Quaternary Geochronology 37 (2017) 97–107

Contents lists available at ScienceDirect

Quaternary Geochronology

journal homepage: www.elsevier.com/locate/quageo

Research paper

Using R for TL dating

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ARTICLE INFO

Article history:
Received 7 March 2016
Received in revised form
29 August 2016
Accepted 10 September 2016
Available online 12 September 2016

Keywords:
R
TL
Thermoluminescence dating
Burnt flint

ABSTRACT

Whilst optically stimulated luminescence (OSL) is commonly more suitable for sediment dating because of faster signal resetting, thermoluminescence (TL) remains important for dating burnt material, e.g. in archaeological contexts, or for studying the luminescence properties of different materials. A lack of user-optimized analysis software for TL data has exacerbated the decline of TL dating in comparison to OSL. However, exciting developments in TL dating of flint and calcite indicate a rise in application of this underused method.

R is a programming language and environment for statistical computing and graphics. It provides a wide variety of statistical and graphical techniques and is highly extendable. A package specifically designed for luminescence data analysis is available. However, it mainly includes functions for the analysis of OSL data. The *TLdating* package is a new **R** package specifically dedicated to TL dating. This package is designed to be fully compatible with the existing *Luminescence* package and is user-friendly. It includes functions for TL data pretreatment and palaeodose estimation using the MAAD and the SAR protocols. The functionality of the *TLdating* package is evaluated using heated flints from Taibeh, Jordan.

1. Introduction

Thermoluminescence (TL) dating has been used since the end of the 1950s to date archaeological material (Houtermans et al., 1957; Tite and Waine, 1962). Although less commonly applied in sedimentary settings than optically stimulated luminescence (OSL) dating because of the presence of an unbleachable residual component (Aitken, 1985, 1998), there are some instances where TL dating remains the method of choice, e.g. for burnt flint (Mercier et al., 2003; Richter et al., 2009; Schmidt et al., 2013), ceramic (Blain et al., 2007; Veronese et al., 2008; Fraser and Price, 2013), and calcite dating (Polikreti et al., 2003; Stirling et al., 2012). It has been demonstrated that the *Single Aliquot Regenerative Dose* (SAR) protocol (Murray and Wintle, 2000), which is routinely used in OSL dating, is also applicable to TL (Fattahi and Stokes, 2000; Bassinet et al., 2006; Hong et al., 2006) and the use of red rather than blue TL provides promising results for burnt flint (Schmidt et al., 2015) and volcanic quartz (Tsukamoto et al., 2007), although this signal is not always present (Krbetschek et al., 1997). Moreover, TL has some general advantages compared to OSL. TL is not, or less, sensitive to light (Aitken, 1985), which is particularly interesting for archaeological samples excavated in bright sunlight. Furthermore, the TL signal is more directly linked to the physical properties of the sample. Each TL peak can be assigned to a specific luminescence trap, which is especially interesting for investigating the use of new materials for dating (Krbetschek et al., 1997). While recent developments demonstrate the relevance of TL dating, its application is still limited by a lack of user-optimized analysis software for TL data.

R is a programming language and environment for statistical computing and graphics. It provides a wide variety of statistical and graphical techniques and is highly versatile (R Core Team, 2016). In 2012, an **R** package especially designed for luminescence data was published (Kreutzer et al., 2012; Dietze et al., 2013; Fuchs et al., 2015). This package, called *Luminescence*, marked a step change in the possibility for statistical analysis, data treatment and plotting of luminescence data. Whereas previously much analysis was done using the established standalone software *Analyst* (Duller, 2015), the *Luminescence* package has enabled the incorporation of the latest statistical and plotting approaches shared by the luminescence dating community (Smedley, 2015; Dietze et al., 2016). In 2013, a second **R** package called *numOSL* was published (Peng et al., 2013). However, these packages focus almost solely on OSL. The *Luminescence* package includes only a few functions dedicated to TL dating. More generally, most software currently available for luminescence dating seems to neglect TL dating. Whilst *Analyst*

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http://dx.doi.org/10.1016/j.quageo.2016.09.001
1871-1014/

(which is bundled with the commonly used *Risø TL/OSL Readers*) still includes functionality to perform the analyses associated with the *Multiple Aliquot Additive Dose* (MAAD) protocol commonly used in TL dating (Aitken, 1985), researchers have to use their own code to perform more advanced analysis of TL data. To close this gap, we present a new **R** package called *TLdating*, which focuses exclusively on the analysis of TL data. It is fully compatible with the existing *Luminescence* package and has been designed to be as easy to use as possible. It is freely available for the community and offers the functionality to perform the entire dose calculation for TL samples, including the import of measurement data, data pretreatment, and palaeodose estimation with both the MAAD and SAR protocols.

2. *TLdating* package and R environment

One of the main differences between this package and the *Luminescence* package is the way the uncertainties are treated. Rather than estimating the uncertainty at the end of the dose estimation process, uncertainties are generated as soon as the data are imported into **R**. Indeed, TL dating requires the combination of different luminescence curves prior to any dose estimation.

Uncertainties on TL measurements can have various origins including purely statistical fluctuations in the number of photons detected by the PMT or fluctuation in the conditions under which the curves are measured (solid angle of light collection, sample orientation, thermal contact,etc.). These uncertainties can be categorised as systematic and random uncertainties. The systematic uncertainties can usually be discarded as long as an experiment uses the same reader with the same radiation source, the time required to take all necessary luminescence curves is not excessively long and the doses are expressed in terms of irradiation time and not in Gy. Therefore, this package considers all uncertainties as random. Errors are propagated in quadrature. Different hypotheses can be used for estimation of TL signal uncertainty. If signal counts obey a Poisson distribution, the initial standard uncertainties on the signal are the square root of the number of counts (Adamiec et al., 2012). When two signals, a and b, are added or subtracted, the resulting absolute uncertainty δs is calculated as $\delta s = \sqrt{(\delta a)^2 + (\delta b)^2}$. When a signal a is divided by a signal b, the resulting relative uncertainty $\delta s/s$ is calculated as $\frac{\delta s}{s} = \sqrt{\left(\frac{\delta a}{a}\right)^2 + \left(\frac{\delta b}{b}\right)^2}$. When a weighted average of the elements i from a signal x is made, $s = \frac{\sum_i (w_i . x_i)}{\sum_i w_i}$ and the resulting absolute uncertainty is: $\delta s = \frac{1}{\sqrt{\sum_i w_i}}$ where $w_i = \frac{1}{(\delta x_i)^2}$. This approach allows a more systematic and homogeneous uncertainty estimation when different steps of data pretreatment and measurement protocols are used and combined.

Another difference between the *Luminescence* and the *TLdating* packages is related to the use of some of the metadata accompanying the luminescence data. Both the *sequence editor* software, used with *Risø TL/OSL readers*, and the *Lexstudio* software, used with *Freiberg Instruments Lexsyg TL/OSL readers*, provide the results of luminescence measurements as a *.BINX* file. This file does not only contain the luminescence response data from the measurements but also a series of *metadata* containing the characteristics of these measurements comprising the irradiation time and sometimes the irradiation dose rate, the type of luminescence signal produced (e.g. TL, OSL, etc.), the heating rate and the maximum temperature, the presence of a temperature plateau and its duration, and the measurement *data-type*. It also includes a comment section that can contain extra information (Duller, 2015). When discs are analyzed using a single aliquot approach, the *data-type* is directly linked to the order of the curves in the measurement sequence. The first curve is always the *Natural*, the second its *Test Dose* response, and then a succession of *Regenerative Dose* and *Test Dose* responses follow. When several discs are combined in a multiple aliquot approach, this is no longer possible, especially if the palaeodose estimation and sub-linearity correction are included in the same sequence. The *data-type* of each measurement is therefore a key feature of the package and it is used by most of the functions, including those for the SAR protocol. The *data-type* has to be defined properly when writing the sequence (or afterwards with *Analyst*). The main existing *data-types* are *Natural*, *Natural + Dose*, *Dose*, *Background*, *Bleach* and *Bleach + Dose*. To make the analysis of luminescence data easier, the package introduces two new *data-types*: *Preheat* and *Testdose*.

The functions of the package can be separated into five categories: (1) the class objects that store the data (*TLum.**); (2) the conversion functions, identified by a 2 between two class names following the *Luminescence* package nomenclature; (3) the data manipulation functions (*mod_**); (4) the analysis functions (*analyse_**); and (5) the predefined scripts (*script_**) (Table 1). The easiest way to use the package is to call the *script_** functions, which gather a complete workflow to produce the desired results (Fig. 1). The *script_** functions also select the most relevant data for output. For more flexibility, users can create their own scripts using the *mod_** and *analyse_** functions. However, this generally requires more advanced knowledge of **R**.

