TECHNISCHE UNIVERSITÄT DRESDEN

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TU BERGAKADEMIE FREIBERG

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Automated identification and separation of quartz CW-OSL signal components with R

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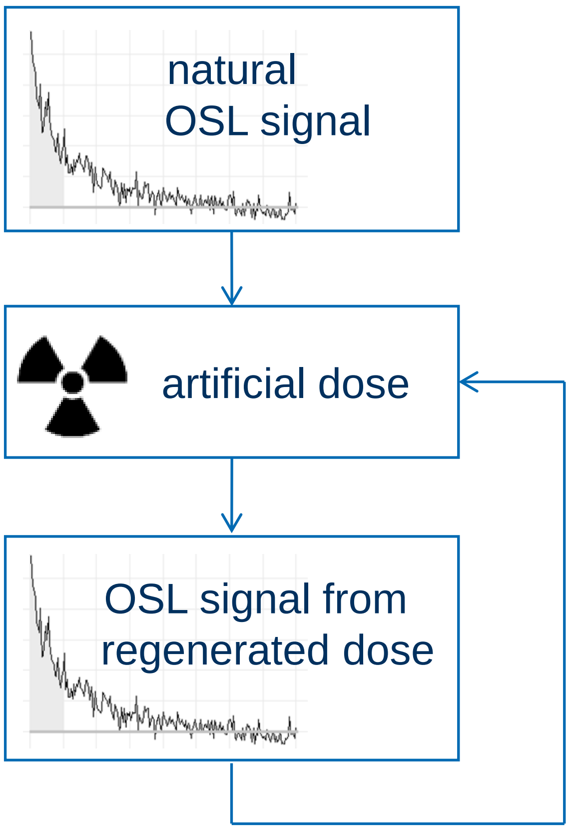
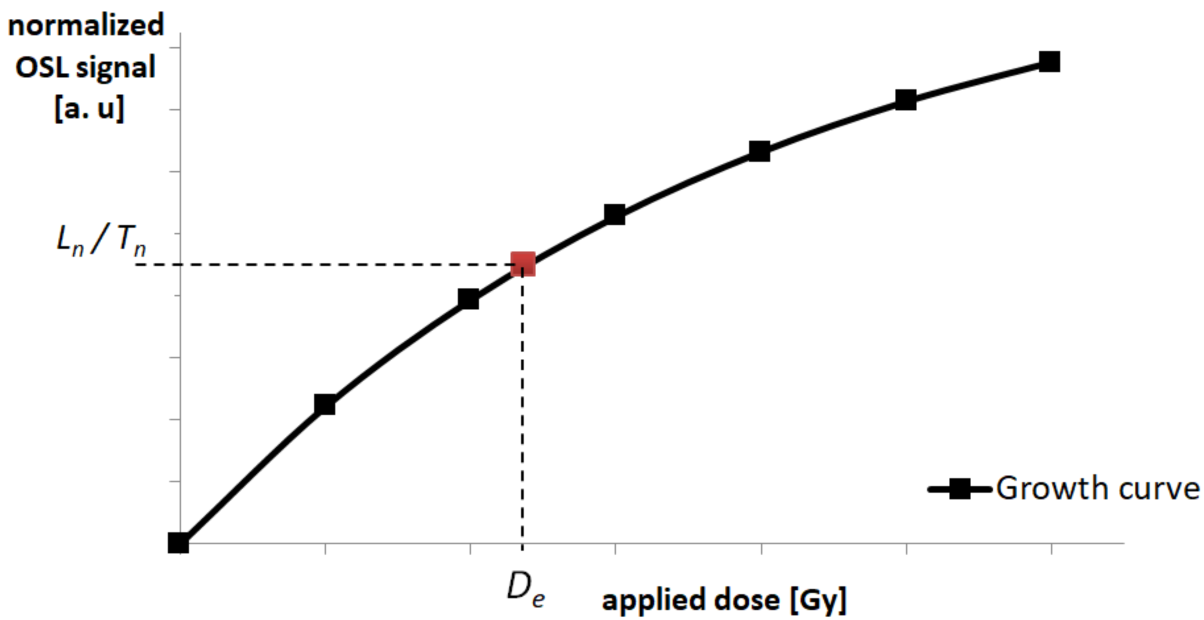
The problem

D_e calculation by SAR protocol

Natural dose is determined by building an artificial dose-signal curve





Protocol see:
Murray, A. S. & Wintle, A. G. Luminescence dating of quartz using an improved single-aliquot regenerative-dose protocol. Radiation Measurements 32, 57–73 (2000).

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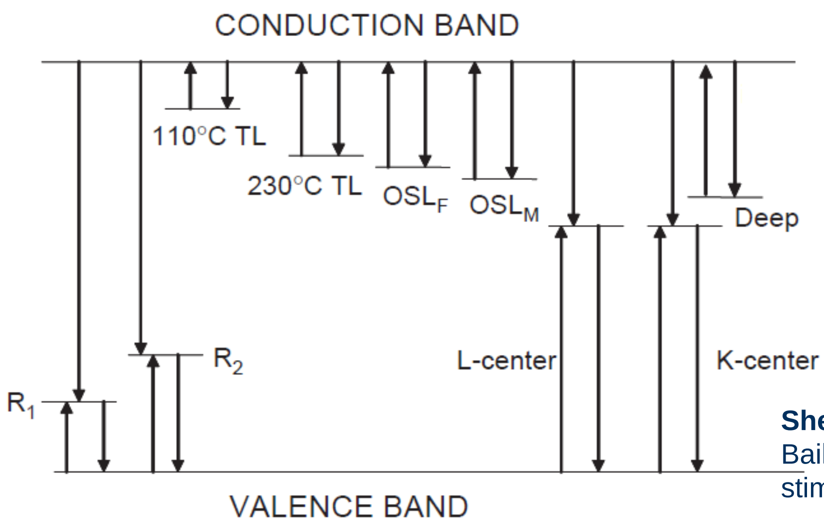
Problem



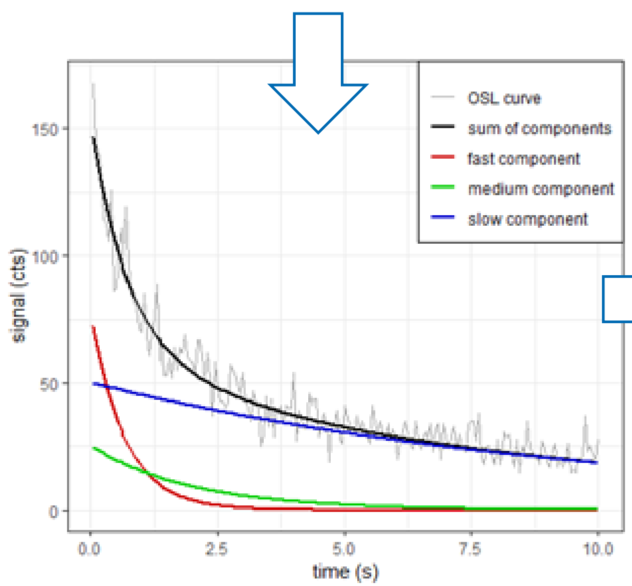
Multiple trap types contribute to the OSL signal

