected to be not accurate. These values may come from high concentrations of uranium or thorium localized in small areas. Considering their low occurrence, we assumed that their contribution to the beta dose rate in the ROI is negligible. A mapping of the uranium content in layers 2 and 3 showed that the uranium is mainly distributed in the matrix surrounding the quartz grains (Fig. 8). This observation leads us to represent in this model quartz grains free of radioactivity.

The grain size distribution and compactness of these grains in each layer were determined by image processing with the ImageJ software (Abràmoff et al., 2004; Rasband, 1997–2012, Schneider et al., 2012) applied to the SEM mapping of the major chemical elements. Assuming that, in a single layer, the grain spatial distribution is isotropic, the compactness of the grains can be calculated as the ratio of the intercepted surfaces of the grains and the surface of measurement (Degallaix and Ilschner, 2007). The apparent porosity of each layer was calculated in the same way, using the ratio between the intercepted surfaces of the apparent porosities and the surface analyzed. This measure is highly dependent on the image resolution, so it can only be applied with accuracy to objects with dimensions much larger than the resolution. As it was not always the case in this study because of a resolution arround 10 μm, the exactness and accuracy of the obtained data are subject to discussion.

For each layer, the sizes of the intercepted grain surfaces were used to calculate the grain size distribution by applying stereological corrections with the CSD (Crystal Size Distributions) software (Higgins, 2000, 2006). These corrections are limited by the image resolution, by the assumptions made on the grains shape and proportions, and by the statistical representativeness of the number of grains.

The density and water content of each layer were chosen by a qualitative approach, considering both their material identification based on the chemical composition and the apparent porosity determined by image processing. Quantitative and accurate data would require much more analysis and processing, which is not the purpose of this sample reconstruction. These properties can be modified afterward in considering other hypotheses in order to assess the influence of these parameters on the calculated dose rates.
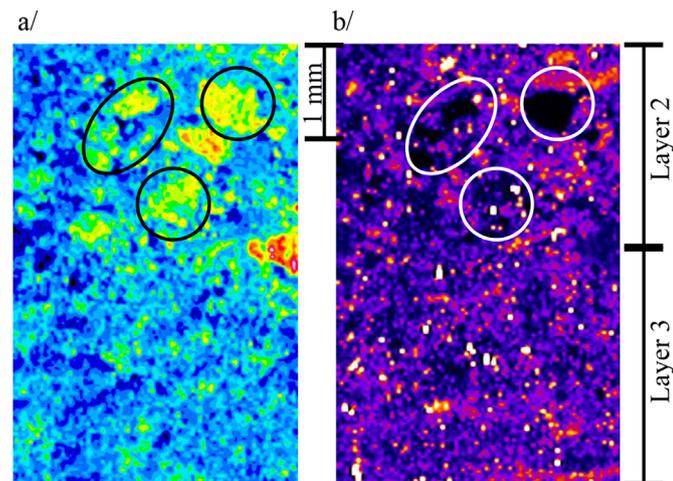
## 4.2. Simulation results

All the data related to chemical composition, water content, grains size and uranium distribution were used to create the material present in each layer of the model. The DosiVox grid was filled with these materials; in this model, each layer of sediment is represented by one or two layers of voxels, according to the stratigraphy (Figs. 6 and 7). Simulations of the beta radioactivity were carried out using the U-series spectrum at secular equilibrium. Non-radioactive quartz grains were explicitly created in the voxels of the central column for recording the doses. Apart from these radioactive free inclusions, Uranium emissions were sampled homogeneously for each Layer, based on its average content, in the matrix between the grains. Fig. 9 represents the obtained dose rate distributions in the different grain sizes (30–40 μm; 60 μm and 90–110 μm). The different fillings on the figure represent the proportion of grains from the corresponding layer. The arrows indicate the beta dose rate which would have been calculated considering the average uranium and water contents in all the ROI, the beta-dose attenuation factors available for the three grain sizes considered here Guérin et al. (2012), and moisture Nathan and Mauz (2008).

For each grain size, a large dispersion in the dose rates is observed. It is noticeable that the dose distributions for grains from layer 1, localized at the top of the simulated sequence, are globally underestimated and skewed towards the lower doses because of edge effects. For the other layers, the dose distribution and the position of standard calculated dose rate compared to the average grain dose rate are different from a grain size to another. In addition to the radioactive heterogeneity, various factors are involved in this phenomenon. The main one is the grain size distribution of the grains in the different layers: Guérin et al. (2012) have already shown that grain size attenuation factors are relative factors depending not only on the size of the grains of interest, but also on the grain size distribution and compactness of the sample taken as a whole. In addition, this local dose rate is affected by attenuation factor variations: the chemical composition, density (as already observed by Nathan, 2011) and water content in a giving layer affect the beta particles diffusion and the matrix stopping power. Similarly, Guérin and Mercier (2012) have shown that water content attenuation factors for gamma dose rates are also grain size dependent. Moreover, we can hypothesize that grains close to the boundary between two strata are affected by the dose rates and the local attenuation factors specific of these two strata.