The *mod_** functions are used for the modification of a *TLum.Analysis* object and create a new *TLum.Analysis* object that can be used for further analysis. All the pretreatment functions are *mod_** functions. The *analyse_** functions combine the curves of a *TLum.Analysis* object to extract information, producing a *TLum.Results* object rather than modifying the original *TLum.Analysis* object. The estimation of an equivalent dose (D_e) is performed using an *analyse_** function. The *TLdating* package also contains a series of *calc_** functions which are internal functions used by the *mod_** and *analyse_** functions. These functions can be used by advanced users who want to develop their own functions.

3. Thermoluminescence dating using the *TLdating* package

3.1. Data import and export

The *script_TL.import* function allows the import of TL data measured on standard *Risø* and *Lexsyg* readers in the form of *.BINX* files into **R**. The only mandatory parameter is the file name. The function will successively call the functions *read_BIN2R*, adopted from the *Luminescence* package (Kreutzer et al., 2012), *Risoe.BINfileData2TLum.BIN.file*, *TLum.BIN.File2TLum.Analysis*, *mod_extract.TL* and *mod_identify.dType*. The creation of the error matrix is fulfilled by *Risoe.BINfileData2TLum.BIN.file* during the conversion of the data created by *read_BIN2R*. This matrix contains an absolute random uncertainty for each data point from the original *Risoe.BINfileData* object. The function default setting is that the initial signal uncertainty is equal to the square root of the intensity. This default can be modified using a corrective factor, k, provided by the user (Adamiec et al., 2012). The function calls a *.BINX* file from the **R** *working directory* unless an alternative path is specified using *file.parameters*.

Once the data are imported, the script calls *mod_extract.TL*, which removes all the data that are not indicated as TL curves (Fig. 2), and *mod_identify.dType*, which updates the *data-type* information of the *TLum.Analysis* object. This function is required to generate the two new *data-types* introduced by the package. These *data-types* are specified using the *comment* metadata of the *.BINX* file. When the data are imported into **R**, this metadata is scanned

Table 1
Summary of the main functions of the *TLdating* package. (1) Class objects containing the data; (2) functions which convert a class object into another class object; (3) functions that manipulate the data; (4) functions that do the data analysis; and (5) scripts that gather a predefined set of functions. For a list of required input arguments see online supplement (Table A.1).

	Functions	Description
(1)	*TLum.BIN.file*	Object containing the information present in *.BINX* file
	TLum.Data.Curve	Objet gathering the information linked to a record
	TLum.Analysis	List of *TLum.Data.Curve*.
	TL.Results	Results provided by *Analyse_** function
(2)	*Risoe.BINfileData2TLum.BIN.file*	Converts *Risoe.BINfileData* object into *TLum.BIN.file* object
	Tlum.BIN.file2Risoe.BINfileData	Converts *TLum.BIN.file* object into *Risoe.BINfileData* object
	Tlum.BIN.file2Tlum.Analysis	Converts *Tlum.BIN.file* object into *TLum.Analysis* object
	RLum.Analysis2TLum.Analysis	Converts *RLum.Analysis* object into *TLum.Analysis* object
(3)	*mod_extract.TL*	Extracts TL curve from *TLum.Analysis*
	mod_identify.dType	Updates *data-type* metadata of *TLum.Analysis*
	mod_remove.aliquot	Removes specified aliquots from *TLum.Analysis*
	mod_remove.preheat	Removes preheat curves from *TLum.Analysis*
	mod_subtract.background	Subtracts background from associated TL curve in *TLum.Analysis*
	mod_align.peaks	Aligns TL peaks from *TLum.Analysis*
(4)	*analyse_TL.plateau*	Performs the heating plateau test for *TLum.Analysis* object
	analyse_TL.MAAD	Estimates D_e using the MAAD protocol for *TLum.Analysis* object
	analyse_TL.SAR	Estimates D_e using the SAR protocol for *TLum.Analysis* object
(5)	*script_TL.import*	Creates *TLum.Analysis* from *.BINX* file
	script_TL.export	Creates *.BINX* file from *TLum.Analysis*
	script_TL.pretreatment	Gathers series of data pretreatments
	script_TL.plateau	Gathers heating plateau test estimation and series of data pretreatments
	script_TL.MAAD	Gathers complete workflow to estimate D_e using MAAD protocol
	script_TL.SAR	Gathers complete workflow to estimate D_e using SAR protocol

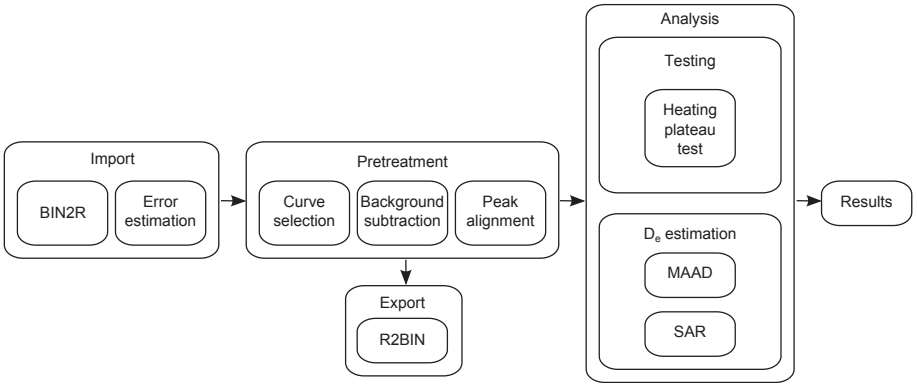

Fig. 1. Workflow followed by the different *script_** functions of the *TLdating* package. First the TL curves are imported into **R**, then they are submitted to a series of pretreatment to prepare the upcoming analysis. Finally, the modified curves can be exported, for use in a different package or software, or they can be combined in **R** to realize the desired analysis.

and the *data-type* section of the *TLum.Analysis* object is automatically updated when it contains the words *Preheat* or *Testdose*. At the same time, the old *data-type* is saved separately for backward compatibility.

The *script_TL.export* function allows the export of TL data stored in a *TLum.Analysis* object as a standard .BINX file. The only mandatory parameter is the file name. The function will successively call the functions *TLum.Analysis2TLum.BIN.File*, *TLum.BIN.File2Risoe.BINfileData* and *write_R2BIN*, from the *Luminescence* package. During the process, the original *data-type* of each curve is restored to avoid a compatibility problem with *Analyst*.

3.2. Pretreatment of TL data

Due to thermal effects, it is generally not possible to directly use measured TL curves to estimate a D_e. A series of data pretreatments are required. *Script_TL.pretreatment* gathers a series of the most often used pretreatments: (1) the removal of selected aliquots; (2) the removal of the preheat curves; (3) the subtraction of the background signal; and (4) the alignment of the main TL peak. To make its use easier, the script directly calls *script_TL.import*. Therefore, the only input parameter which is required is the file name. Although the function can automatically detect the TL peak used for the D_e estimation, specifying an initial temperature interval including the TL peak maximum can improve the analysis, especially in case of TL curves with multiple peaks or a strong black body radiation signal. This can be done using the user defined *aligning.parameters* option.

First, the script calls *mod_remove.aliquot*, which allows the removal of specified aliquots. These aliquots are generally identified after a first estimation of the D_e and may not fulfil the selection criteria or be outliers. Secondly, it calls *mod_remove.preheat*. This function removes the preheat curves from the data object (Fig. 3a, b). It relies on their prior identification in the *data-type* metadata to work properly. Afterwards, *mod_subtract.background* subtracts the background curve associated with each of the TL curves (Fig. 3c). Once again, it relies on a prior identification of the background curves in the *data-type* metadata to work properly. Finally, *mod_align.peaks* is called. This last function will align the main peak of each luminescence curve. It realizes a polynomial smoothing of the curves, using *smooth.spline* from the *stats* package (R Core Team, 2016), to identify the position of the maximum in the interval

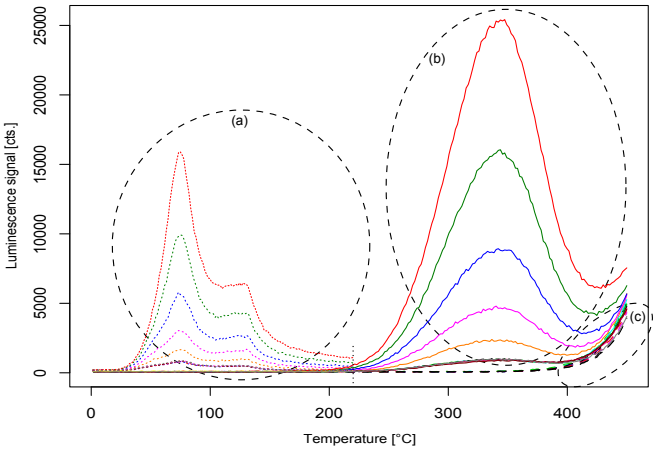

Fig. 2. Selection of TL curves. As this package focuses exclusively on TL, all non-TL data are automatically removed from the *.BINX* file. In this figure, it is possible to identify (a) preheat curves (dotted lines), (b) L_x and T_x curves (solid lines) and (c) background curves (dashed lines). The aliquot is composed of 150–200 μm quartz grains, extracted from sample NS2 (flint) that were bleached at 360 °C for 2 h and then irradiated with regenerative doses ranging from 80 to 1200 Gy.

specified by the user. If the protocol includes a test dose response, it uses the average position of their maxima as the alignment reference point, otherwise it uses the average position of all the maxima. Once the alignment reference point has been defined, it shifts the original TL curves to align their maxima with the reference point (Fig. 3d).