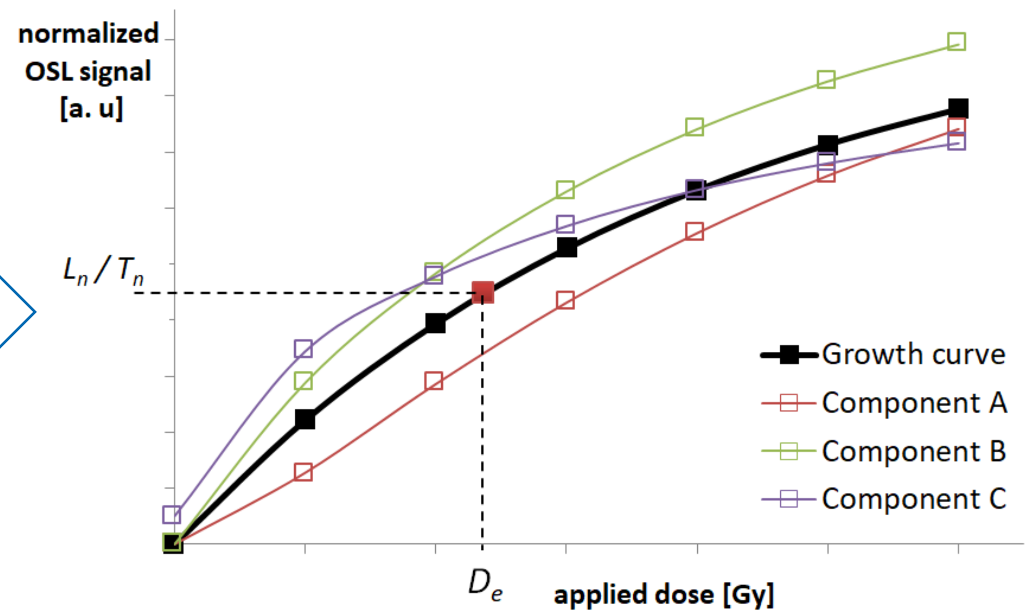
→ Traps differ in thermal stability, saturation dose, recuperation etc.

→ Complex dose-signal curve

→ D_e includes unknown systematic errors

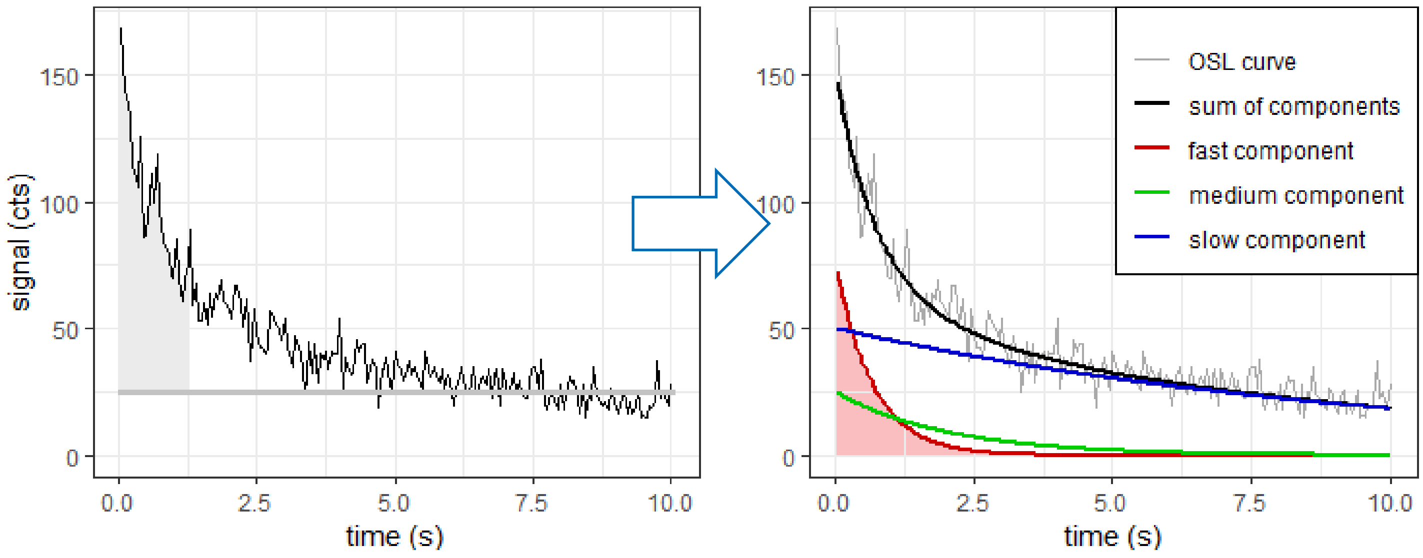
Sheme from:
Bailey, R. M. Towards a general kinetic model for optically and thermally stimulated luminescence of quartz. *Rad. Meas.* **33**, 17–45 (2001).





Goal

Provide a mathematic method for CW-OSL decomposition



- Identification of the components and their decay constants for each sample individually
- Applicable for a large variety of instrumental conditions
- Maximum robustness against instrumental noise
- **easy-to-use → applicable in daily lab routine**

The approach

Basic idea

Step 1
Find decay parameters λ_k for all K components globally

↓

Step 2
Calculate amplitudes n_k for each OSL curve

↓

Step 3
Evaluate dose for each component separately

Assumption 1

CW-OSL measurements can be sufficiently described by:

$$I(t) = \sum_{k=1}^{K} n_k \lambda_k e^{-\lambda_k t}$$

$I(t)$	OSL signal
K	number of components
n_i	component amplitude
λ_i	component decay constant

