User's Manual for

Diffuser
v1.0

A program for diffusion modeling

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www.geoapp.cn

Li-Guang Wu, Yang Li, Michael C. Jollands, Pieter Vermeesch, Xian-Hua Li, 2022. Diffuser: a user-friendly program for diffusion chronometry with robust uncertainty estimation. Computers and Geosciences. In revision.

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Diffuser is a program written in MATLAB to model elemental diffusion in minerals. It is coded with robust uncertainty propagation of curve fitting, temperature, and experimentally determined diffusion coefficients. A web version is recommended for all users and available at http://www.geoapp.cn/. Diffuser can also be downloaded and installed from https://github.com/liguangwu/diffuser.git.

A user manual is given below.

1 Data input

For running Diffuser directly in MATLAB, the user can run 'Diffuser.mlapp' or type 'Diffuser' in the MATLAB command window. After opening the graphical user interface (GUI) of Diffuser, the user can click the '**Select file**' button from the import data panel (Figure 1).

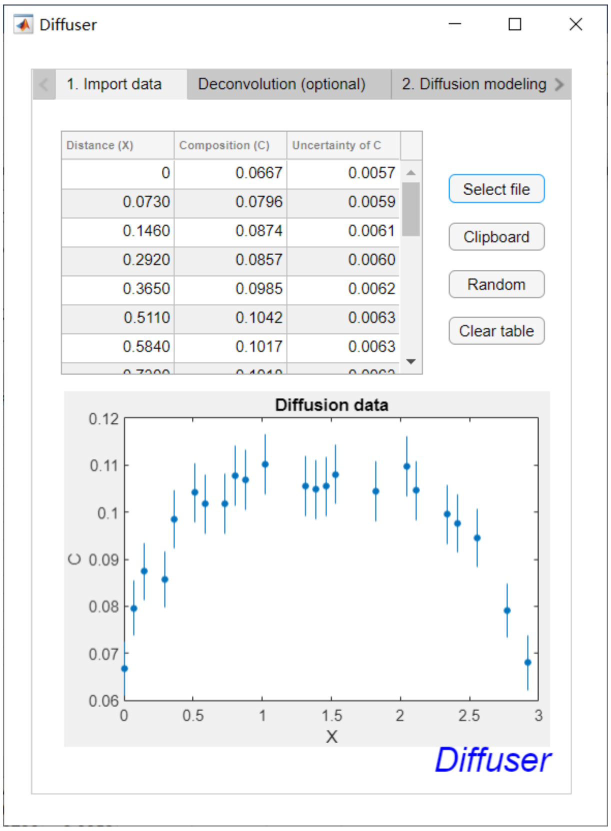

Figure 1. Import data panel of Diffuser

The measured diffusion profile can be fed into the software through a delimited text (e.g., txt or csv format), spreadsheet file (Microsoft® Excel), or clipboard (by the '**clipboard**' button; Figure 1). The distance and composition data should be in two columns (Figure 2). If composition uncertainties (σ) are assigned in another column (Figure 3), Diffuser will ask the user to input the column names of x, C, and σ (Figure 4). Otherwise, all composition data will be treated with equal weights. After data import,

the data is directly visualized by means of a pre-formatted plot (Figure 1).
To play with Diffuser, the user can click the ‘**Random**’ button and Diffuser will generate a random diffusion profile. Then the user can deal with the example data in the same way. For clearing the table and plot of diffusion data, just click the ‘**Clear table**’ button.

	A	B	C	D	E	F
1	Rubin et al., 2007	Fig. 1, Peak 2	put uncertainty in third column if you have			
2	X(m)	7Li ppb				
3	3.401E-05	0.000				
4	3.470E-05	1.187				
5	3.500E-05	0.591				
6	3.569E-05	1.778				
7	3.598E-05	0.000				
8	3.668E-05	1.179				
9	3.707E-05	3.534				
10	3.737E-05	6.524				
11	3.806E-05	6.553				
12	3.836E-05	16.015				
13	3.905E-05	25.248				
14	3.935E-05	48.689				
15	4.004E-05	75.149				

Figure 2. A diffusion profile containing two-column data.