Once all the pretreatment has been completed, the script calls *script_TL.export* to save the results into a new *.BINX* file. This allows *Analyst*, or any other compatible software, to be used to visualize the results or estimate the D_e separately.

3.3. Identification of sufficiently heated objects using the heating plateau test

The heating plateau test was proposed to verify if a TL signal of a ceramic sample has been properly reset throughout heating (Aitken, 1985). Initially designed for the MAAD protocol, the test consists of dividing the signal emitted from a natural sample (N) by the signal emitted from a natural sample that has received an additive dose ($N + \beta$). If the natural sample has not been fully reset by heating, the shape of the two curves will be different. If the sample was completely reset, the shape of the curves should be the same and the signals proportional. Therefore $N/(N + \beta)$ should present a plateau in the region of the peak maximum (Fig. 4). This plateau defines the region suitable for D_e estimation. Although the heating plateau forms a robust test, second order kinetics may exhibit an effect similar to partial resetting of the signal (Grün, 1994). Furthermore, if the natural signal is close to saturation, $N/(N + \beta)$ will tend towards 1, preceding any inference of partial resetting.

A similar approach can be used when applying the SAR protocol. If the signal has been properly reset and presents no second order kinetic effects, N/β should form a plateau. Once again, this plateau defines the region that could be used for the D_e estimation. It must be noted that for regenerative doses smaller than the natural dose the ratio will be greater than 1, which is impossible when using additive doses.

When the natural, additive and regenerative dose responses are followed by test dose responses, these can be used to normalize the results. This may be necessary when the aliquots have heterogeneous luminescence properties or when successive irradiations modify their sensitivity.

script_TL.plateau first calls *script_TL.pretreatment* to prepare the TL data. Once again, only the name of the *.BINX* file is required. It then calls *analyse_TL.plateau*. For each temperature channel L_n/L_x is estimated, where L_n is the average natural signal and L_x is the average signal obtained for each additive or regenerative dose. When the dose responses are followed by test dose responses, T_x, the function will also estimate $(L_n/T_n)/(L_x/T_x)$ for each temperature channel (Fig. 5).

It has to be noted that having a heating plateau does not guarantee the accuracy of the final D_e. However, when a sample does not fulfil this test, it is a strong indication that it will not provide any meaningful D_e.

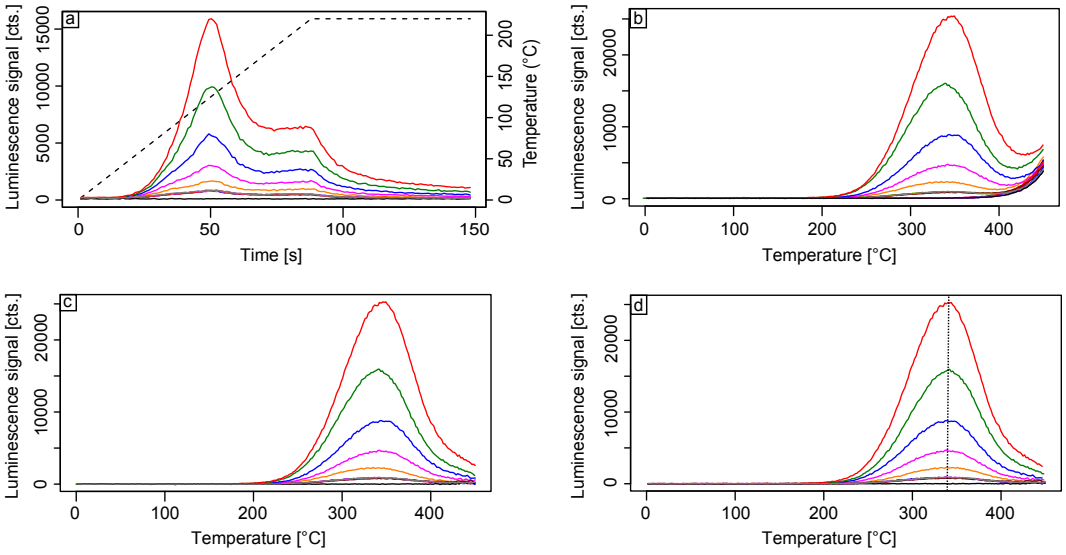

Fig. 3. Pretreatment of the TL data: Separation of (a) the preheat signals from (b) the signals used for dating. In (a), the solid lines are the preheat signals and the dashed line is the temperature. (c) Creation of the L_x and T_x signal that will be used for dating by subtracting the corresponding background signal. (d) Alignment of the TL peaks of the L_x and T_x signals. The aliquot is composed of 150–200 μm quartz grains, extracted from sample NS2 (flint) that were bleached at 360 °C for 2 h, and then irradiated with regenerative doses ranging from 80 to 1200 Gy.

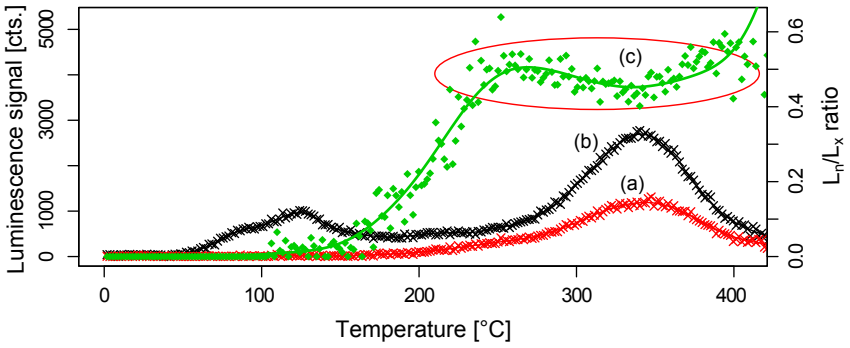

Fig. 4. Heating plateau for quartz extracted from sample NS2 (flint). When a natural signal L_n (a) is divided by an additive dose signal L_x (b), the L_n/L_x ratio should present a plateau around the maximum peak position (c). The *natural* aliquot is composed of 150–200 μm quartz grains that were bleached at 360 °C for 2 h, then irradiated with 80 Gy and preheated at 180 °C for 3 min. The *additive dose* aliquot was prepared as described for the *natural* aliquot and then irradiated with an additive dose of 80 Gy.