Assumption 2

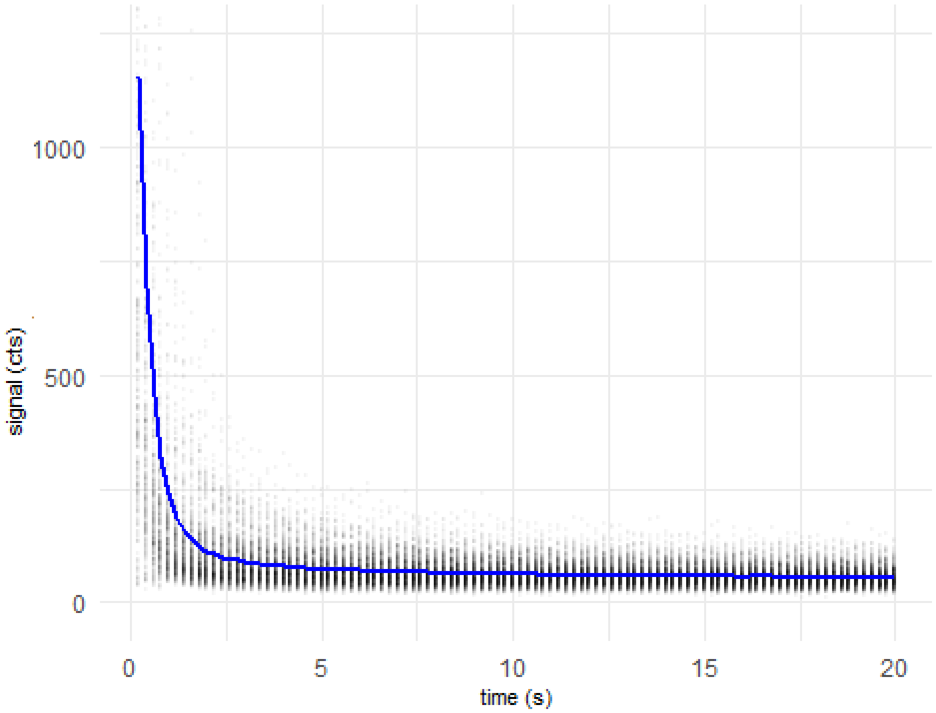
Set of λ_1 ... λ_K is the same for each OSL curve in a data set

Step 1: Workflow

Task:

Find decay parameters λ_k for all K components globally

Combine all curves to one global mean curve



Step 1: Workflow

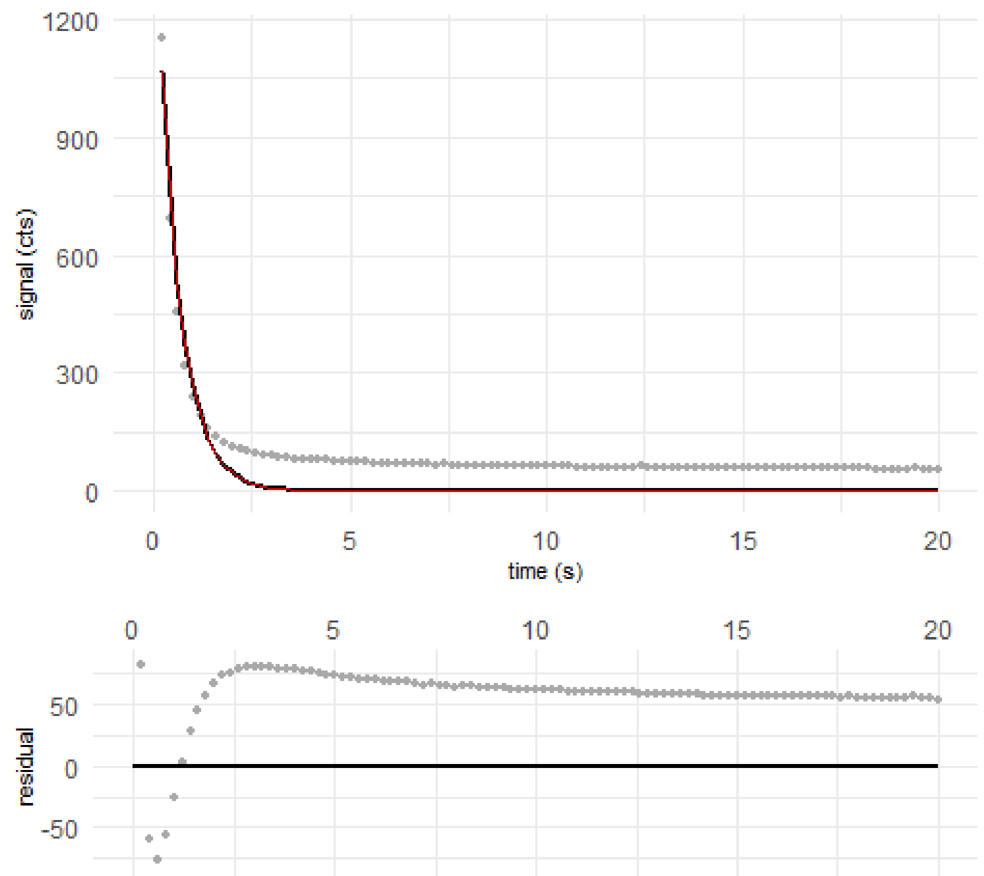
Task:
Find decay parameters λ_k for all K components globally

Combine all curves to one global mean curve

↓

Fit with increasing number of components K

K = 1



Step 1: Workflow

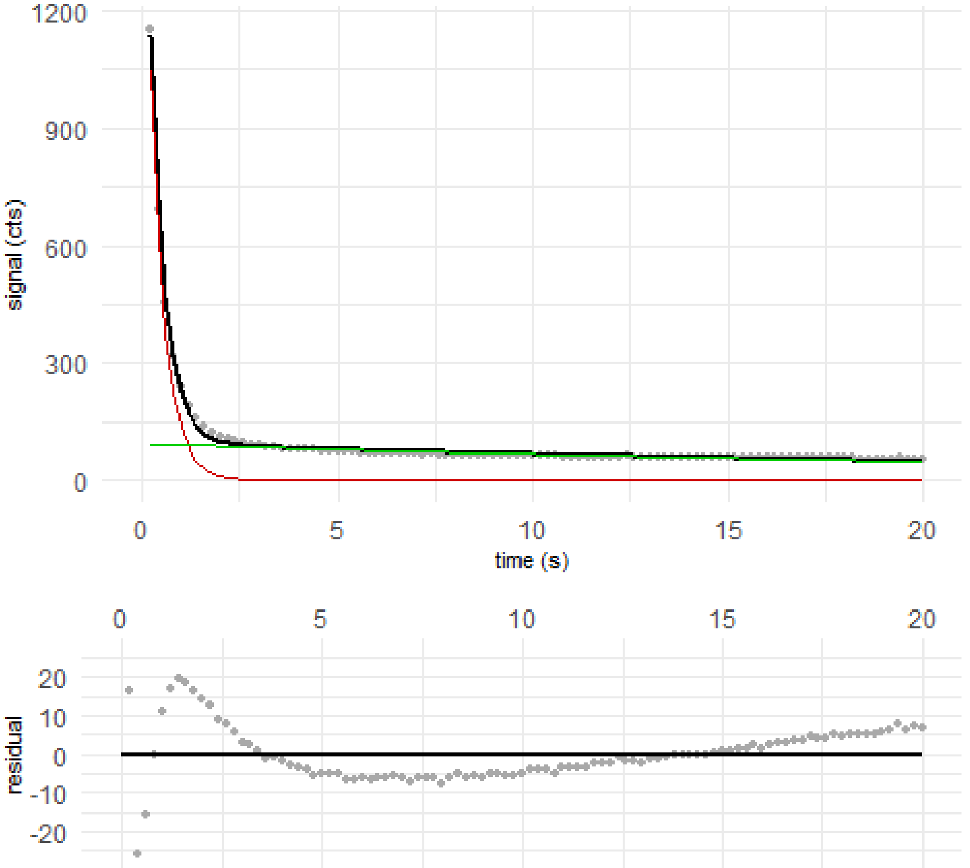
Task:
Find decay parameters λ_k for all K components globally