	A	B	C	D	E	F
1	x (mm)	CaO (wt%)	1s			
2	0	0.0667	0.0057			
3	0.073	0.0796	0.0059			
4	0.146	0.0874	0.0061			
5	0.292	0.0857	0.006			
6	0.365	0.0985	0.0062			
7	0.511	0.1042	0.0063			
8	0.584	0.1017	0.0063			
9	0.73	0.1018	0.0063			
10	0.803	0.1077	0.0064			

Figure 3. A diffusion profile containing three-column data.

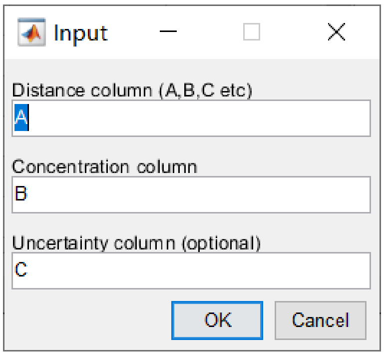

Figure 4. A pop-up window to input the column names of x, C, and σ.

2 Deconvolution (optional)

All in-situ analytical techniques have a non-zero beam size, so the measured diffusion profiles will suffer from convolution to some degree, especially when the diffusion length approaches the resolution of the analytical technique. Jollands (2020) has developed a program for numerically deconvoluting diffusion profiles acquired using techniques with Gaussian, Lorentzian, (pseudo-)Voigt, circular/elliptical, or square/rectangular interaction volumes (PACE-the Program for Assessing Convolution Effects), which has been incorporated into Diffuser.

The user can evaluate the convolution effect and determine whether deconvolution is needed or not using Diffuser. If the user chooses deconvolution by ticking the box of '**Deconvolute analytical beam effects**' on the 'Deconvolution' panel (Figure 5), the beam shape and size and the **size unit** can be set. If a **circular/elliptical** beam shape is selected, the **diameter** of the beam is required. If a **square/rectangular** beam shape is selected, the **width** of the beam is required. If a **Gaussian or Lorentzian** beam shape is selected, the **full width of half maximum** (FWHM) of the beam is required. If a **pseudo-Voigt** beam shape is selected, the **FWHM of both the Gaussian and Lorentzian** components is needed. After the beam shape and size set, Diffuser will deconvolute the diffusion profile to calculate the *Dt* value in the next diffusion modeling step.

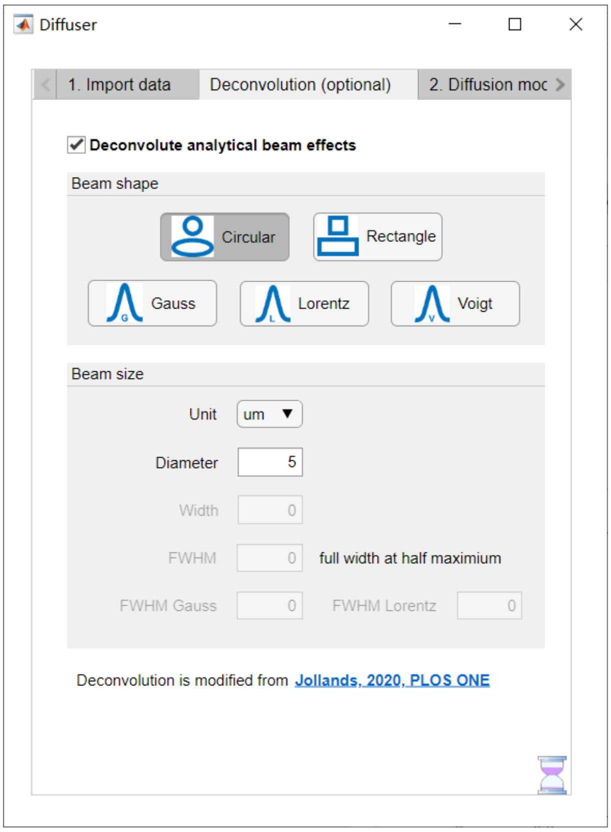

Figure 5. Deconvolution panel of Diffuser

3 Diffusion modeling

After diffusion data import, the user can choose the relevant solution to Fick's second law graphically, on the diffusion modeling panel ('**Diffusion profile**' in Figure 6) by comparing the shape of the measured data profile with diffusion profiles in Figure 6. The **unit of position** $\boldsymbol{x}$ should be set and **initial compositions (**C_1**,** C_2**, and** C_3**)** can be fixed or determined by fitting. Definitions of C_1, C_2, and C_3 can be looked up through the '**See C_1, C_2, and C_3 definitions**' button. It should be emphasized that a wrong unit of x can lead to an unusual timescale which should be carefully checked. Then, the user can start a diffusion model and a modeled *Dt* value can be acquired (Figure 7) and saved in a file (Figure 8). The user can evaluate the diffusion data quality through the goodness of fit (R^2). Usually, a low R^2 value (e.g., <0.8) indicates that the curve fitting of the diffusion profile will contribute to a large uncertainty of the timescale. Such a diffusion profile may not be good or valid to further calculate the timescale.