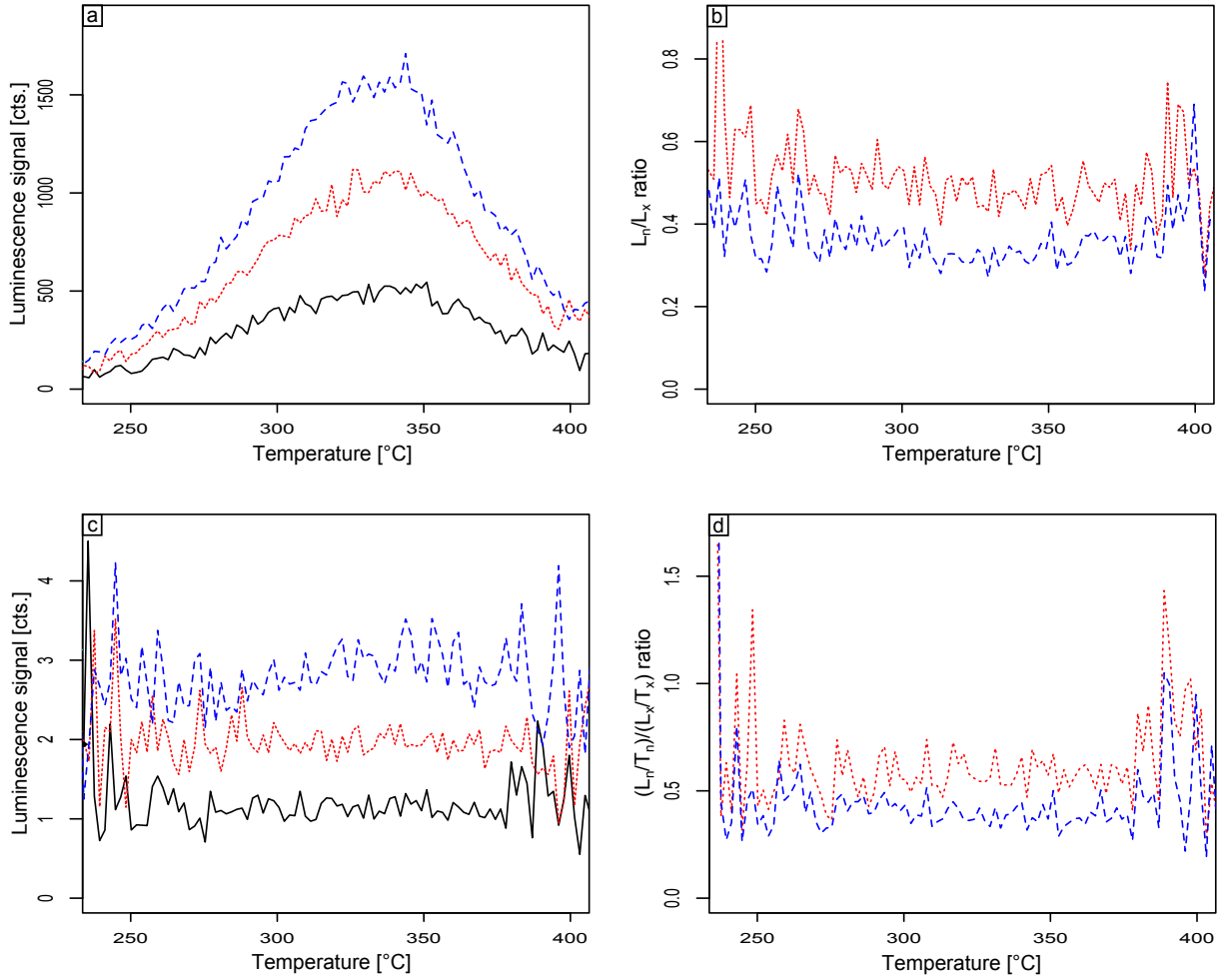

Fig. 5. Heating plateau test for (a) two additive dose TL signals (L_x) and (c) the corresponding sensitivity corrected signals (L_x/T_x). In both cases, a heating plateau can be observed between 290 and 360 °C, i.e. around the position of the TL peak (b, d). The *natural* aliquot is composed of 150–200 μm quartz grains from sample NS2 (flint) that were bleached at 360 °C for 2 h, then irradiated with 40 Gy. The *additive dose* aliquots were prepared as described for the *natural* aliquot and then irradiated with additive doses of 30 and 60 Gy.

3.4. Equivalent dose calculation from TL data

3.4.1. The growth curve and dose plateau approach

In TL dating, the usual approach for extracting luminescence signals from TL curves is to integrate the signal over a given temperature interval which is centered on the luminescence peak maximum (Fig. 6 a, b). As previously seen, an appropriate interval can be estimated using the heating plateau test. These integrals define the luminescence response for each of the measured additive and regenerative doses. A single regression curve then allows the determination of the D_e; this approach is called the growth curve (GC) approach (Wintle, 1997).

Another approach is to define a D_e value for each data channel or temperature interval. This second approach generally requires the

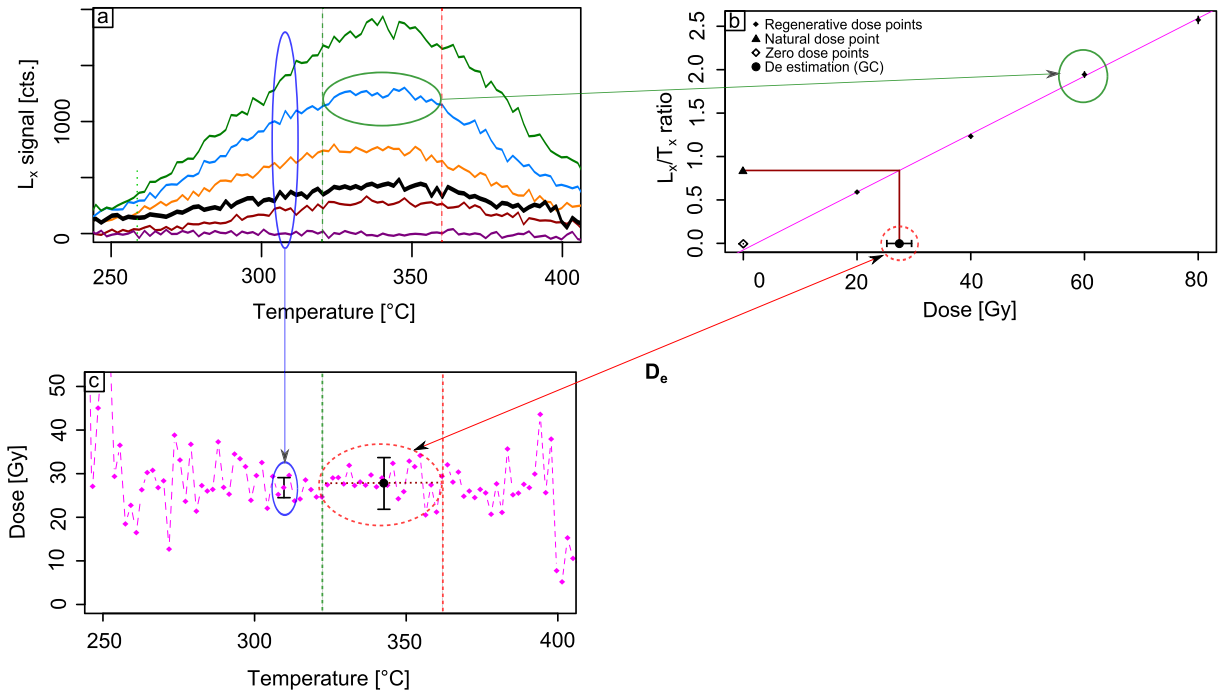

Fig. 6. Comparison between the growth curve and the dose plateau approach. In both cases, the same TL data are used (a); in the GC approach the response for each additive/regenerative dose is averaged over the heating plateau before estimating the D_e (b); in the DP approach, a D_e is estimated for each temperature interval before averaging the results (c). This aliquot is composed of 150–200 μm quartz grains from sample NS2 (flint) that were bleached at 360 °C for 2 h, then irradiated with 30 Gy to simulate the natural dose. The aliquot was then irradiated with increasing *regenerative dose* from 20 to 80 Gy.

realization of many more regression curves. Once we have a D_e for each temperature interval, we can average the results over the same temperature interval as used in the previous approach (Fig. 6a, c). This approach is called the dose plateau (DP) approach (Wintle, 1997).

As previously shown for the heating plateau test, the signal is proportional to the absorbed dose if it does not saturate. Therefore, a D_e plateau should form around the peak maximum position. The plateau obtained with the DP approach is generally shorter than the one obtained with the heating plateau test. Therefore, it is generally a better indicator for defining the temperature interval for the D_e estimation.

3.4.2. D_e calculation with the MAAD protocol

Since the development of SAR protocols, the MAAD protocol remains widely used only for TL dating (Zöller et al., 2014). The luminescence signal can be considered as linear before starting to saturate and is relatively well described by an exponential function with a linear term. However, for low doses, this signal also exhibits non-linear behavior that must be corrected for. The dose received by the sample after the last resetting event is called the palaeodose, *P*. Its best estimation is usually called the equivalent dose, D_e.

When using the MAAD protocol, the D_e can be separated into two components, the *extrapolated* equivalent dose (Q) and the supralinear correction (I) (Fig. 7). Q is estimated using a series of natural aliquots irradiated with different *additive* doses. A luminescence response is evaluated for each of these *additive* doses and a regression function enables determination of Q. I is estimated using bleached aliquots irradiated by different *regenerative* doses. Once again, the luminescence response is evaluated for each of these *regenerative* doses and a regression function can be used to determine I. The final D_e is the sum of Q and I, both of which produce an uncertainty which is propagated in quadrature on the D_e. For TL, in most cases, the regression function is a simple linear fit.