Combine all curves to one global mean curve

↓

Fit with increasing number of components *K*

K = 2



Step 1: Workflow

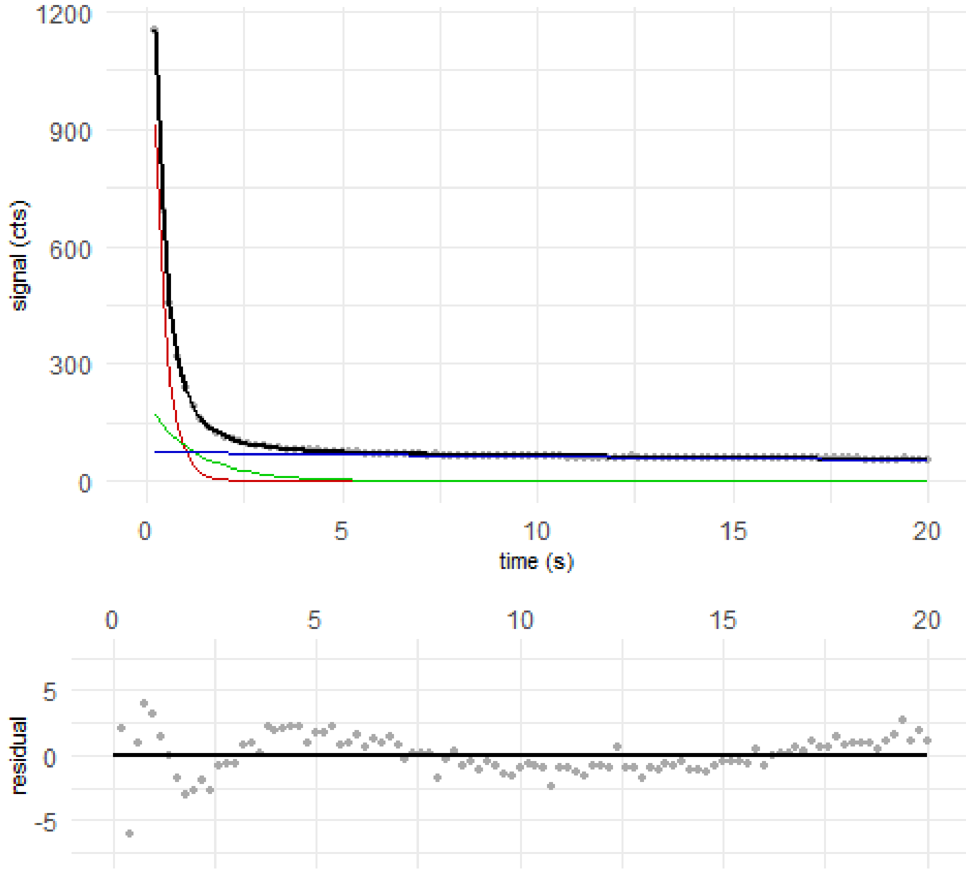
Task:
Find decay parameters λ_k for all K components globally

Combine all curves to one global mean curve

↓

Fit with increasing number of components *K*

K = 3



Step 1: Workflow

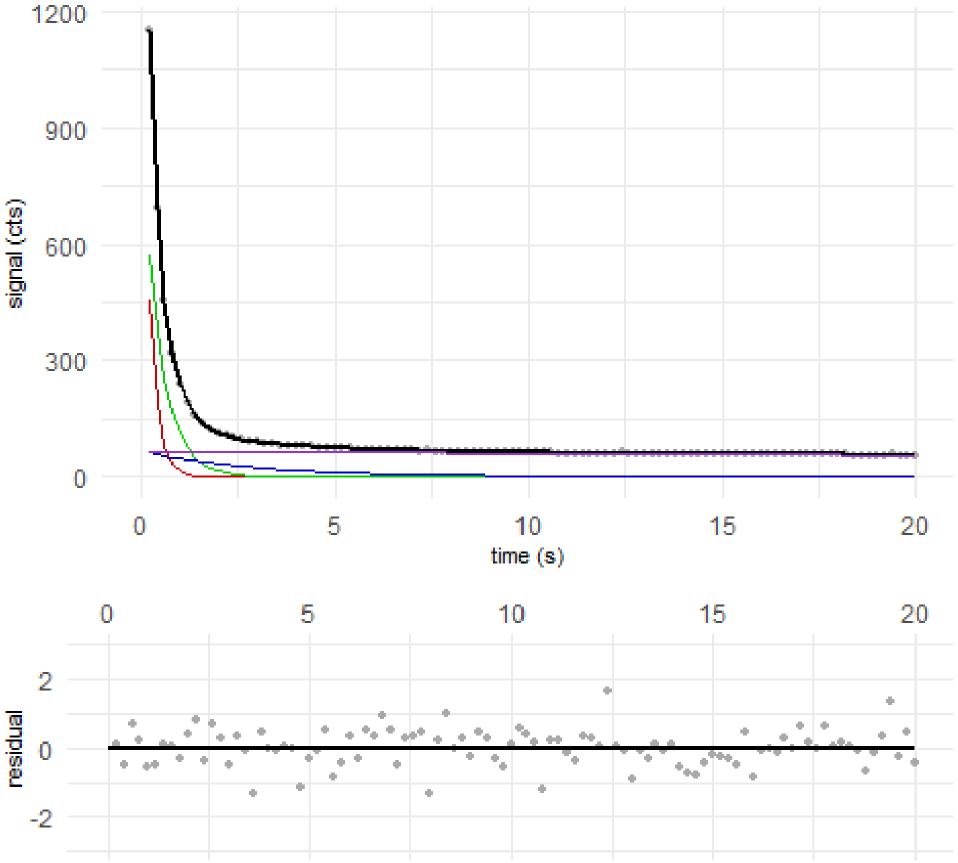
Task:
Find decay parameters λ_k for all K components globally