Specifically, if the user does not define an initial flat peak or trough for profiles I–L (i.e., no tick in the box of '**a flat peak/trough can be seen in profile I-L**'), more parameters including **minimum and maximum values and a step length of the assumed flat peak or trough (C_0)** are required (Figure 9). Diffuser will plot the modeled *Dt* values vs. assumed initial flat compositions (Figure 10).

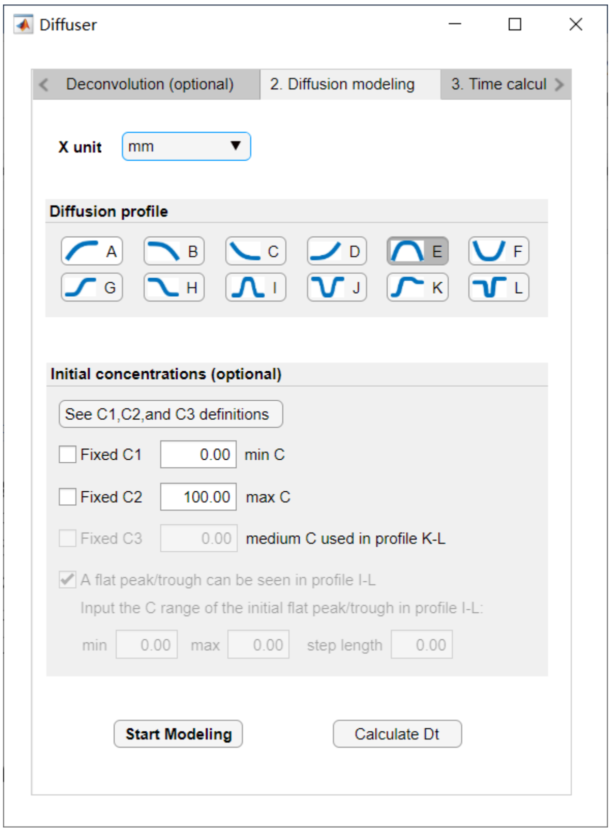

Figure 6. Diffusion modeling panel of Diffuser.

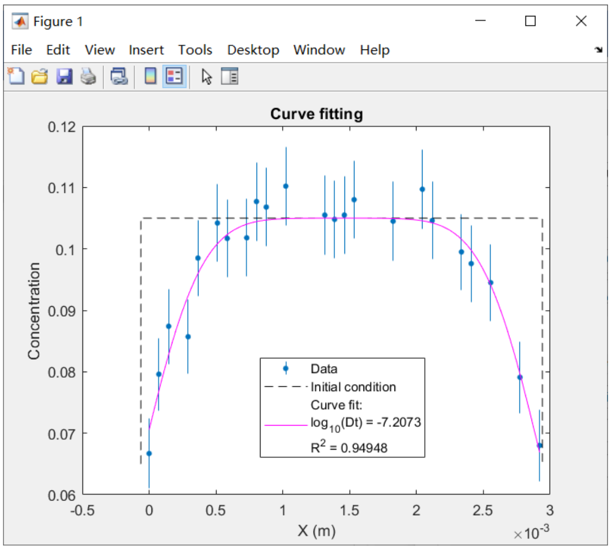

Figure 7. Curve fitting of the diffusion profile.

	A	B	C	D	E	F	G	H	I	J
1	Trials	R2	C1	C2	x0 (m)	rim-to-core length (m)	log10(Dt)	2s	Dt	2s
2	1	0.949479683	0.065	0.105	0.001439735	0.001501448	-7.207316696	0.169266205	6.20416E-08	2.41807E-08
3										

Figure 8. Curve fitting result of the diffusion profile.