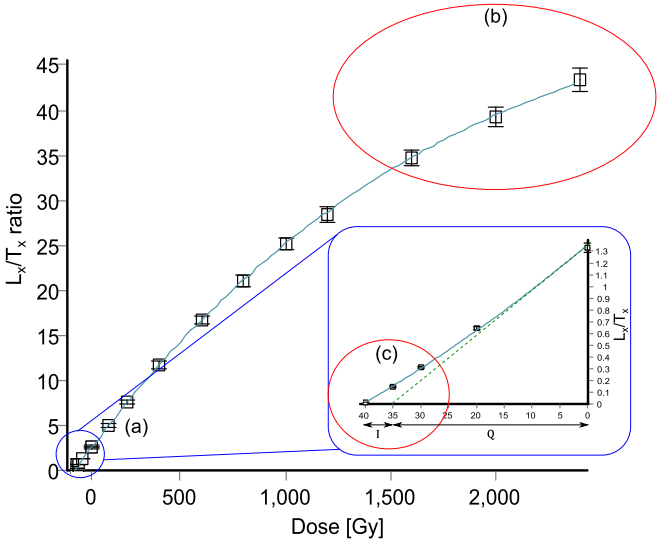

Fig. 7. The growth curve can be separated into 3 parts (a) the linear region, (b) the saturation region and (c) the supralinear region. When using the MAAD protocol, the linear approximation (Q) does not take into account this supralinearity. Therefore, a supralinear correction (I) has to be added to obtain a final D_e which reflects the real palaeodose (P). This aliquot is composed of 150–200 μm quartz grains, extracted from sample NS2 (flint) that were bleached at 360 °C for 2 h and then irradiated with doses ranging from 0 to 2400 Gy.

Indeed, the MAAD protocol is not applicable beyond the linear dose region.

It is possible to estimate a *test dose* response after each *additive* and *regenerative* dose. Since the MAAD protocol combines signals from different discs, this *test dose* does not allow sample sensitivity change to be tracked. However, it does allow normalization of the

results, especially when the aliquots are heterogeneous.

The initial MAAD protocol requires that the samples rest for several weeks before estimation of the dose response; this enables thermally unstable signal components to decay (Aitken, 1985). However, it has been shown that this step can be replaced by preheating at lower temperature, as done in SAR measurement protocols (Dutkiewicz and Prescott, 1997; Feathers, 1997). It allows a more routine application of the MAAD protocol since the aliquots do not have to be removed from the luminescence reader in the middle of the procedure.

The *script_TL.MAAD* function calculates an estimation of the D_e using both the DP and the GC approaches. In theory, both approaches should provide similar results. Once again, it starts by calling *script_TL.pretreatment* to prepare the curves for the D_e estimation. Then it calls *analyse_TL.MAAD*. This function requires two extra parameters: *eval.Tmin* and *eval.Tmax*, which are the temperature boundaries for signal integration. A series of complementary parameters enable improvement of the fitting: (1) the type of fitting, at present only linear fitting is supported; (2) whether the fitting should be weighted; (3) the additive dose interval used for the estimation of *Q*, which is required when the highest dose signals are in saturation or when the natural signal is in the supralinear region; (4) the regenerative dose interval used for *I* estimation, which allows exclusion of the regenerative dose signals in the supralinear region; and (5) if the slope defined for *Q* should be reused to estimate *I*. The method relies on the hypothesis that all the aliquots should have the same dose response curve (Aitken, 1985), i.e the *additive* and *regenerative* growth curves must have the same slope (Fig. 8). Therefore, option (5) will produce more coherent results. Another solution would have been to combine all the aliquots to estimate the slope of the growth curve (Prescott et al., 1993). However this approach has not been implemented as it can mask differences of slope when heating induced sensitivity changes occurs.

The function starts by identifying the dose responses (L_x) and the *test dose* response (T_x) to calculate L_x/T_x. The average L_x and T_x for each additive/regenerative dose is estimated before plotting the heating plateau test of all the resulting curves. Finally, the function estimates the D_e using both the DP and GC approaches (Fig. 9). When the measurement protocol does not include *test doses*, the function considers that $T_x = 1$ and $L_x/T_x = L_x$. When the measurement protocol does not include *regenerative* doses, only *Q* is estimated. When the measurement protocol does not include *additive* doses, only *I* is estimated.

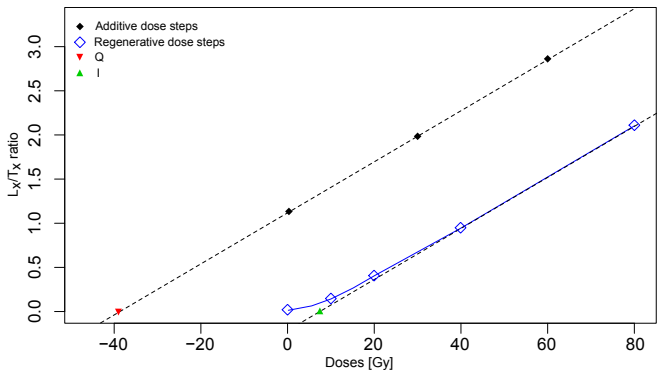

Fig. 8. When there is no sensitivity change during the measurement, the additive and the regenerative dose response curve slopes are identical. In this example, the first three regenerative dose responses are above the regression line and should not be used to determine the supralinear correction. The 18 aliquots are composed of 150–200 μm quartz grains, extracted from sample NS2 (flint) that were bleached at 360 °C for 2 h. Six aliquots were irradiated with 40 Gy to simulate the natural dose and four of them received extra *additive doses* of 30 and 60 Gy. Eight further aliquots received *regenerative doses* from 10 to 80 Gy.

3.4.3. D_e calculation with the SAR protocol

Initially developed for OSL dating, the SAR protocol can also be applied in TL dating (Richter and Krbetschek, 2006; Bluszcz and Bøtter-Jensen, 1995). This protocol has the advantage of requiring less sample material and allows D_e estimation for samples close to saturation, using non-linear fit. Moreover, the supralinear correction is no longer required. For this protocol, the D_e is estimated by comparing the natural signal L_n to the dose response curve created by irradiating the aliquot with a series of *regenerative* doses L_x. However, the successive heatings that are required to reset the material between each *regenerative* dose may influence its sensitivity. This sensitivity change can be partially corrected for by the monitoring of a *test dose* response T_x after each *regenerative* dose irradiation. However, common to most SAR protocols, the initial sensitivity change between measurement of the *natural* and the first *regenerative* dose cannot be detected (Wintle and Murray, 2006). Modified SAR protocols were developed to overcome this problem, for example the Natural Sensitivity Corrected-SAR (NSC-SAR) which uses the 110 °C peak present in the preheat signal when the sample is submitted to a pre-dose before the recording of L_n (Singhvi et al., 2011), but they are not yet included in the *TLdating* package.

Since we estimate a D_e for each aliquot, this method is far less sensitive to the heterogeneous luminescence behaviour between aliquots. The final D_e is estimated by averaging the D_e obtained for each aliquot.

Like *script_TL.MAAD*, *script_TL.SAR* starts with calling *script_TL.pretreatment*. Once again, two parameters, *eval.Tmin* and *eval.Tmax*, are required to define the temperature boundary between which the signal will be integrated. A series of other parameters enables refinement of the final result: (1) fitting type, although at present only a linear fit has been implemented; (2) whether the fitting should be weighted; and (3) dose interval used to define the fitting. As for the MAAD protocol, the function starts by separating L_x and T_x and calculating L_x/T_x for each regenerative curve. It then realizes the heating plateau test for all of the curves before estimating the D_e using both the GC and the DP approach (Fig. 10).

4. Discussion

The package was used to estimate the D_e values of a series of burnt flint samples from the palaeolithic site of Taibeh, Jordan. These artefacts are associated with the Masraqan technocomplex, currently dated between 25 and 20 ka BC (MIS2) (Belfer-Cohen and Goring-Morris, 2007). All the aliquots were measured on a *Risø* TL/OSL-DA-20 reader equipped with the optical filters BG39, GG400, 7–59 and HA3. The results obtained with **R** were compared with the results obtained with *Analyst 4.14.6* before and after using the pretreatment script.

For the results obtained with the *TLdating* package, we used a weighted linear fitting. To avoid the supralinear region, we removed the first *regenerative* dose points from the fitting, i.e *regenerative* doses below 16 Gy. For the MAAD protocol, we reuse the slope obtained when estimating *Q* to determine *I*. The same aliquots were used for estimation of *Q* and *I*. To calculate a D_e value using the SAR protocol in *Analyst* it is necessary to provide a channel interval for the background estimation, even if it has already been subtracted from the signal. We decided to use the first channels of the TL peaks (channel 120 to channel 125 of 250, i.e from 216 to 225 °C), to provide background signals as low as possible whist ensuring that values remain positive.