Combine all curves to one global mean curve

↓

Fit with increasing number of components *K*

***K* = 4**



Step 1: Workflow

Task:
Find decay parameters λ_k for all K components globally

Combine all curves to one global mean curve

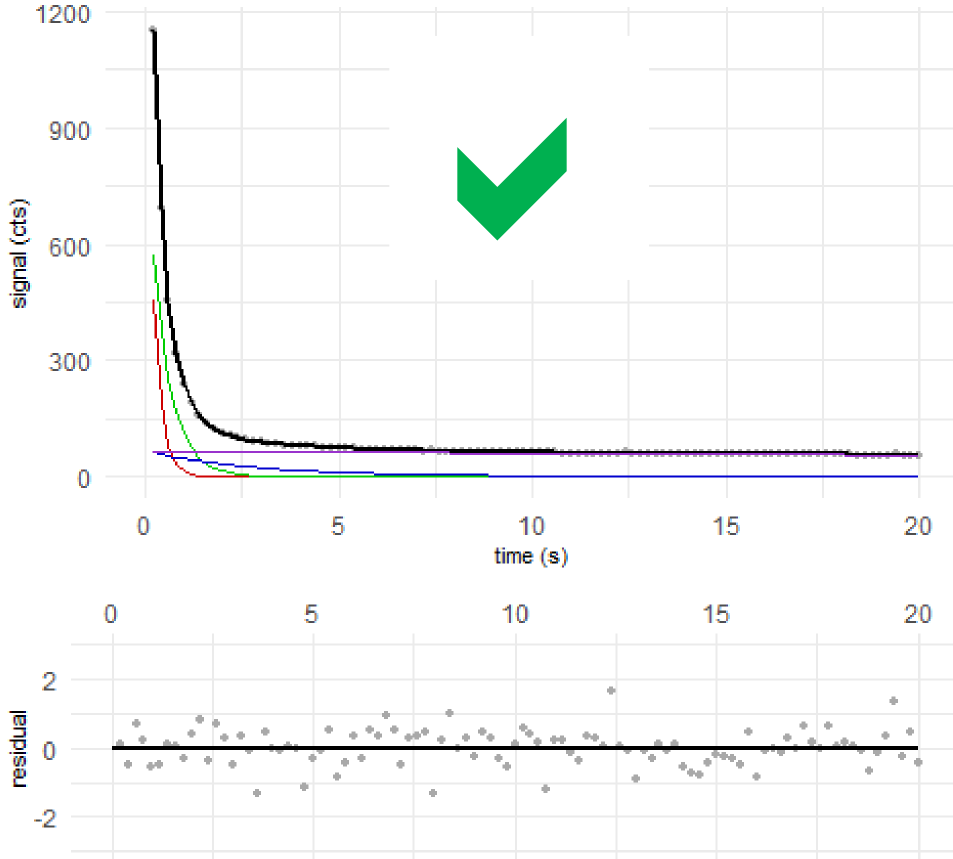
↓

Fit with increasing number of components *K*

↓

Decide which fit is the best via a statistical test

K = 4



Method proposed by:
Bluszcz, A. & Adamiec, G. Application of differential evolution to fitting OSL decay curves. *Radiation Measurements* **41**, 886–891 (2006).

Step 1: Testing

Verification by simulation:

- Virtual global mean OSL curves
- Includes noise simulation
- Set of varying parameters, with 10386 combinations
- **10386 different global OSL curves simulated and fitted**

	Parameter	Input variants
OSL components	Fast ($\lambda = 2\ s^{-1}$)	$n = 0, 1000, 3000, 10000$
	Medium ($\lambda = 0.5\ s^{-1}$)	$n = 0, 1000, 3000, 10000$
	Slow1 ($\lambda = 0.1\ s^{-1}$)	$n = 0, 3000, 10000$
	Slow2 ($\lambda = 0.02\ s^{-1}$)	$n = 10000, 30000, 100000$
Detection settings	Channel width	$\Delta t = 0.1, 0.2, 0.5$ s
	Number of channels	$N = 100, 400$
	Background signal	$b = 0, 20, 40$ cts s^{-1}
	Curve additions	$M = 100, 400$
Method options	χ^2 weighting	$\sigma^2 = 1, I_i$

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Step 1: Testing

Approximated accuracy:

		Component			
		Fast	Medium	Slow1	Slow2
Component found?		99.95%	95.8%	73.8%	96.6%
Within ±10% range?		97.4%	83.7%	65.5%	52.8%
$\lambda_{out} / \lambda_{in}$	$Q_{0.05}$	0.943	0.785	0.691	0.631
	$Q_{0.25}$	0.988	0.953	0.909	0.897
	Median	**0.998**	**0.991**	**0.981**	**0.995**
	$Q_{0.75}$	1.003	1.004	1.013	1.042
	$Q_{0.95}$	1.021	1.065	1.507	2.266

Key dependencies :

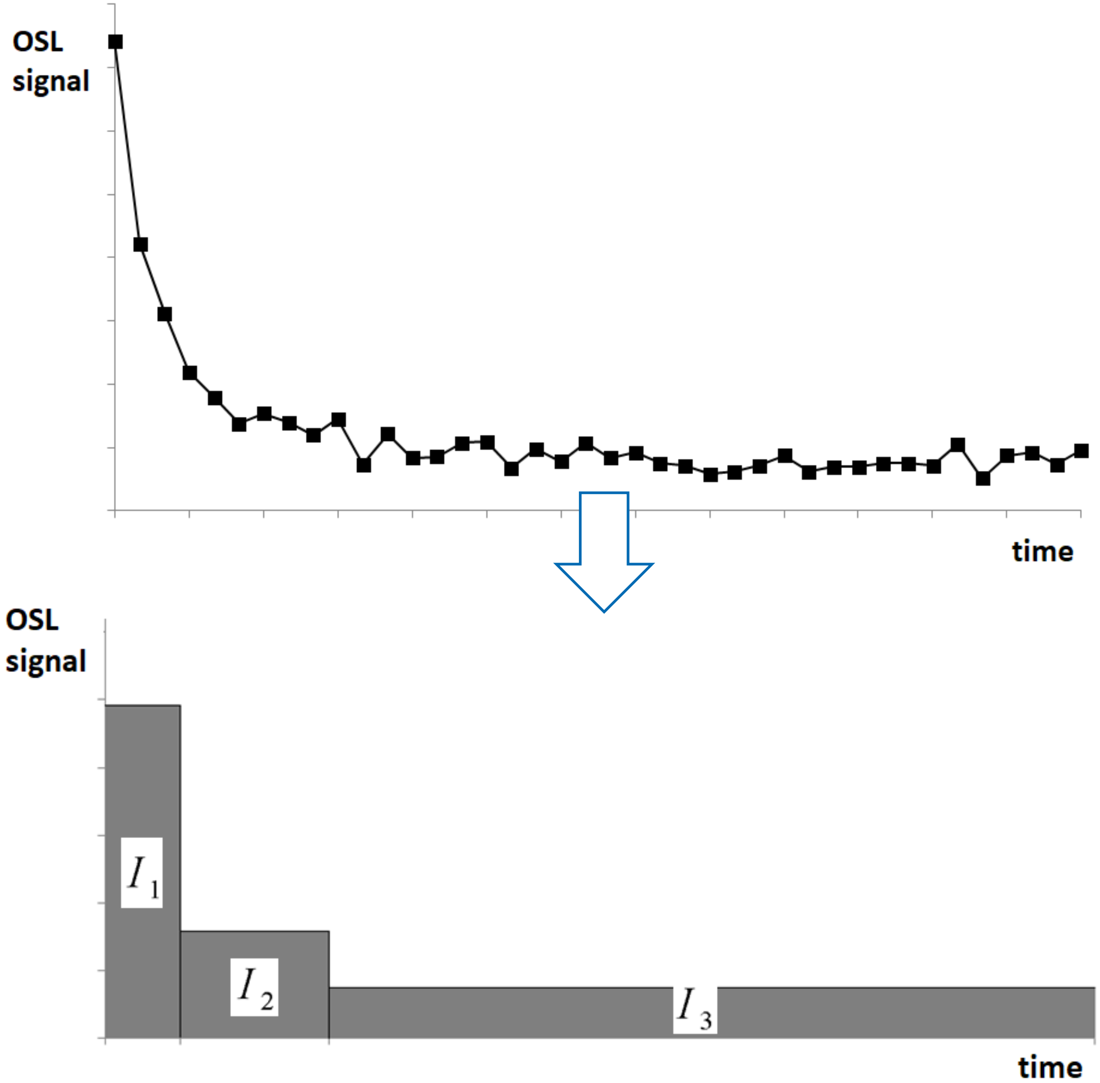
- Long measurement times → Higher chance of over-fitting (too much components)
- Short measurement time → Higher chance of under-fitting (components missed)
- Over-fitting shifts decay constants
- Insignificant correlation between background and λ_{fast} accuracy
- Just weak correlation between background and λ_{slow}accuracy