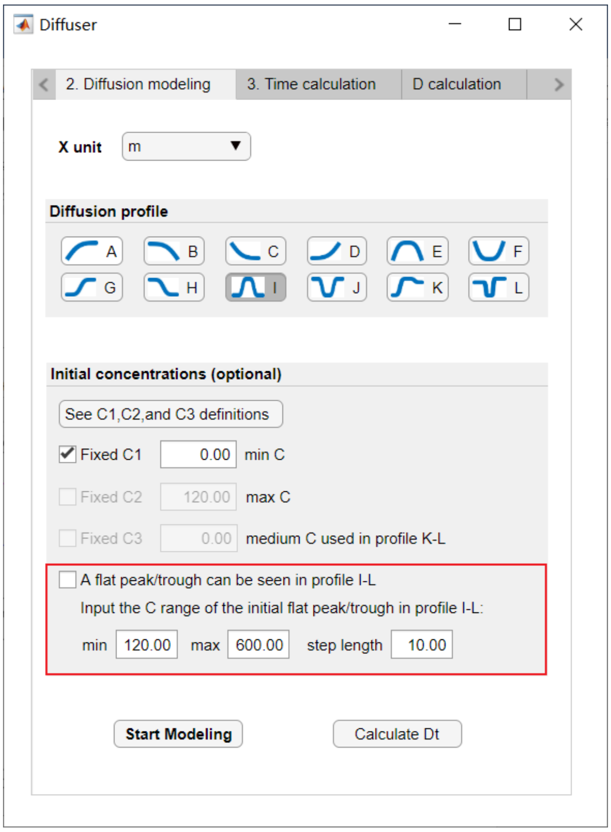

Figure 9. Minimum and maximum values and a step length of the assumed flat peak or trough for profiles I–L.

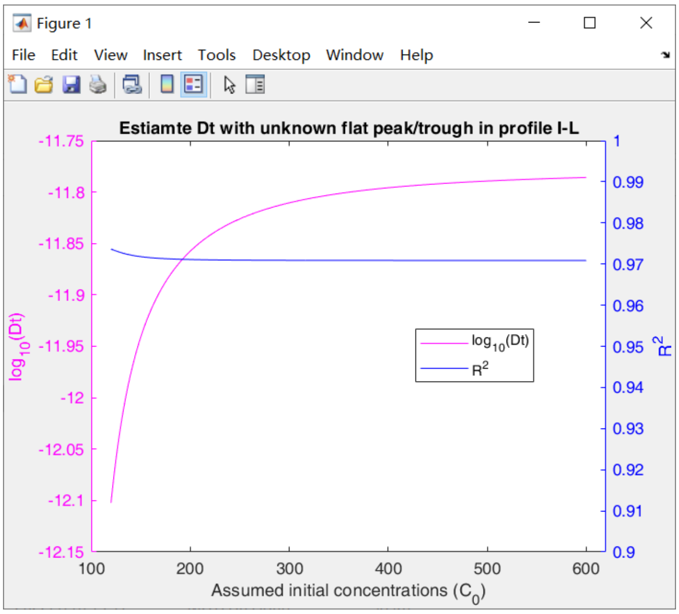

Figure 10. Estimate *Dt* with an assumed composition range of the initial flat peak/trough for profiles I–L.

4 Time calculation

To model timescales in natural samples, the **diffusion coefficient, initial temperature with its uncertainty, trials for the Monte Carlo calculation, and cooling path (linear, exponential, or parabolic) with its constant coefficient** should be set on the time calculation panel **when using an Arrhenius equation for *D*** (Figure 11A). Otherwise, if the user uses **a constant *D***, the ***D* value and its uncertainty** can be set instead of the above parameters (Figure 11B).