As a conclusion, the calculation of the dose rate based on a method consisting in averaging the characteristic values of each individual layer is not adapted for laminated sediments when the thickness of the layers is similar to the beta particles range. At least, the grain size distribution of the sediment and the water content variations between the different layers should be taken into account to make a correct estimate of the dose rate.

## 5. Discussion

It is noticeable that all the results presented in this paper concern only the beta dose contribution. The total dose distribution to the grains will be affected by the intensity and the heterogeneity of the different contributions. The gamma and cosmic rays contribution would tend to reduce the relative dispersion in dose rates because their variations are little significant at the scale of the models presented in this study (few centimeters of height). At the opposite, it is likely that the effective alpha contribution would tend to widen the dose dispersion because, like the beta contribution, it is sensitive to the emitter distribution in the sample and to the properties of the
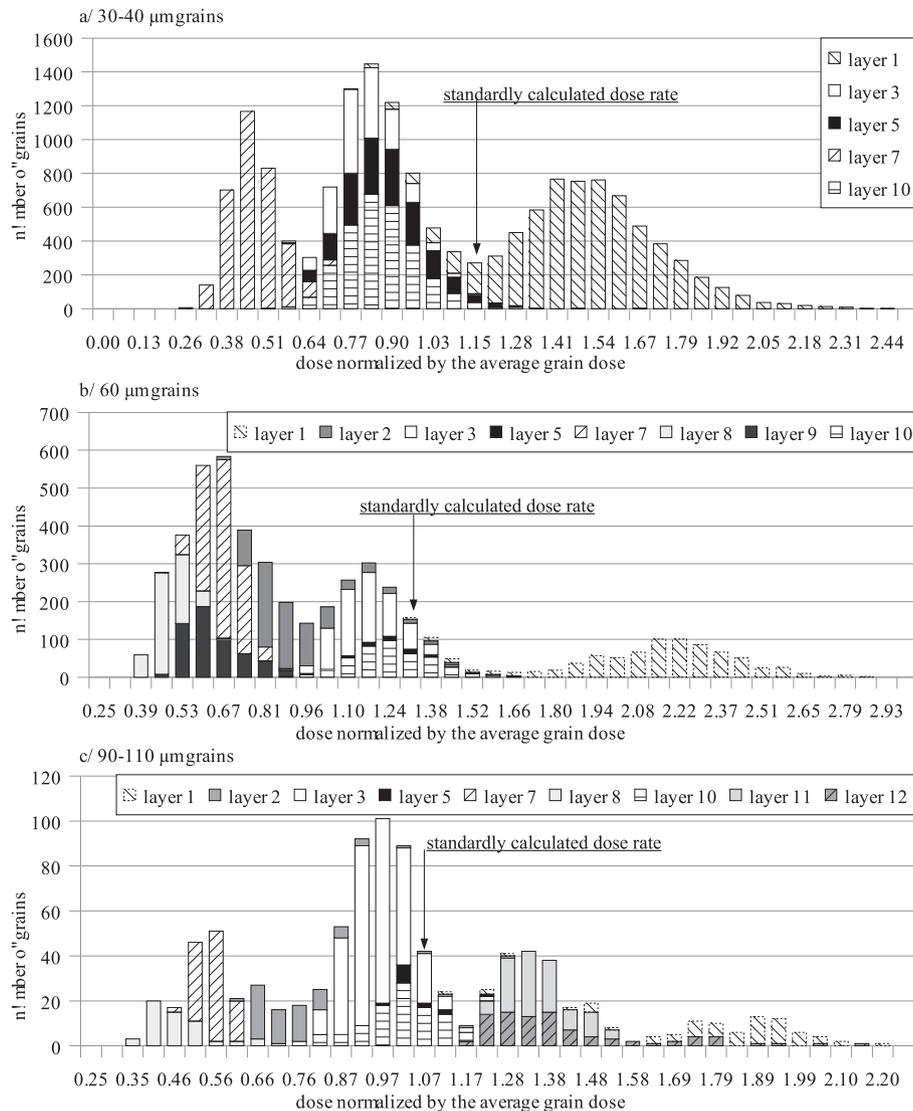


a/  b/

**Fig. 8.** a-Silica and b-Uranium mapping on the layers 2 and 3. The circled areas correspond to coarse quartz grains.

**Fig. 9.** Dose distribution for different grain sizes from the simulation model of the Mas d'Azil sediment.

different matrices. For grain sizes significantly affected by the alpha contribution, similar simulations for the alpha radioactivity would be required. However, if etched coarse grains are considered, the beta dose usually counts for the two third of the total dose, when the remaining third comes from the cosmic and gamma contributions. In this case, the weight of the calculated beta dose distribution to the grains remain considerable.