Step 2: Workflow

Task:

Calculate amplitudes n_k for each OSL curve

Divide each curve into *K* intervals and create *K* signal bins



Step 2: Workflow

Task:
Calculate amplitudes n_k for each OSL curve

Divide each curve into *K* intervals and create *K* signal bins

↓

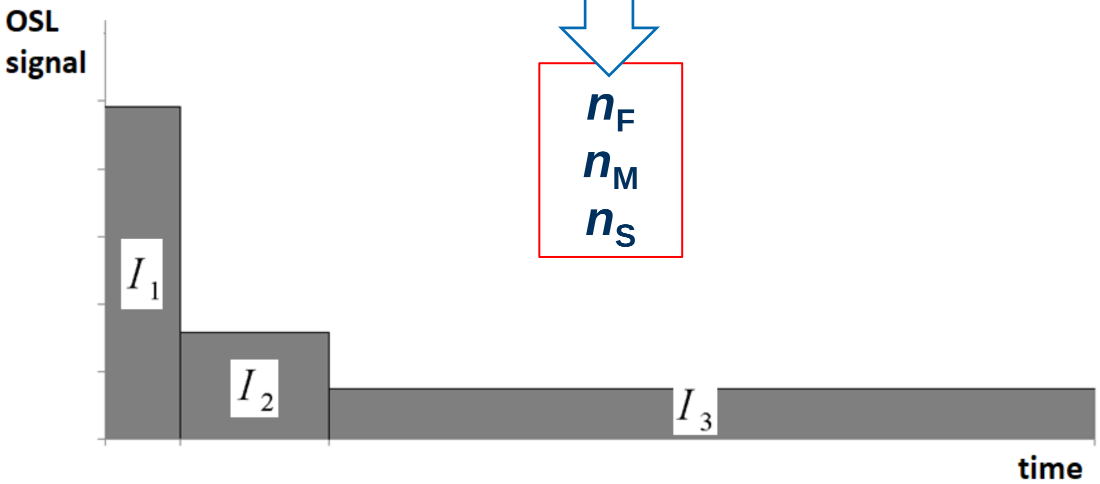
Build and solve equation system

$$I_1 = \int_{t_0}^{t_1} I(t) = n_F P_{F1} + n_M P_{M1} + n_S P_{S1}$$

$$I_2 = \int_{t_1}^{t_2} I(t) = n_F P_{F2} + n_M P_{M2} + n_S P_{S2}$$

$$I_3 = \int_{t_2}^{t_3} I(t) = n_F P_{F3} + n_M P_{M3} + n_S P_{S3}$$

with $P_{i,k} = \mathrm{e}^{-f_k t_{i-1}} - \mathrm{e}^{-f_k t_i}$



n_F
n_M
n_S

Step 2: Workflow

Task:
Calculate amplitudes n_k for each OSL curve

Divide each curve into K intervals and create K signal bins

↓

Build and solve equation system

↓

Apply propagation of uncertainty method

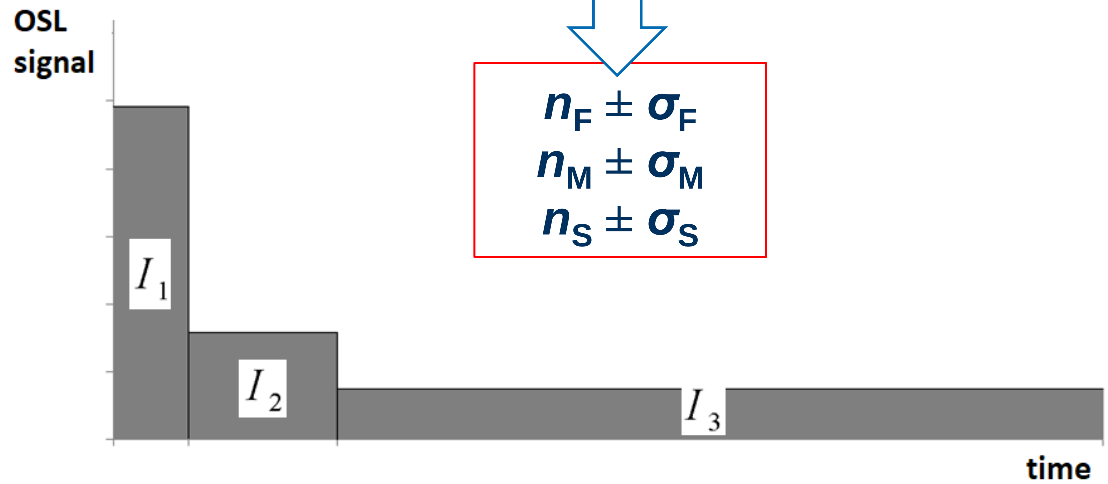
$$I_1 = \int_{t_0}^{t_1} I(t) = n_F P_{F1} + n_M P_{M1} + n_S P_{S1}$$

$$I_2 = \int_{t_1}^{t_2} I(t) = n_F P_{F2} + n_M P_{M2} + n_S P_{S2}$$

$$I_3 = \int_{t_2}^{t_3} I(t) = n_F P_{F3} + n_M P_{M3} + n_S P_{S3}$$

with $P_{i,k} = e^{-f_k t_{i-1}} - e^{-f_k t_i}$

$n_F \pm \sigma_F$
$n_M \pm \sigma_M$
$n_S \pm \sigma_S$



Step 2: Testing

Verification by simulation:

