iDADwigl - a R package for open-system uranium-thorium dating

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1 Introduction

Open-system uranium-thorium (U-Th) dating of teeth and bones, while challenging, has revolutionised our ability to provide reliable chronology for humans and fauna (Eggins et al., 2005; Sambridge et al., 2012; Grün et al., 2014). Thus, this approach has significantly improved our understanding of human evolution (e.g. Dirks et al., 2017; Sutikna et al., 2016). Uranium-thorium dating is based on the premise that a material takes up U but no Th, so all the ²³⁰Th in the sample comes from decay of ²³⁸U. If detrital Th is included to the sample, a correction must be included to account for the fraction of ²³⁰Th which is detrital and not derived from ²³⁸U decay. Another requirement is that there is no gain or loss of ²³⁰Th, ²³⁴U or ²³⁸U after formation of the material. While it is often the case for many geological samples such as corals or speleothems, this requirement is rarely met for teeth and bone (although enamel can sometimes be quite impervious to isotope gain or loss). Thus, for teeth and bone, U-Th dating requires to take into account open system behaviour. 11 The diffusion-adsorption-decay (DAD) model developed by Sambridge et al. (2012) was instrumental to 12 implement successfully open-system U-Th dating. It allows for advective and diffusive transport of uranium 13 and thorium isotopes, while include synchronous radioactive decay. The software implementation was written in Fortran and is available as a Java GUI (http://www.iearth.org.au/codes/iDaD/). In this article, we 15 propose a R package which implements the model of Sambridge et al. (2012).

The motivation for providing this model as an R package is to increase the transparency, reproducibility, 17 and flexibility of the analytical workflow for computing U-Th ages. Currently it is difficult to include the 18 Java GUI in a fully scripted data analysis so the method for computing the DAD model is not highly 19 transparent. This can obscure steps where key decisions are made that are important for others to see to verify the reliability of the analysis. Enabling a scripted workflow for computational analysis of geoscience data is important for improving the reproducibility of results. Reproducibility refers the ability to recreate the results or retest the hypotheses leading to a scientific claim, either by rerunning the same code used by the original authors, or by writing new code. High rates of irreproducibility of research results have been estimated in several fields and disciplines (Ioannidis, 2005; Institute, 2013; Collaboration and others, 2015; Freedman et al., 2015; Medical Sciences, 2015; Camerer et al., 2016; Camerer et al., 2018). Consequently, the transparency, openness, and reproducibility of results and methods are receiving increased attention, and the norms of research in many fields are changing (Miguel et al., 2014; Nosek et al., 2015). 28

There is strong interest in open, transparent, and reusable research in the geoscience community (Gil et al., 2016) and substantial progress toward open data has been made in the geosciences with the widespread use of data services of NASA, USGS, NOAA and community-built data portals such as OneGeology, EarthChem, RRUFF, PANGAEA, PaleoBioDB, and others (Kattge et al., 2014; Ma, 2018). However, the use of open source software such as R (Pebesma et al., 2012), and sharing of scripted data analysis workflows with research publications is not yet widespread (Hutton et al., 2016). With this R package our goal is to make

- scripted and reproducible data analysis easy for open-system uranium-thorium dating. This will improve
- the transparency of geochronology research, and provide a more credible and robust foundation for scientific
- advancement (Hutton et al., 2016).

$_{18}$ Methods

- Data required for the DAD model are $(^{230}\text{Th}/^{238}\text{U})$ and $(^{234}\text{U}/^{238}\text{U})$ activity ratios collected along a transect
- 40 perpendicular to the surface of the tooth or bone (brackets denote activity ratios throughout this article).
- 41 Sampling for analysis can be done by micro-drilling or laser ablation. If the former, aliquots are then dissolved,
- 42 followed by separation of U and Th using ion exchange chromatography. This is more time consuming (at
- least one week of work) than laser ablation, where the material sampled by the laser is directly sent to the
- 44 mass spectrometer.
- While laser ablation also offers a better spatial resolution than micro-drilling, the precision of the data is
- inferior because of the much smaller amount of material sampled. Uranium and thorium isotope ratios are
- then analysed by multi-collector inductively-coupled plasma mass spectrometry. A plasma ionise all U and
- Th atoms, their isotopes are separated through a magnetic field and each collected in a different collector. If
- using laser ablation, it is best to have two ion counters so ²³⁰Th and ²³⁴U can be collected simultaneously.
- 50 The distance of each analysis location from the inner and outer surfaces of the bones, for instance, needs to
- be recorded. One surface is given a coordinate of 1 and the other one -1, thus coordinates of analyses take
- values in between (Figure 1).