If using an Arrhenius equation for *D*, the user can choose propagation of the experimentally determined parameters (by ticking the box of '**Calculate with D_0 and E_a errors**') to see the contributions to the uncertainty of the calculated timescale compared with curve fitting and temperature. If the user selects an isothermal diffusion history for the 'cooling path', the program will calculate a constant *D* using the input initial temperature. If selecting a linear, exponential, or parabolic cooling path, the user should input the constant coefficient (A) in the form of:

T=T_0-At for linear cooling

T=T_0e^{-At} for exponential cooling

T=T_0-At^2 for parabolic cooling

In addition, diffusion coefficient data collected from the literature are provided in a built-in excel file ('DiffusionCoefficient.xlsx', Figure 12). The program will read it automatically and users can access them via a drop-list ('**Mineral-Element-Reference**') on the diffusion coefficient panel (Figure 11A) once the program interface is opened. Currently, Diffuser contains a library of diffusion coefficients for the following systems: olivine (Ca, Al, P, REE, Ti, H, Li, Be), garnet (Hf, REE, H), quartz (Ti, Al, H), zircon

(REE, Ti, Al, Li), orthopyroxene (REE, Ti, Cr, H), clinopyroxene (Ti, REE, H), and feldspar (Sr, Ba, REE, H). These have been compiled in Diffuser by refitting original experimental data in previous studies. Thus, the algorithm of parameter uncertainties in Diffuser is internally consistent. Diffusion coefficients from other systems can be added into Diffuser by modifying the relevant template file. Requests to add other diffusion coefficients to future web versions of Diffuser can be sent to the first author (wuliguang@mail.iggcas.ac.cn).

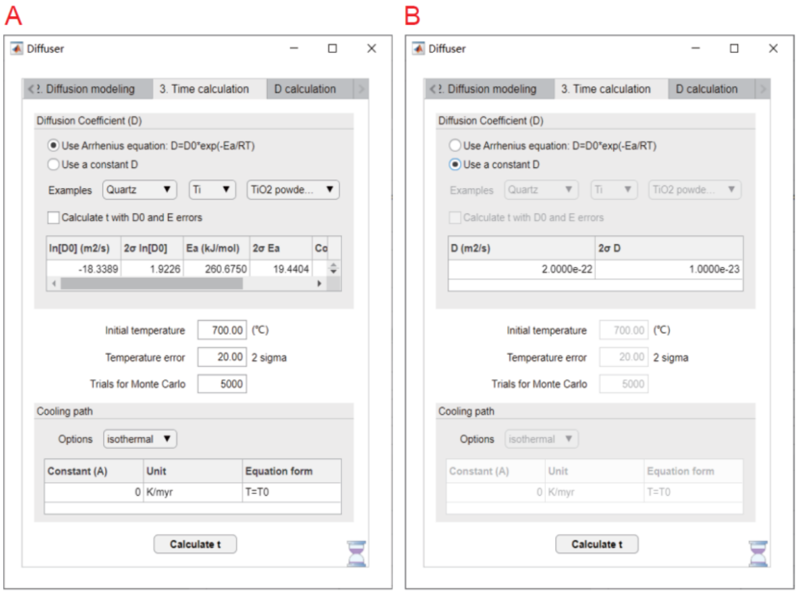

Figure 11. Time calculation panel of Diffuser.