- Noise function simulating Poisson-distributed shot noise & instrumental noise
- 15552 parameter combinations
- **15.6 million OSL curves simulated and decomposed**

	Parameter	Input variants
OSL components	Fast $(\lambda = 2\ s^{-1})$	$n = 0, 1000, 3000, 10000$
	Medium $(\lambda = 0.5\ s^{-1})$	$n = 0, 1000, 3000, 10000$
	Slow1 $(\lambda = 0.1\ s^{-1})$	$n = 0, 3000, 10000$
	Slow2 $(\lambda = 0.02\ s^{-1})$	$n = 10000, 30000, 100000$
Detection settings	Channel width	$\Delta t = 0.1, 0.2, 0.5\ s$
	Number of channels	$N = 100, 400$
	Background signal	$b = 0, 20, 40\ cts\ s^{-1}$
Method options	Determine signal offset	`TRUE`, `FALSE`
	Decomposition algorithm	`det`, `nls`, `det+nls`

Step 2: Testing

Results:

- 100 % calculation success rate
- Accurate intensity results in all cases with corrected signal background
- High precision: 0% - 7% relative uncertainty, caused by decomposition method
- Accurate error estimation

Key dependencies:

- Lack of background correction leads to …
  - Slow2 intensity overestimation
  - Slow1 intensity underestimation
- Long channel widths decrease precision and error estimation accuracy

  → Channel width = 0.1 s recommended

$\frac{\bar{n}_{out}}{n_{in}}$	Component: Fast	Medium	Slow1	Slow2
	Background = 0 cts / s			
$Q_{0.05}$	1.00	0.99	0.99	1.00
Median	1.00	1.00	1.00	1.00
$Q_{0.95}$	1.00	1.01	1.01	1.00
	Background = 20 cts / s			
$Q_{0.05}$	0.98	0.94	0.71	1.01
Median	1.00	1.00	0.94	1.06
$Q_{0.95}$	1.01	1.11	0.99	1.25
	Background = 40 cts / s			
$Q_{0.05}$	0.96	0.89	0.43	1.03
Median	1.00	1.01	0.89	1.12
$Q_{0.95}$	1.03	1.23	0.98	1.50

Step 3: Workflow

Task:
Evaluate dose for each component separately

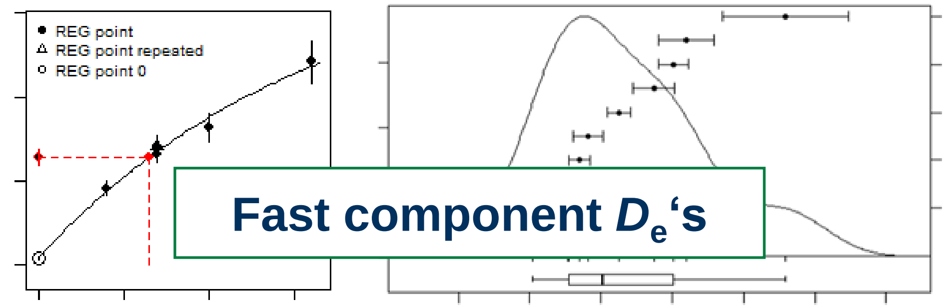
Define amplitudes n_k as component signals

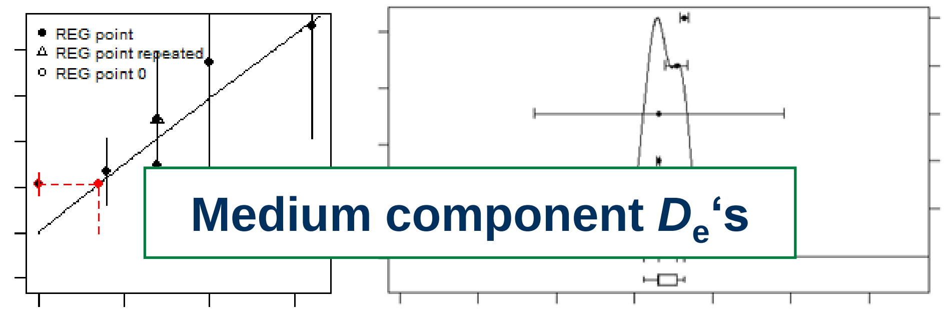
↓

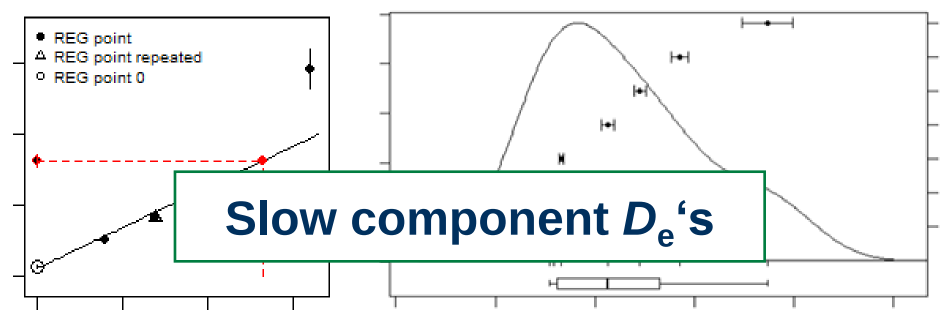
Build *LxTx* tables

↓

Calculate and plot De’s and paleodoses with all the functionality of the **R** `Luminescence` package







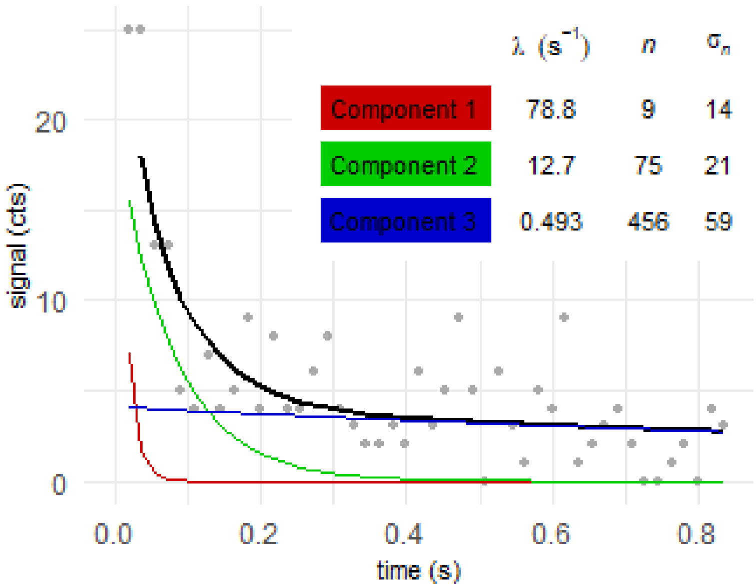
Applications

Result overview

Some application tests were performed ...