53 Installing and attaching the package

- 54 To run the model, start R and install the package from GitHub. There are many ways to do this, one simple
- method is shown in the line below. This only needs to be done once per computer.

```
source("https://install-github.me/tonydoss/iDADwigl")
```

- 56 For routine data analysis, R scripts need to contain this line to attach the package to the current working
- environment. This line needs to be run at the start of each analysis:

```
# attach the package
library(iDADwigl)
```

58 Input data format

59 The key function of our package, iDADwigl() requires a data frame with the following column names:

```
    iDAD.position
    U234_U238_CORR
    U234_U238_CORR_Int2SE
    iDAD.position.1
    Th230_U238_CORR
    Th230_U238_CORR_Int2SE
    U_ppm
    U_ppm_Int2SE
```

This data frame can be created by importing an Excel or CSV file into the R environment using a generic function such as read.csv or read_excel from the readxl package (Wickham and Bryan, 2018). To help with preparing data for input into our function, we have included two examples of input files. The code chunk below shows how to access one of the two example CSV files included in the package, and how to read it into the R environment.

Table 1 Data contained in the example CSV file Hobbit_MH2T_for_iDAD.csv included in the package

iDAD.position	U234_U238_CORR	U234_U238_CORR_Int2SE	iDAD.position.1	Th230_U238_CORR	Th230_U238_CORR_Int2SE	U_ppm	U_ppm_Int2SE
-0.9558824	1.2696216	0.00421	-0.9558824	0.0733000	0.00226	12.3	0.615
-0.8568112	1.2729341	0.00424	-0.8568112	0.0732000	0.00238	12.7	0.635
-0.7577400	1.2654235	0.00372	-0.7577400	0.0760000	0.00177	12.5	0.625
-0.6586688	1.2673451	0.00454	-0.6586688	0.0770000	0.00193	14.2	0.710
-0.5595975	1.2691554	0.00291	-0.5595975	0.0721000	0.00188	19.8	0.990
-0.4605263	1.2655151	0.00284	-0.4605263	0.0769000	0.00167	18.0	0.900
-0.3614551	1.2669790	0.00255	-0.3614551	0.0816000	0.00169	20.0	1.000
-0.2623839	1.2760185	0.00231	-0.2623839	0.0786000	0.00126	27.2	1.360
-0.1633127	1.2655140	0.00228	-0.1633127	0.0753000	0.00124	26.7	1.335
-0.0642000	1.2766815	0.00210	-0.0642000	0.0731000	0.00098	0.3	0.015
0.0348000	1.2706338	0.00223	0.0348000	0.0779000	0.00152	33.9	1.695
0.1339009	1.2642664	0.00242	0.1339009	0.0666000	0.00138	37.7	1.885
0.2329721	1.2662201	0.00273	0.2329721	0.0713000	0.00138	30.7	1.535
0.3320434	1.2649714	0.00224	0.3320434	0.0773000	0.00185	25.2	1.260
0.4311145	1.2655783	0.00284	0.4311145	0.0832000	0.00147	28.9	1.445
0.5301858	1.2679863	0.00264	0.5301858	0.0864000	0.00162	29.0	1.450
0.6292570	1.2644629	0.00258	0.6292570	0.0816000	0.00162	27.5	1.375
0.7283282	1.2627009	0.00200	0.7283282	0.0704000	0.00132	30.1	1.505
0.8273994	1.2658484	0.00471	0.8273994	0.0953000	0.00175	19.6	0.980
0.9264706	1.2626681	0.00316	0.9264706	0.0958000	0.00232	14.8	0.740

The columns iDAD.position, U234_U238_CORR, U234_U238_CORR_Int2SE, Th230_U238_CORR and
Th230_U238_CORR_Int2SE must be present in the input data frame with these exact names for the
model to function. The iDADwig1() function will check if the input data frame has these columns, and will
stop with an error message if it does not find these columns. The names function can be used to update
column names of a data frame to ensure they match the names that the model function requires. The order
of the columns in the data frame is not important.

The iDAD.position column corresponds to the coordinates of the (²³⁴U/²³⁸U) analyses, which as indicated above take values between -1 and 1 (Figure 1). The second iDAD.position.1 column is used if the coordinates of the (²³⁰Th/²³⁸U) analyses are different from those of the (²³⁴U/²³⁸U) analyses.