This study does not intend to use simulations for calculating accurate beta dose rates in complex sedimentary environments. It only shows a possible way to construct models for samples of interest, in order to study the parameters influencing the dose rate, in particular their impact on the beta dose. Although DosiVox allows constructing models with a relatively high resolution (since the number of voxels is mainly limited by the RAM memory of the computer), it has been chosen to limit to 8000 the number of voxels managed by the graphical interface in order to keep conditions compatible with a relatively fast and user-friendly system of modeling. This low resolution (20 voxels on each axis) is the most obvious limit of the models. Other limitations concerning the accuracy of the calculated doses which result from the software itself are detailed in Martin et al. (submitted). In addition to these limitations, it is obvious that the accuracy of the analysis performed on

a sample, the data processing and the unavoidable assumptions made during modeling have to be carefully considered too. All these error terms can contribute to limit the representativeness of the model and of the results. Although it is hard to believe that an accurate model of a complex sediment sample such as the Mas d'Azil one can be created, even a simple modeling can be considered better than applying the infinite matrix model which is not suitable in this case. However, the assumptions and limitations of his model should be borne in mind to avoid any over-interpretation of modeling results.

In our opinion, the main contribution of any dosimetric simulation tool like DosiVox for such complex sediment case is to allow the user assessing the impact of the different parameters which influence the dose rate, for example in running several simulations with different sets of values. The comparison of these results should give indications on the possible scatter of the dose absorbed by the grains (as presented above for the Mas d'Azil sample) and information on the most influencing parameters, leading the experimenter to consider them carefully. In this scope, a "coarse" model (*i.e.* with a low resolution, qualitative data) can often be a good option since it is faster to construct and requires less accurate data than a detailed one.

The set of analysis presented in this study constitutes only a possible way for getting the required data. Any protocol and technique allowing getting information about the material composition and its properties, the grain size distribution and the distribution of the radioactivity with the requested resolution can be implemented. In every case, the sampling process have to be considered in regard of the sample complexity at the beta scale, because of the considerable dependence of the model on the input data. Similarly, the simulation models discussed here constitute only one approach offered by DosiVox: actually, faster simulations can be carried out without explicitly creating the quartz grains. In that case, the user needs to balance afterward the doses recorded in each stratum with appropriate attenuation factors and grain proportions to calculate the dose rate distribution. Even if the attenuation factors can vary with the sediment properties, this modeling remains correct when the effect of dose rate heterogeneity between strata is much more important than the possible variations of these factors. It is also possible to create a 3D detailed image of the sample with a software like ImageJ, and to use this image as an input data for DosiVox to create a model with a higher resolution (Martin et al., submitted).

## 6. Conclusion

The DosiVox software allows to model dose rates in sediments with different levels of complexity. It is possible to study and calculate dose rate distributions in grains using this modeling. This can bring useful dosimetric information, in particular for single grain dating. Taking into account both the software limitations and the imprecision of the requested data for the model construction, the pertinence of quantitative results from the dose rate modeling of complex samples remains limited. However, a qualitative modeling still allows to access information on the sample dose rate and on the influence of various factors. DosiVox seems to be an efficient tool for studying the dosimetry in samples where the infinite matrix assumption is not valid. Taking into account the simulation results during the dating process could lead to a better understanding of the equivalent dose dispersion and contribute to reduce the uncertainty on the calculated age.

The simulations presented in this study highlight different factors having a significant influence on the beta dosimetry, in addition to the heterogeneous distribution of the radio-elements. Our results indicate that the most important parameter seems to be the proportion and distribution of the grains in the different layers of the sample. Ignoring this parameter can make the dose rates received by the grains in the sediment considerably different from the dose rate calculated in averaging the different levels of radioactivity present in the sample and considering the mean water content of the entire sample. Actually, our results show that the water content heterogeneity, the compactness of the grains in the matrix and its chemical composition may have a significant impact on the beta dose rate and its dispersion. These parameters should be taken into account for dating samples for which the infinite matrix assumption cannot be assumed. Dosimetric simulation models can be a solution for evaluating the impact of these various factors, but at the condition of considering sampling processes adapted to the complexity of the sediments in order to construct adapted models; it should also be noted that different superposed layers may indicate different deposition ages for the corresponding grains. As a consequence, such dose rate modeling associated with high resolution sampling may lead to enhanced chronological models. Since a software like DosiVox does not require any programming skills, it represents an interesting tool for this task if coupled with sample analysis protocols allowing to access the data at the beta radiation scale.