For the SAR protocol, the **R** package provides results very similar

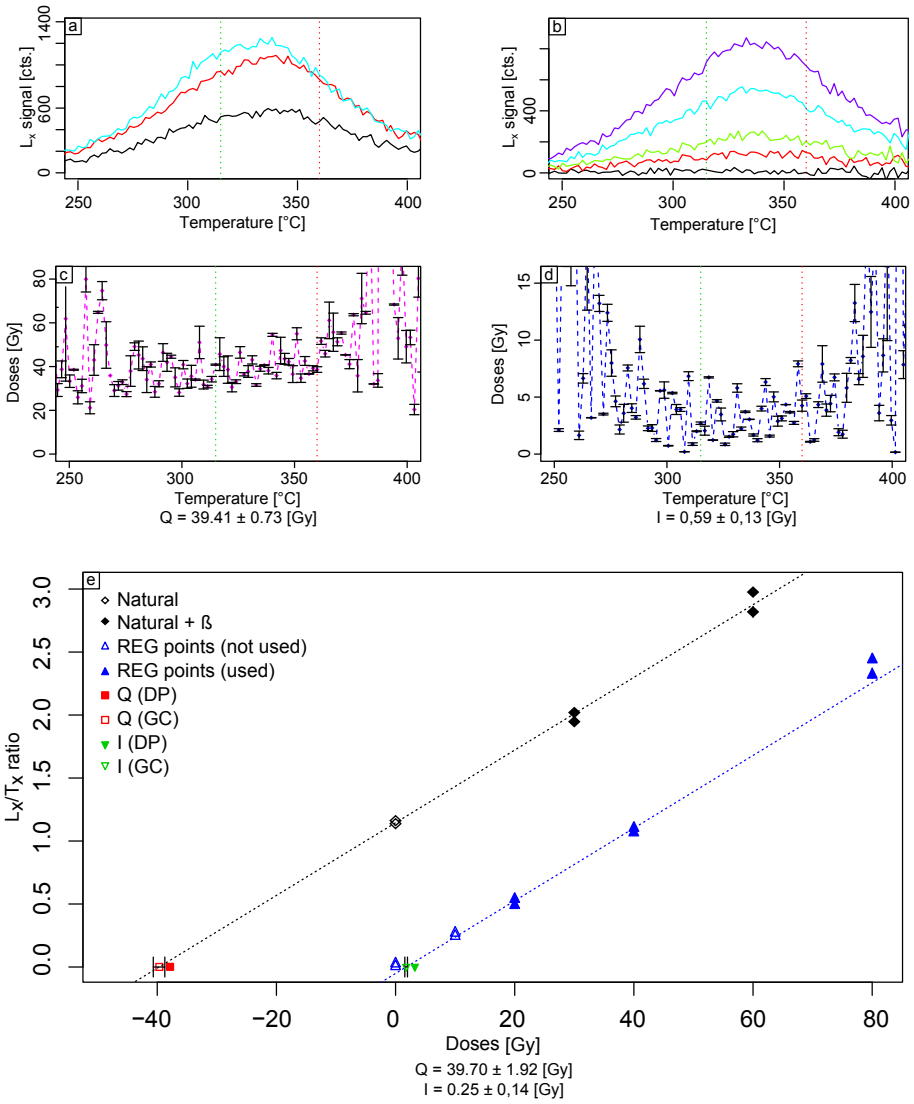

Fig. 9. Estimation of the D_e with the MAAD protocol using the *TLdating* package. Two aliquots were irradiated with *additive doses* of 0, 30 and 60 Gy (a). Their test dose response signals allow correction for the very low signal emitted by one of the 60 Gy aliquots. Five regenerative dose steps were used (from 0 up to 80 Gy) to identify the supralinear effect (b). Both the DP (c, d) and the GC (e) approaches provide very similar results, around 40 Gy D_e values below 20 Gy are very sensitive to variations in the supralinear correction.

to those obtained with *Analyst*. However, for the MAAD protocol, the results are sometimes very different (Fig. 11a). Within the **R** package, the difference between the SAR-TL and the MAAD-TL is generally below 5%; both protocols provide similar results (Fig. 11b). The only notable exception is the sample TAI 20. This sample presents all the characteristics of a flint that was heated at lower temperature or during a shorter period. The significantly lower D_e obtained using the SAR protocol can be explained by a sensitivity change which occurs after the recording of the natural signal. The difference between the GC and the DP results are generally below 2%. However, within *Analyst*, the difference between the SAR-TL and the MAAD-TL can be above 33%. These differences can be easily explained by the heterogeneous luminescence properties of the aliquots, each of them containing only a small amount of material. The use of a test dose allows the **R** package to partially correct for this heterogeneity. This feature is not supported by *Analyst* where problematic samples can be easily identified by the user but have to be removed manually.

One of the main strengths of the *TLdating* package is the ability to easily modify the fitting parameters. We compared results using different fitting parameter combinations: (1) unweighted average using all dose responses and an independent *I* estimation; (2) unweighted average using all dose responses but reusing the *Q* slope for *I* estimation; (3) weighted average using all dose responses and an independent *I* estimation; (4) weighted average using all dose responses but reusing the *Q* slope for *I* estimation; (5) unweighted average using only the higher dose responses and independent *I* estimation; (6) unweighted average using only the higher dose responses but reusing the *Q* slope for *I* estimation; and (7) weighted average using only the higher dose responses but with an independent *I* estimation (for the complete dataset see online supplement, Table A.3).

For these samples, the SAR protocol provides relatively similar results despite the parameter changes. However, using a weighted average systematically underestimates the high dose values when all of the dose responses are taken into account. It is not a problem for low D_e values, as presented in the example data, but it can become problematic for higher D_e values with the deviation being directly proportional to dose.

For the MAAD protocol, using all the dose response values

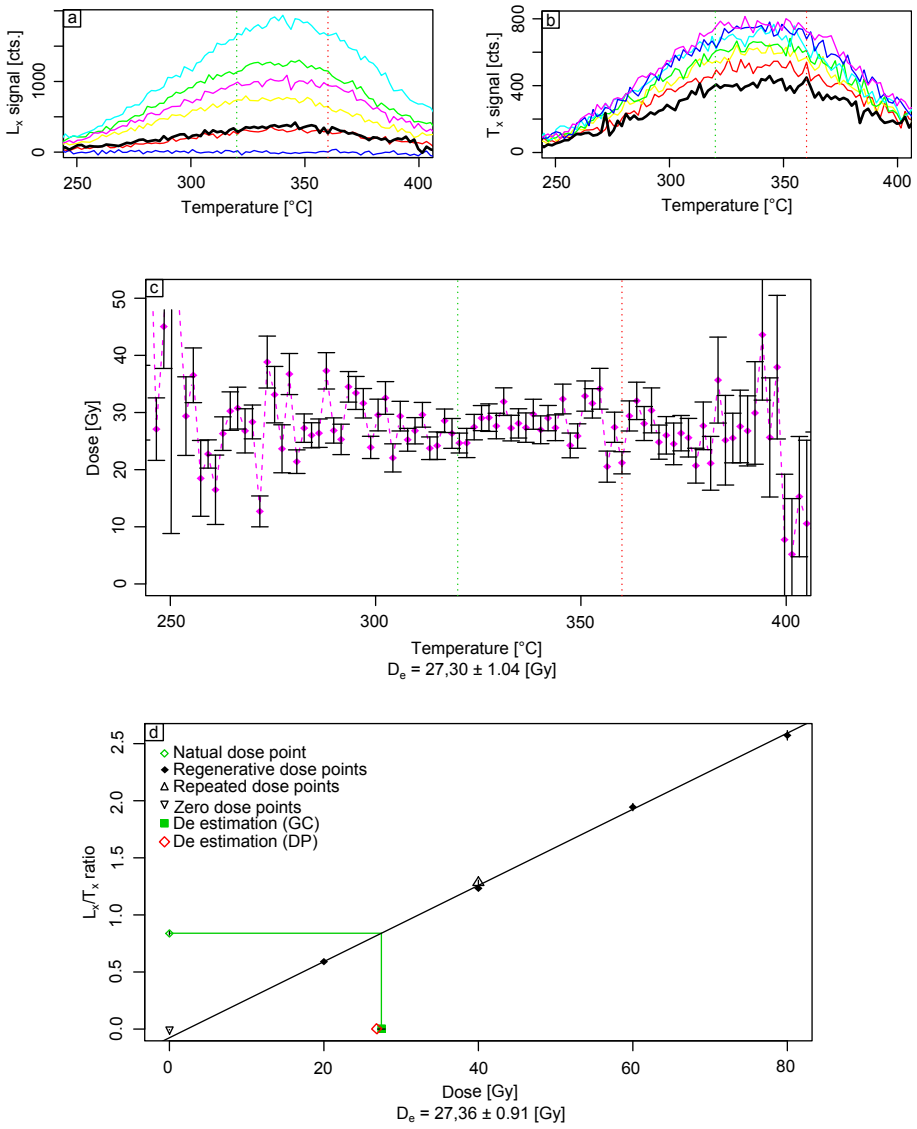

Fig. 10. Estimation of the D_e with the SAR protocol for one aliquot using the *TLdating* package. Six regenerative doses, including a zero dose and a repeated dose point, were administered (a). The test dose responses show a sensitisation of the material with increasing doses (b). The dose plateau (c) and growth curve approach (d) provide a similar D_e estimation, around 27 Gy. The DP approach allows an easy assessment of the temperature interval for D_e estimation whereas the GC approach allows an easy assessment of the regenerative dose interval which avoids the supralinear and the saturation regions.

systematically underestimates the D_e. Indeed, most of the *regenerative* doses are still in the supralinear region. Moreover, the zero dose response has the lowest absolute uncertainty and, therefore, the greatest weight, which forces the linear regression to pass near zero and annihilates the supralinear correction. The difference between the DP and GC results are maximized when the Q slopes are not reused for estimation of I. Indeed, once again, when averaging I in the DP approach, the lowest values are overweighted which induces an underestimation of I. This effect is not as strong within the GC approach which therefore provides a more robust D_e.