Columns U234_U238_CORR and U234_U238_CORR_Int2SE are the $(^{234}\text{U}/^{238}\text{U})$ activity ratios and their 2σ

- errors. Columns Th230_U238_CORR and Th230_U238_CORR_Int2SE are the $(^{230}\text{Th}/^{238}\text{U})$ activity ratios and
- their 2σ errors.
- ⁸⁶ Columns U_ppm and U_ppm_Int2SE and calculated uranium concentrations (in ppm) and their 2σ errors.
- 87 Uranium concentrations are not necessary for the model and only used for display of the U concentration
- 88 profile in a figure.



Figure 1. Modern human femur (132A/LB/27D/03) from Liang Bua, Flores, Indonesia. Two analysis transects can be seen. For a given transect, the outer and inner surface of the bone are given 1 and -1 reference coordinates, and the position of each analysis is calculated accordingly. Modified from Sutikna et al. (2016).

Details of the input parameters

- 90 Our key function, iDADwigl() has several arguments that need to be set before we can get meaningful
- 91 results.
- nbit is the number of iterations. For the first run, set to 1.
- 93 fsum_target is the sum of the squared differences between the calculated and observed activity ratios. Give
- 94 it a low value to start with (e.g. 0.05). If script takes too long, try a higher value for fsum_target.
- $_{95}$ U48_0_min and U48_0_max are the minimum and maximum values allowed for the (234 U/ 238 U) activity ratio
- 96 at the surface of the sample. Since $(^{234}\mathrm{U}/^{238}\mathrm{U})$ does not vary greatly over the time period generally studied,
- $_{97}$ the values measured near the surface of the sample can be used as a guide. These values can be adjusted
- 98 if the model fit to the data is not optimal. For Hobbit_1-1T they are taken to be 1.3 and 1.4, and for
- 99 Hobbit_MH2T, 1.265 and 1.275, respectively.
- $_{100}$ 1 is the thickness of the sample in centimeters. For Hobbit_1-1T it is 3.5 cm, for Hobbit_MH2T it is 5.35
- 101 cm
- U_0 is the uranium concentration at the surface in ppm. This value does not significantly affect the model
- results and values from analyses near either surface of the sample can be used as a guide. For Hobbit_1-1T
- it is taken to be 15 ppm; for Hobbit MH2T, 25 ppm.
- 105 K_min and K_max are the minimum and maximum values allowed for the uranium diffusion coefficient (in
- cm^2/s). Values between 10^{-13} and 10^{-11} cm²/s are generally appropriate.
- 107 T_min and T_max are the minimum and maximum values for the age of the specimen (yr). If there is no
- estimated knowledge of the sample age, the range of values can be 1,000 to 500,000 yr and adjusted later.
- For Hobbit_1-1-T, in the final model run, they are taken to be 50,000 and 100,000 yr, and for Hobbit_MH2T,
- 1,000 and 20,000 yr, respectively.
- After setting the U480 maximum and minimum values, run the function and adjust these min and max
- values by looking at the calculated U48_0_final, K_final, and T_final. Adjust T_min and T_max using
- 113 first estimates of the age. As you iterate, increase the nbit value to reduce the error.

$_{\scriptscriptstyle 114}$ How to run the model

- To run the function, attach the package and then run iDADwigl(), specifying the input data frame and the
- input parameters as described above. The code block below shows a quick example that will execute in less

than five seconds on a typical 2.3 GHz Intel Core i5 laptop:

When run on the R console, this function will print a confirmation that the input data frame has the required columns, and print the resulting age value with an error reported as the 67% and 33% quantiles, as follows:

```
All required columns are present in the input data [1] "Age: 7 +0.6/-0.7 ka"
```

In a typical analysis we will explore the model fit by changing the range of allowed values for the (²³⁴U/²³⁸U) ratio at the surface and the age of the sample, and by decreasing the fsum_target. Once we obtain a satisfying fit (by visual inspection of the produced figures), we would increase nbit to a higher value (e.g. 1000) and run the model again.

Inspecting the model's output

The model computes a Monte Carlo simulation where age of the sample, U diffusion coefficient and (234U/238U) ratio at the surface of the sample are taken randomly within the range of values allowed.

Results are only kept if the calculated sum of the squared differences between the calculated and observed activity ratios is less than the value set in fsum_target. If this is the case, the calculated ratios and the set of solutions for age of the sample, U diffusion coefficient and (234U/238U) ratio at the surface of the sample are saved. The model stops once the number of sets of solutions reaches nbit.