	A	B	C	D	E	F	G	H
1	Mineral	Element	Reference	ln(D0) (m^2/s)	2σ ln(D0)	E (kJ/mol)	2σ E (kJ/mol)	Covariance (ln(D0),E)
2	Quartz	Ti	//[001], Cherniak et al. (2007), Chem. Geol.	-16.27	1.83	275.66	18.86	8.58
3	Quartz	Ti	//[001], Jollands et al. (2020), Geology	-19.21	2.74	310.61	34.48	23.42
4	Quartz	Al	Tailby et al. (2018), Am. Mineral.	-24.85	8.61	196.65	75.98	163.35
5	Zircon	Li	Trail et al. (2016) and Cherniak and Watson (2010), Contrib.	-13.85	2.08	278.27	19.93	10.34
6	Olivine	P	//[010], Watson et al. (2015), Am. Mineral.	-22.96	3.03	230.35	26.86	20.33
7	Olivine	Ca	//[100], Bloch et al. (2019), Geochim. Cosmochim. Acta	-19.34	1.38	250.05	13.87	4.75
8	Olivine	Ca	//[100], log[fO2]=-10 (bars), Coogan et al. (2005), Geochim.	-20.19	3.19	233.64	40.25	31.99
9	Olivine	Ca	//[010], log[fO2]=-10 (bars), Coogan et al. (2005), Geochim.	-23.59	2.54	189.64	32.02	20.25
10	Olivine	Ca	//[001], log[fO2]=-10 (bars), Coogan et al. (2005), Geochim.	-23.20	3.54	188.40	44.84	39.55
11	Olivine	Al	//[100] (fo+en+crd), log[fO2]=-0.68, Zhukova et al. (2017), C	-2.86	12.50	367.63	164.74	514.36
12	Olivine	REE	Cherniak (2010), Am. Mineral.	-21.81	3.26	276.93	35.38	28.79
13	Olivine	REE	assumed a sensible E of 225±25 kJ/mol; Spandler and O'Ne	-17.88	3.84	225.00	25.00	23.77
14	Olivine	Ti	forsterite, Cherniak and Liang (2014), Geochim. Cosmochim.	-29.56	2.84	212.77	33.63	23.82
15	Olivine	Ti	San Carlos olivine, Cherniak and Liang (2014), Geochim. Co	-31.36	2.67	196.81	31.72	21.12
16	Olivine	H	//[100], Jollands et al. (2016), Am. Mineral.	-6.28	3.52	229.92	32.80	28.84
17	Olivine	H	//[100], Demouchy and Mackwell (2003), Phys Chem Mineral	-9.90	3.68	225.00	40.00	36.47
18	Olivine	H	//[100], Kohlstedt and Mackwell (1998), Zeitschrift für Physik	-8.88	5.26	144.81	50.87	66.71
19	Olivine	H	//[010], Demouchy and Mackwell (2003), Phys Chem Mineral	-10.59	2.76	205.00	31.00	21.20
20	Olivine	H	//[010], Kohlstedt and Mackwell (1998), Zeitschrift für Physik	-8.23	10.44	185.87	103.02	268.46
21	Olivine	H	//[001], Jollands et al. (2016), Am. Mineral.	-5.29	1.95	220.69	17.54	8.50
22	Olivine	H	//[001], Demouchy and Mackwell (2003), Phys Chem Mineral	-8.75	2.99	210.00	33.00	24.45
23	Olivine	H	//[001], Kohlstedt and Mackwell (1998), Zeitschrift für Physik	-16.40	9.92	103.69	97.83	242.20
24	Olivine	Li	//[001], interstitial site, Dohmen et al. (2010), Geochim. Cosm	-5.34	1.23	214.24	12.50	3.82
25	Olivine	Li	//[001], octahedral site, Dohmen et al. (2010), Geochim. Cos	-15.38	5.53	188.04	56.10	76.91
26	Olivine	Be	//[001], Jollands et al. (2016), Earth Planet. Sci. Lett.	-13.06	0.71	231.96	8.55	1.52

Figure 12. Library of diffusion coefficients in Diffuser.

After all parameters are set, the user can start calculating the timescale. If the temperature is assigned a non-zero error, a marginal plot will show distributions of the temperature and diffusion timescale and the trade-off between these two parameters (Figure 13). Otherwise, only a histogram will show the distributions of the timescale (Figure 14). A histogram will also show the uncertainty budget of the modeled timescale (Figure 15). so that the user can evaluate the main contributions of the timescale uncertainty. This helps users find more ways for better timescale constraints, e.g., a diffusion profile with a better R^2 value of curve fitting, a more precious method of temperature estimation, or a diffusive element with a more precious diffusion coefficient.

If the temperature decreases with time (non-isothermal systems), Monte Carlo results of timescales are displayed on histograms to show whether they are log-normally distributed based on the Lilliefors test (Figure 16). If not, the user should consider using a larger number of trials for Monte Carlo modeling. A dialog will appear saying that 'Trials are not enough to estimate ln[t] uncertainty budget' (Figure 17) when the calculated uncertainty of the timescale using propagation of three error sources (curve fitting, temperature, and D_0 and E_a) is even smaller than that using propagation of two error sources (or two sources<one source). In this case, the user also should consider using a larger number of trials for Monte Carlo modeling.

Finally, the modeled timescale and Monte Carlo results can be saved in individual files (Figures 18 and 19). Specifically, if the user does not define an initial flat peak or trough for profiles I–L, Diffuser will plot the modeled timescales vs. assumed initial flat compositions (Figure 20).