## References

Abràmoff, M.D., Magalhães, P.J., Ram, S.J., 2004. Image processing with ImageJ. Biophot. Int. 11 (7), 36–42.

Agostinelli, et al., 2003. Geant4 – a simulation toolkit. Nucl. Instrum. Methods Phys. Res. A 506, 250–303.

Aitken, M.J., 1985. Thermoluminescence Dating. Academic Press, London.

Cunningham, A.C., DeVries, D.J., Schaart, D.R., 2012. Experimental and computational simulation of beta-dose heterogeneity in sediment. Radiat. Meas. 47, 1060–1067.

Degallaix, S., Ilschner, B., 2007. Caractérisation expérimentale des matériaux I (TM volume 2), chapter 3. Presses Polytechniques et Universitaires Romandes, 9782880745677.

Duller, G.A.T., Bøtter-Jensen, L., Murray, A.S., Truscott, A.J., 1999. Single grain laser luminescence (SGLL) measurements using a novel automated reader. Nucl. Instrum. Methods B 155, 506–514.

Guérin, G., Mercier, N., 2012. Preliminary insight into dose deposition processes in sedimentary media on a grain scale: Monte Carlo modelling of the effect of water on gamma dose-rates. Radiat. Meas. 47, 541–547.

Guérin, G., Mercier, N., Nathan, R., Adamiec, G., Lefrais, Y., 2012. On the use of the infinite matrix assumption and associated concepts: a critical review. Radiat. Meas. 47, 778–785.

Higgins, M.D., 2000. Measurement of Crystal size distributions. Am. Mineral. 85, 1105–1116.

Higgins, M.D., 2006. Quantitative Textural Measurements in Igneous and Metamorphic Petrology. Cambridge University Press. http://dx.doi.org/10.2277/0521847826, 10.2277/0521847826.

Kalchgruber, R., Fuchs, M., Murray, A.S., Wagner, G.A., 2003. Evaluating dose-rate distributions in natural sediments using a-Al2O3:C grains. Radiat. Meas. 37, 293–297.

Martin, L., Incerti, S., Mercier, N., 2015. DosiVox: a Geant 4-based software for dosimetry simulations relevant to luminescence and ESR dating techniques. Anc. TL (being reviewed).

Mayya, Y.S., Morthekai, P., Murari, M.K., Singhvi, A.K., 2006. Towards quantifying beta microdosimetric effects in single-grain quartz dose distribution. Radiat. Meas. 41, 1032–1039.

Nathan, R.P., Thomas, P.J., Jain, M., Murray, A.S., Rhodes, E.J., 2003. Environmental dose rate heterogeneity of beta radiation and its implications for luminescence dating: Monte Carlo modelling and experimental validation. Radiat. Meas. 37, 305–313.

Nathan, R.P., 2010. Numerical Modelling of Environmental Dose Rate and its Application to Trapped-charge Dating. D.Phil. Thesis. St Hugh's College, University of Oxford, p. 207.

Nathan, R.P., Mauz, B., 2008. On the dose-rate estimate of carbonate-rich sediments for trapped charge dating. Radiat. Meas. 43 (1), 14–25.

Pallier C., Jarry M., Fritz C., Lacombe S., Lelouvier L.-A., Martin H., Rabanit M., Tosello G., Wattez J., Arrighi V., Salmon C., Bon F., Bruxelles L., Soumis, Evolution karstique et occupation humaine dans la grotte du Mas d'Azil (Ariège, France), Quaternaire, (submitted).

Rasband, W.S., 1997–2012. ImageJ. U.S. National Institutes of Health, Bethesda, Maryland, USA. imagej.nih.gov/ij/.

Ruffer, D., Preusser, F., 2009. Potential of autoradiography to detect spatially resolved radiation patterns in the context of trapped charge dating. Geochronometria 34, 1–13.

Schneider, C.A., Rasband, W.S., Eliceiri, K.W., 2012. NIH image to ImageJ: 25 years of image analysis. Nat. Methods 671. http://dx.doi.org/10.1038/nmeth.2089.