The final calculated age T_final (in yr), U diffusion coefficient K_final (in cm²/s) and (²³⁴U/²³⁸U) ratio at the surface of the sample U48_0_final are the set of solutions where the solution age is the closest to the median age of the population of solutions. The uncertainty on each output paramter is calculated as the 67% and 33% quantiles of the population of solution sets. T_final, K_final and U48_0_final are included in the model's output, along with their uncertainties. The function also includes a one-row data frame summarising the age:

Table 2 Summary table of the computed age and error values

Age (ka)	Age 67% quantile (ka)	Age 33% quantile (ka)	U234_U238_0	U234_U238_0 67% quantile	U234_U238_0 33% quantile
7.016351	0.591806	0.649064	1.268382	0.006618	0.003382

The last item in the output is a copy of the input data with two additional columns, the calculated activity ratios, $(^{234}\text{U}/^{238}\text{U})$ and $(^{230}\text{Th}/^{238}\text{U})$, for each measurement location on the sample.

Table 3 Output table including the input data and two new columns

iDAD.position	U234_U238_CORR	U234_U238_CORR_Int2SE	iDAD.position.1	Th230_U238_CORR	Th230_U238_CORR_Int2SE	$_{ m Uppm}$	U_ppm_Int2SE	U234_U238_CALC	Th230_U238_CALC
-0.955882	1.269622	0.00421	-0.9558824	0.0733	0.002260	12.3	0.615	1.270050	0.0797486
-0.856811	1.272934	0.00424	-0.8568112	0.0732	0.002380	12.7	0.635	1.269722	0.0794946
-0.757740	1.265424	0.00372	-0.7577400	0.0760	0.001770	12.5	0.625	1.269429	0.0791309
-0.658669	1.267345	0.00454	-0.6586688	0.0770	0.001930	14.2	0.710	1.269173	0.0788680
-0.559598	1.269155	0.00291	-0.5595975	0.0721	0.001880	19.8	0.990	1.268952	0.0787486
-0.460526	1.265515	0.00284	-0.4605263	0.0769	0.001670	18.0	0.900	1.268768	0.0787728
-0.361455	1.266979	0.00255	-0.3614551	0.0816	0.001690	20.0	1.000	1.268619	0.0789402
-0.262384	1.276018	0.00231	-0.2623839	0.0786	0.001260	27.2	1.360	1.268506	0.0792494
-0.163313	1.265514	0.00228	-0.1633127	0.0753	0.001240	26.7	1.335	1.268429	0.0795461
-0.064200	1.276682	0.00210	-0.0642000	0.0731	0.000983	0.3	0.015	1.268388	0.0797043
0.034800	1.270634	0.00223	0.0348000	0.0779	0.001520	33.9	1.695	1.268383	0.0797247
0.133901	1.264266	0.00242	0.1339009	0.0666	0.001380	37.7	1.885	1.268414	0.0796075
0.232972	1.266220	0.00273	0.2329721	0.0713	0.001380	30.7	1.535	1.268480	0.0793522
0.332043	1.264971	0.00224	0.3320434	0.0773	0.001850	25.2	1.260	1.268582	0.0790173
0.431115	1.265578	0.00284	0.4311145	0.0832	0.001470	28.9	1.445	1.268720	0.0788076
0.530186	1.267986	0.00264	0.5301858	0.0864	0.001620	29.0	1.450	1.268894	0.0787408
0.629257	1.264463	0.00258	0.6292570	0.0816	0.001620	27.5	1.375	1.269104	0.0788175
0.728328	1.262701	0.00200	0.7283282	0.0704	0.001320	30.1	1.505	1.269349	0.0790378
0.827399	1.265848	0.00471	0.8273994	0.0953	0.001750	19.6	0.980	1.269631	0.0793927
0.926471	1.262668	0.00316	0.9264706	0.0958	0.002320	14.8	0.740	1.269949	0.0796877

Visualising the model's output

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142 iDADwigl() returns several figures useful for visualisation of the model results along with the data:

- 1. a histogram of the solution ages (Figure 2 A)
- 2. the U concentrations in the sample as a function of the relative distance from the center (Figure 2 B)
- 3. the measured (in blue) and modelled (in red) $(^{234}\text{U}/^{238}\text{U})$ activity ratios as a function of the relative distance from the center (Figure 2 C), and
- 4. the measured (in blue) and modelled (in red) (²³⁰Th/²³⁸U) activity ratios as a function of the relative distance from the center (Figure 2 D).