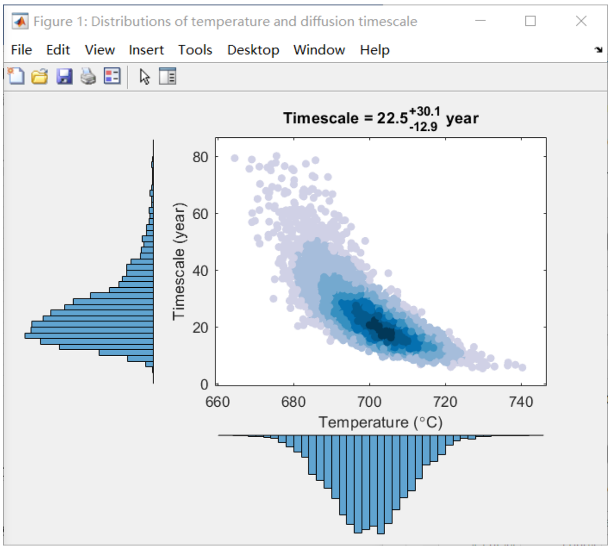

Figure 13. Marginal plot showing distributions of the temperature and diffusion timescale when the temperature is assigned a non-zero error.

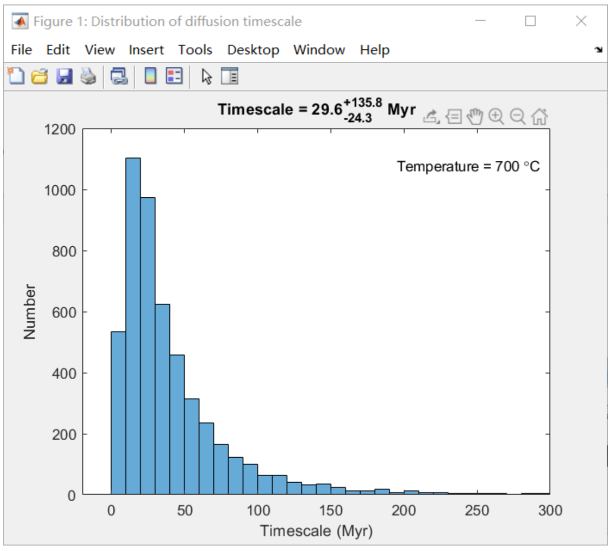

Figure 14. Histogram showing the distribution of the modeled timescale when the temperature is assigned an error of zero.

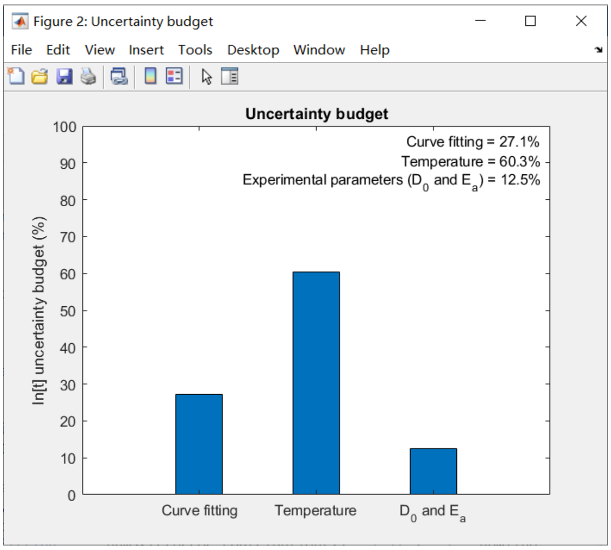

Figure 15. Histogram showing the uncertainty budget of the modeled timescale.

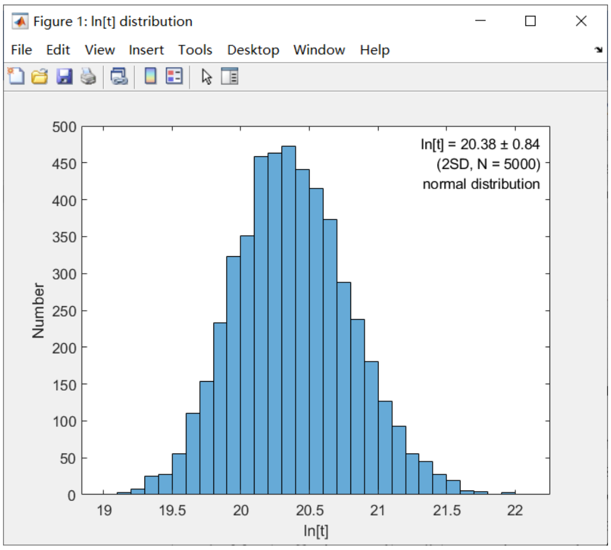

Figure 16. Histogram showing whether ln[t] are log-normally distributed for non-isothermal diffusion modeling.