... at standard SAR samples (blue LEDs; Risø reader)

- Component identification (Step 1) approach found 4 (and once 5) components
- But 3 components matched expectations better → Step 1: *F*-test is insufficient
- Nonetheless, all *fast* component D_e's are either in accordance with late background subtraction results or match expected age even better



... at single grain data sets (green laser; Risø reader)

- Component identification found 3 components
- Component-resolved single grain dose calculation is feasible

SAR without thermal treatment

Roberts et al. (2018) proposed new protocol for OSL measurements at room temperature:

Step	Treatment	Details	Comment
1	Reg. Dose	Give dose D_i	D_i = 10 Gy at first cycle (pre-dose)
2	OSL	470 nm stimulation for 100 s	at room temperature
3	Test dose	Give dose D_T	
4	OSL	470 nm stimulation for 100 s	at room temperature
		Return to step 1	

Roberts, H. M. *et al.* Strategies for equivalent dose determination without heating, suitable for portable luminescence readers. *Rad. Meas.* (2018)

- A pre-dose fills up 110°-TL-traps
- Decomposition extracts fast-component-OSL signal from 110°-TL-trap-OSL signal

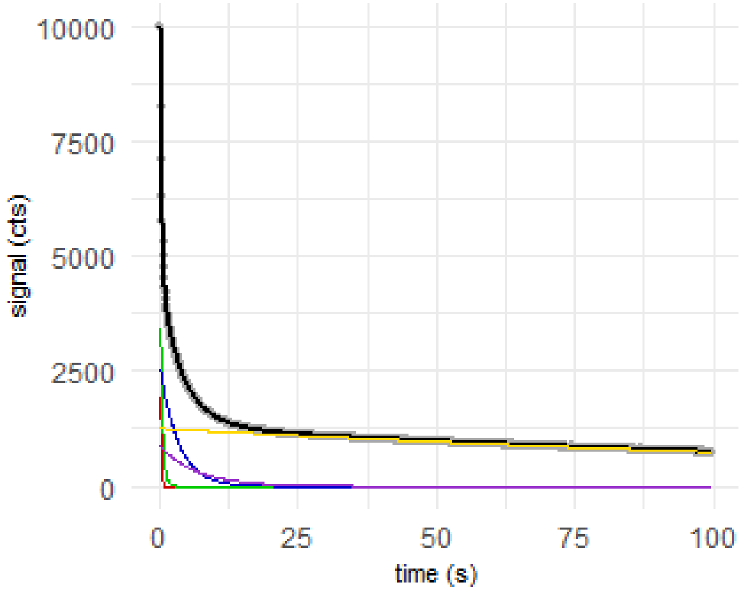
Potential gains:

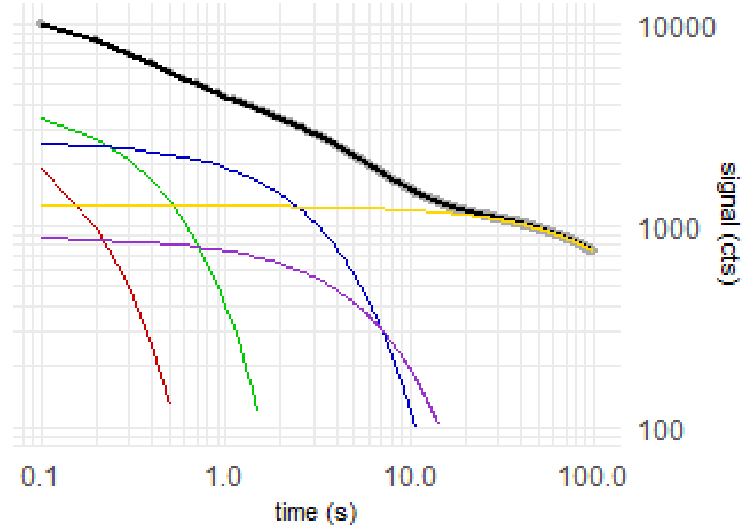
- Simplified instrumentation
- Faster measurements (no preheat/cutheat steps)
- May circumvent sensitivity change from pre-dose effect

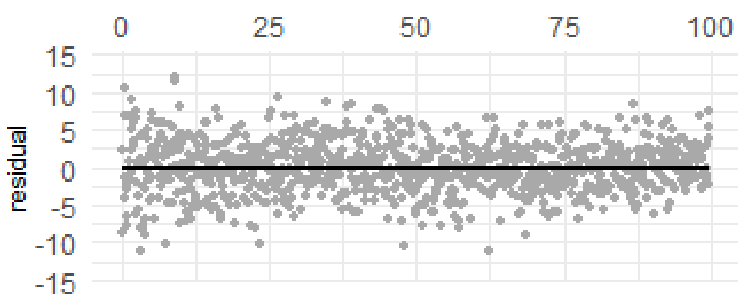
SAR without thermal treatment

Global OSL curve of ‘Fontainebleau’ reference quartz measured at room temperature

(Lexsyg research; 525 nm stimulation; ~10 Gy recovery dose; 10 aliquots + 1 background aliquot)







	λ (s^{-1})	n	σ_n	$tail_n$
Component 1	6.77	3917	88	0
Component 2	2.39	16070	158	0
Component 3	0.306	84607	300	0
Component 4	0.15	57978	298	0
Component 5	0.00529	2376637	313	1401751

SAR without thermal treatment

Fast component parameter

		decay constant (s^{-1})	Signal amplitude
Fontainebleau	heated SAR	0.79	1.20E+05
	RT SAR	0.31	8.46E+04
BT1713	heated SAR	0.98	2.00E+03
	RT SAR	0.35	2.80E+03

Fast component D_e's

		Passed rejection crit.	expected dose (Gy)	CAM (Gy)	Overdispersion
Fontainebleau	heated SAR	10 of 10	10.5	8.7 ± 0.1	1%
	RT SAR	7 of 10		10.2 ± 0.5	0%
BT1713	heated SAR	6 of 10	11.3	12.3 ± 1.7	33%
	RT SAR	1 of 10		31.8	-

High potential, but further investigations needed

Summary

Conclusion: 'OSLdecomposition' is an accurate and useful tool

What is this method useful for?

- Component-resolved OSL dating
- As tool for thermochronometry or rock surface dating (?)
- As tool to simplify measurement protocols or enable new ones
- Can be adapted for spectrometer and EM-CCD measurements

R Package download from CRAN will be available in spring 2020

Thank you

For your attention

Get notification when method goes online
send an (empty) email at: info@luminescence.de

Interested in beta-testing or any cooperation?
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