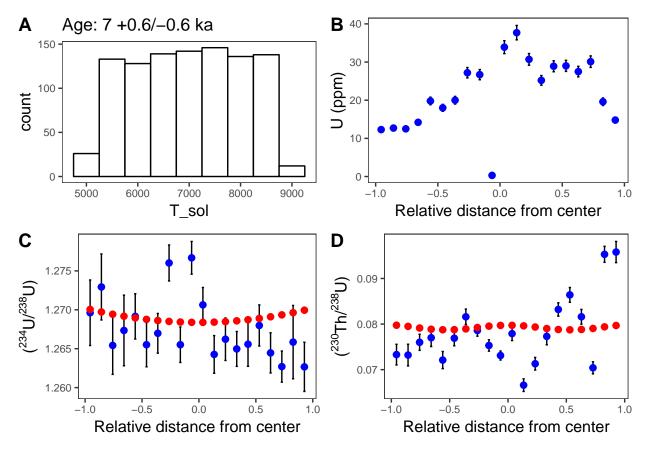


Figure 2. A: Histogram of the solution ages, B: Uranium concentration profile for transect 2 of modern human femur 132A/LB/27D/03. C: Calculated (red) and observed (blue) ($^{234}U/^{238}U$) activity ratios for transect 2 of modern human femur 132A/LB/27D/03. D: Calculated (red) and observed (blue) ($^{230}Th/^{238}U$) activity ratios for transect 2 of modern human femur 132A/LB/27D/03..

¹⁴⁹ Case study of the age of the Hobbit from Sutikna et al. 2016

```
The package includes two sample data sets derived from Sutikna et al. (2016): "Hobbit_MH2T_for_iDAD.csv" is data from transect 2 for modern human femur 132A/LB/27D/03. "Hobbit_1-1T_for_iDAD.csv" is data from transect 1 for Homo floresiensis ulna LB1/52. For the latter, six analyses were removed from the set as in Sutikna et al. (2016).
```

- For transect 2 of 132A/LB/27D/03, Sutikna et al. (2016) reported an age of 7.4 ± 0.5 ka (thousand years before 2014). With iDADwigl, we obtain an age of 7.1 + 0.6/-0.7 ka (Figs. 2-4).
- For transect 1 of LB1/52, Sutikna et al. (2016) reported an age of 79.0 ± 3.7 ka. With iDADwigl, we obtain an age of 75.4 ± 0.9 ka.

Table 4 Preview of the data from transect 2 for modern human femur 132A/LB/27D/03, included in our R package.

iDAD.position	U234_U238_CORR	$U234_U238_CORR_Int2SE$	iDAD.position.1
-0.9558824	1.269622	0.00421	-0.9558824
-0.8568112	1.272934	0.00424	-0.8568112
-0.7577400	1.265424	0.00372	-0.7577400
-0.6586688	1.267345	0.00454	-0.6586688
-0.5595976	1.269155	0.00291	-0.5595976
-0.4605263	1.265515	0.00284	-0.4605263
-0.3614551	1.266979	0.00255	-0.3614551
-0.2623839	1.276018	0.00231	-0.2623839
-0.1633127	1.265514	0.00228	-0.1633127
-0.0642000	1.276682	0.00210	-0.0642000
0.0348000	1.270634	0.00223	0.0348000
0.1339009	1.264266	0.00242	0.1339009
0.2329721	1.266220	0.00273	0.2329721
0.3320434	1.264971	0.00224	0.3320434
0.4311146	1.265578	0.00284	0.4311146
0.5301858	1.267986	0.00264	0.5301858
0.6292570	1.264463	0.00258	0.6292570
0.7283282	1.262701	0.00200	0.7283282
0.8273994	1.265848	0.00471	0.8273994
0.9264706	1.262668	0.00316	0.9264706

Table 5 Preview of the data from transect 1 for Homo floresiensis ulna LB1/52, included in our R package.

iDAD.position	U234_U238_CORR	$U234_U238_CORR_Int2SE$	iDAD.position.1
-0.1425926	1.368887	0.00244	-0.1425926
0.1648148	1.370083	0.00236	0.1648148
0.3185185	1.363741	0.00215	0.3185185
0.4722222	1.362125	0.00252	0.4722222
0.6259259	1.365091	0.00292	0.6259259
0.7796296	1.373882	0.00273	0.7796296

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