Finally, it seems that combining the three parameters provides the most robust D_e estimation. The limitation of the dose interval ensures that all the doses used are within or near the linear region. The weighting of the fitting ensures a better mathematical coherence throughout the complete process and finally the reuse of the Q slope smooths the results obtained for the I estimation, especially in the DP approach. The order of the operations should not have an impact on the results. Therefore, a divergence between the DP and the GC approach results is a good indication of problematic samples or parameter selection.

For the Taibeh samples, the SAR protocol seems to provide more robust results despite potential sensitivity changes. It is probably linked to the properties and heterogeneity of the aliquots. The main exception is the TAI 20 sample for which the D_e obtained using the MAAD protocol is more consistent with the D_e obtained for the other samples. This can be explained by a significant sensitivity change between the natural doses and the regenerative doses due to a laboratory heating higher than the archaeological resetting event. It must be noted that the reuse of the same aliquots to estimate I could also introduce an unwanted sensitivity change. Using different aliquots for the Q and I estimation is therefore recommended.

5. Conclusions

The new **R** package allows the estimation of a D_e from TL data with both the MAAD and the SAR protocols. It provides a series of

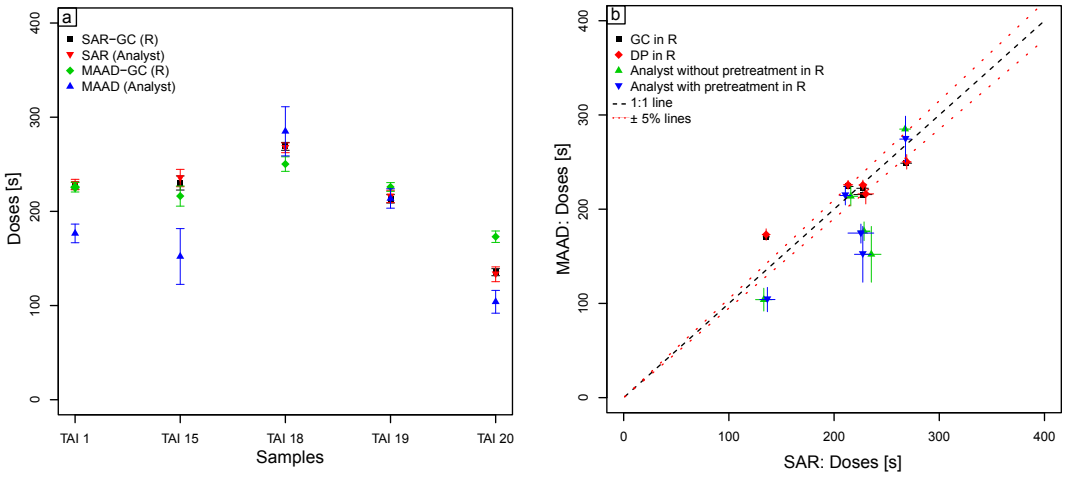

Fig. 11. D_e estimation for burnt flint from Taibeh, Jordan. (a) Estimation of the D_e for 5 samples (TAI 1, TAI 15, TAI 18, TAI 19, TAI 20) using different protocols (SAR & MAAD), and software (**R** & *Analyst*); (b) Comparison of the results of the SAR and MAAD protocols using the GC and the DP approach in **R** and using *Analyst* with and without a pretreatment in **R**. For the complete dataset see online supplement (Table A.2).

tools to pre-process the data but also to evaluate the validity of the results.

The package relies on an extended documentation of the sequence, especially the dose and *data-type* of all luminescence curves. Moreover, extra information, saved in the *comment* metadata, are required. However, well-documented .BINX files facilitate easy identification of the protocol used and of the *test dose*, *zero dose* and *repeated dose* curves, which is especially important for unconventional protocols.

The *script_** functions make the package easy to use for people without programming experience. However, advanced users can still utilize the specific functions to implement their own scripts which makes the package versatile.

Future development of the package will comprise: (1) implementing more functions to manipulate the .BINX file, especially the *data-type* metadata, and merging different .BINX files; (2) implementing more fitting types, especially exponential and exponential plus linear fitting; (3) supporting the *Single Aliquot Regenerated and Added Dose* (SARA) protocol (Mejdahl and Botter-Jensen, 1994); and (4) implementing functions for isothermal decay data analysis, once again for MAAD, SAR and SARA measurement protocols.

The *TLdating* package is available on GitHub (https://github.com/dstreble/TLdating) and on CRAN (http://cran.r-project.org/package=TLdating).

Acknowledgements

This project is realized in the context of the CRC 806 “Our Way to Europe” (http://www.sfb806.uni-koeln.de/) which is funded by the German Research Foundation (DFG). We would like to thank Prof. Dr. Jürgen Richter, Cologne, and Shumon T. Hussain, Cologne, for assistance with sampling the Taibeh site; Dr. Christoph Schmidt, Bayreuth, who collected the samples; Dr. Anja Zander, Cologne, for having been our first beta tester; Dr. Sebastian Kreutzer, Bordeaux, from the **R** *Luminescence* package, for helpful advice and comments; and Dr. Georgina King, Cologne, for numerous corrections of the manuscript.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.quageo.2016.09.001.

References

Adamiec, G., Heer, A.J., Bluszcz, A., 2012. Statistics of count numbers from a photomultiplier tube and its implications for error estimation. Radiat. Meas. 47 (9), 746–751. Proceedings of the 13th International Conference on Luminescence and Electron Spin Resonance Dating, 10-14 July, 2011, Torun, Poland. http://www.sciencedirect.com/science/article/pii/S1350448711005750.

Aitken, M., 1985. Thermoluminescence Dating. Academic Press, Oxford.

Aitken, M., 1998. An Introduction to Optical Dating. The Dating of Quaternary Sediments by the Use of Photon-stimulated Luminescence. Oxford University Press, Oxford.

Bassinet, C., Mercier, N., Miallier, D., Pilleyre, T., Sanzelle, S., Valladas, H., 2006. Thermoluminescence of heated quartz grains: intercomparisons between SAR and multiple-aliquot additive dose techniques. Radiat. Meas. 41 (7–8), 803–808. http://www.sciencedirect.com/science/article/pii/S1350448706000333.

Belfer-Cohen, A., Goring-Morris, A., 2007. Rethinking the Human Revolution. McDonald Institute for Archaeological Research, Cambridge, Ch. From the Beginning: Levantine Upper Paleolithic Cultural Change and Chronology, pp. 199–206.

Blain, S., Guibert, P., Bouvier, A., Vieillevigne, E., Bechtel, F., Sapin, C., Baylè, M., 2007. Tl-dating applied to building archaeology: the case of the medieval church notre-dame-sous-terre (mont-saint-michel, France). Radiat. Meas. 42 (9), 1483–1491. http://www.sciencedirect.com/science/article/pii/S1350448707003149.

Bluszcz, A., Bøtter-Jensen, L., 1995. Dosimetric properties of natural quartz grains extracted from fired materials. Radiat. Meas. 24 (4), 465–468. Proceedings of the International Symposium on Luminescent Detectors and Transformers of Ionizing Radiation. http://www.sciencedirect.com/science/article/pii/135044879500005Y.

Dietze, M., Kreutzer, S., Burow, C., Fuchs, M.C., Fischer, M., Schmidt, C., 2016. The abanico plot: visualising chronometric data with individual standard errors. Quat. Geochronol. 31, 12–18. URL http://www.sciencedirect.com/science/article/pii/S1871101415300583.

Dietze, M., Kreutzer, S., Fuchs, M.C., Burow, C., Fischer, M., Schmidt, C., June 2013. A practical guide to the R package luminescence. Anc. TL 31 (1), 11–18. http://www.aberystwyth.ac.uk/en/media/departmental/iges/ancienttl/pdf/vol31no1/dietze%5fatl13(1)%5f11.pdf.

Duller, G., 2015. The Analyst software package for luminescence data: overview and recent improvements. Anc. TL 33 (1), 35–42. http://www.ecu.edu/cs-cas/physics/Ancient-Timeline/upload/ATL_33-1_Duller_p35-42.pdf.

Dutkiewicz, A., Prescott, J., 1997. Thermoluminescence ages and paleoclimate from the Lake-Malata Lake-greenly complex, Eyre Peninsula, South Australia. Quat. Sci. Rev. 16 (3–5), 367–385.

Fattahi, M., Stokes, S., 2000. Extending the time range of luminescence dating using red TL (RTL) from volcanic quartz. Radiat. Meas. 32 (5–6), 479–485. http://www.sciencedirect.com/science/article/pii/S1350448700001050.

Feathers, J., 1997. Luminescence dating of sediment samples from White paintings Rockshelter, Botswana. Quat. Sci. Rev. 16 (3–5), 321–331. http://www.sciencedirect.com/science/article/pii/S0277379196000832.

Fraser, J.A., Price, D.M., 2013. A thermoluminescence (TL) analysis of ceramics from cairns in Jordan: using TL to integrate off-site features into regional chronologies. Appl. Clay Sci. 82, 24–30. Ancient Ceramics - Analysis and Components. http://www.sciencedirect.com/science/article/pii/S0169131713001804.

Fuchs, M.C., Kreutzer, S., Burow, C., Dietze, M., Fischer, M., Schmidt, C., Fuchs, M.,

2015. Data processing in luminescence dating analysis: an exemplary workflow using the r package 'luminescence'. Quat. Int. 362, 8–13, 9th New World Luminescence Dating Workshop. http://www.sciencedirect.com/science/article/pii/S104061821400408X.

Grün, R., 1994. The kinetics of TL peaks and its effect on the dose versus temperature plot. Radiat. Meas. 23 (1), 175–194. http://www.sciencedirect.com/science/article/pii/1350448794900345.

Hong, D., Kim, M., Choi, J., El-Faramawy, N., Göksu, H., 2006. Equivalent dose determination of single aliquot regenerative-dose (SAR) protocol using thermoluminescence on heated quartz. Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. Atoms 243 (1), 174–178. http://www.sciencedirect.com/science/article/pii/S0168583X05014151.

Houtermans, F.G., Jäger, E., Schön, M., Stauffer, H., 1957. Messungen der Thermolumineszenz als Mittel zur Untersuchung der thermischen und der Strahlungsgeschichte von natürlichen Mineralien und Gesteinen. Ann. Phys. 455 (1–6), 283–292. http://dx.doi.org/10.1002/andp.19574550129.

Krbetschek, M., Götze, J., Dietrich, A., Trautmann, T., 1997. Spectral information from minerals relevant for luminescence dating. Radiat. Meas. 27 (5–6), 695–748. http://www.sciencedirect.com/science/article/pii/S1350448797002230.

Kreutzer, S., Schmidt, C., Fuchs, M.C., Dietze, M., Fischer, M., Fuchs, M., 2012. Introducing an R package for luminescence dating analysis. Anc. TL 30, 1–8. http://www.ecu.edu/cs-cas/physics/Ancient-Timeline/upload/ATL_30-1-Kreutzer.pdf.

Mejdahl, V., Botter-Jensen, L., 1994. Luminescence dating of archaeological materials using a new technique based on single aliquot measurements. Quat. Sci. Rev. 13 (5–7), 551–554. http://www.sciencedirect.com/science/article/pii/0277379194900760.

Mercier, N., Valladas, H., Froget, L., Joron, J.-L., Reyss, J.-L., Balescu, S., Escutenaire, C., Kozlowski, J., Sitlivy, V., Sobczyk, K., Zieba, A., 2003. Luminescence dates for the palaeolithic site of Piekary IIa (Poland): comparison between TL of burnt flints and OSL of a loess-like deposit. Quat. Sci. Rev. 22 (10–13), 1245–1249. http://www.sciencedirect.com/science/article/pii/S0277379103000258.

Murray, A., Wintle, A., 2000. Luminescence dating of quartz using an improved single-aliquot regenerative dose protocol. Radiat. Meas. 32, 57–73. http://www.sciencedirect.com/science/article/pii/S135044879900253X.

Peng, J., Dong, Z., Han, F., Long, H., Liu, X., 2013. R package numosl: numeric routines for optically stimulated luminescence dating. Anc. TL 31 (2), 41–48. http://www.ecu.edu/cs-cas/physics/Ancient-Timeline/upload/ATL_31_2-JunPeng.pdf.

Polikreti, K., Michael, C., Maniatis, Y., 2003. Thermoluminescence characteristics of marble and dating of freshly excavated marble objects. Radiat. Meas. 37 (1), 87–94. http://www.sciencedirect.com/science/article/pii/S1350448702000884.

Prescott, J., Huntley, D., Hutton, J., 1993. Estimation of equivalent dose in thermoluminescence dating - the australian slide method. Anc. TL 11 (1), 1–5.

R Core Team, 2016. R: a Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria. https://www.R-project.org/.

Richter, D., Krbetschek, M., 2006. A new thermoluminescence dating thechnique for heated flints. Archaeometry 48 (4), 695–705. http://dx.doi.org/10.1111/j.1475-4754.2006.00281.x.

Richter, D., Tostevin, G., Škrdla, P., Davies, W., 2009. New radiometric ages for the Early Upper Palaeolithic type locality of Brno-Bohunice (Czech Republic): comparison of OSL, IRSL, TL and 14C dating results. J. Archaeol. Sci. 36 (3), 708–720. http://www.sciencedirect.com/science/article/pii/S0305440308002574.

Schmidt, C., Kindermann, K., van Peer, P., Bubenzer, O., 2015. Multi-emission luminescence dating of heated chert from the middle stone age sequence at Sodmein cave (Red Sea Mountains, Egypt). J. Archaeol. Sci. 63, 94–103. http://www.sciencedirect.com/science/article/pii/S0305440315002629.

Schmidt, C., Sitlivy, V., Anghelinu, M., Chabai, V., Kels, H., Uthmeier, T., Hauck, T., Bălteanu, I., Hilgers, A., Richter, J., Radtke, U., 2013. First chronometric dates (tl and osl) for the aurignacian open-air site of Românești-Dumbrăviţa I, Romania. J. Archaeol. Sci. 40 (10), 3740–3753. http://www.sciencedirect.com/science/article/pii/S0305440313001258.

Singhvi, A.K., Stokes, S.C., Chauhan, N., Nagar, Y.C., Jaiswal, M.K., 2011. Changes in natural osl sensitivity during single aliquot regeneration procedure and their implications for equivalent dose determination. Geochronometria 38 (3), 231–241. http://dx.doi.org/10.2478/s13386-011-0028-3.

Smedley, R.K., 2015. A new R function for the internal external uncertainty (IEU) model. Anc. TL 33 (1), 16–21. http://www.ecu.edu/cs-cas/physics/Ancient-Timeline/upload/ATL_33-1_Smedley.pdf.

Stirling, R., Duller, G., Roberts, H., 2012. Developing a single-aliquot protocol for measuring equivalent dose in biogenic carbonates. Radiat. Meas. 47 (9), 725–731. Proceedings of the 13th International Conference on Luminescence and Electron Spin Resonance Dating, 10-14 July, 2011, Toruń, Poland. http://www.sciencedirect.com/science/article/pii/S1350448712000248.

Tite, M., Waine, J., 1962. Thermoluminescence dating : a re-appraisal. archaeometry 5 (1), 53–79. http://dx.doi.org/10.1111/j.1475-4754.1962.tb00554.x.

Tsukamoto, S., Murray, A., Huot, S., Watanuki, T., Denby, P., Bøtter-Jensen, L., 2007. Luminescence property of volcanic quartz and the use of red isothermal TL for dating tephras. Radiat. Meas. 42 (2), 190–197. http://www.sciencedirect.com/science/article/pii/S1350448706001260.

Veronese, I., Göksu, H., Schwenk, P., Herzig, F., 2008. Thermoluminescence dating of a mikveh in Ichenhausen, Germany. J. Environ. Radioact. 99, 621–630. http://www.sciencedirect.com/science/article/pii/S0265931X07002378.

Wintle, A., 1997. Luminescence dating: laboratory procedures and protocols. Radiat. Meas. 27 (5–6), 769–817. http://www.sciencedirect.com/science/article/pii/S1350448797002205.

Wintle, A., Murray, A., 2006. A review of quartz optically stimulated luminescence characteristics and their relevance in single-aliquot regeneration dating protocols. Radiat. Meas. 41 (4), 369–391. http://www.sciencedirect.com/science/article/pii/S1350448705003227.

Zöller, L., Richter, D., Blanchard, H., Einwögerer, T., Händel, M., Neugebauer-Maresch, C., 2014. Our oldest children: age constraints for the krems-wachtberg site obtained from various thermoluminescence dating approaches. Quat. Int. 351, 83–87. Loess and the record of upper Palaeolithic cultures in the Danube Basin. http://www.sciencedirect.com/science/article/pii/S1040618213002449.