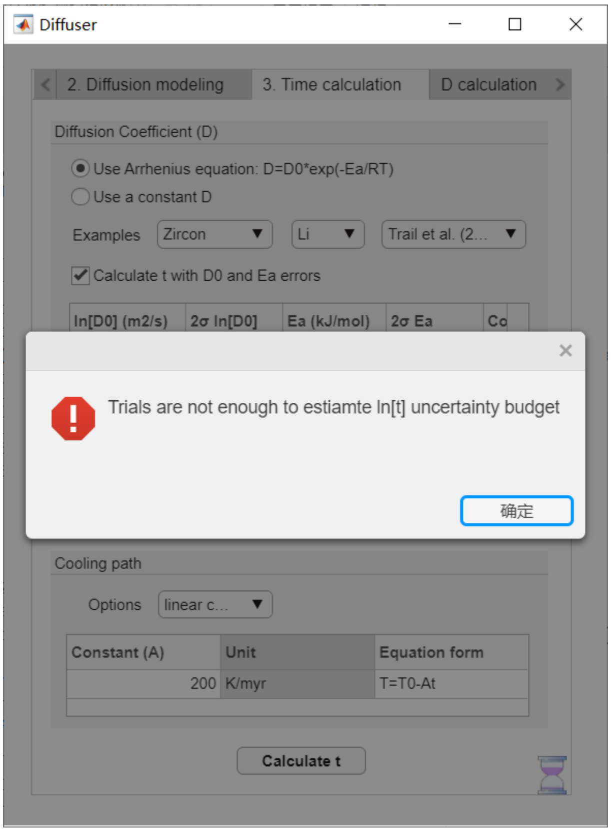

Figure 17. A dialog showing that trials of Monte Carlo are not enough for estimating the uncertainty budget.

	A	B	C	D	E	F	G	H	I	J	K
1	Trials	R2	C1	C2	x0 (m)	half band width (m)	log10(Dt)	2s	Timescale (year)	-2s	+2s
2	1	0.973650013	0	120	4.17501E-05	2.07095E-06	-12.10230331	0.100196679	22.45246895	-12.85626035	30.08008665

Figure 18. Timescale results in Diffuser.

	A	B	C	D	E	F
1	Temperature (Celciu	log10(Dt)=-12.1023				
2		t(s), normal distribution				
3	710.7161606	622360713.6				
4	697.5291488	931498663.9				
5	682.472005	1340934549				
6	711.1874759	433521960.5				
7	701.281457	401655661.9				
8	715.9695517	364050624.4				
9	710.0678741	419741180.7				
10	708.4771519	516501900.9				
11	696.5475219	837155248.1				
12	708.7940009	492472844.6				
13	696.0567776	1002808464				
14	702.7083621	841303567.1				
15	699.355481	910018888.6				

Figure 19. Monte Carlo results in Diffuser.

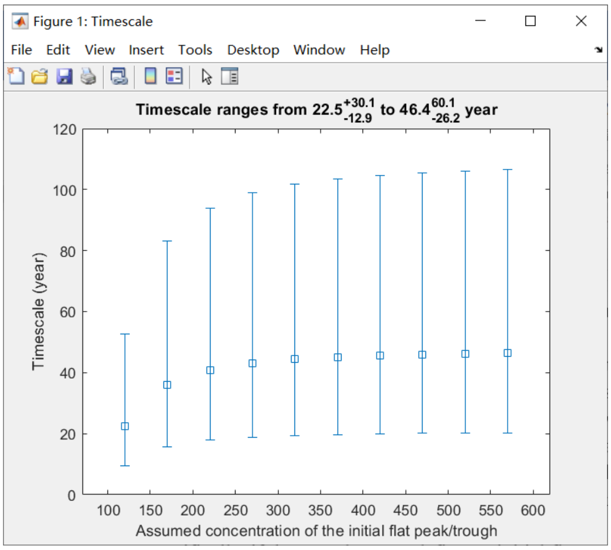

Figure 20. Timescales calculated for an assumed composition range of the flat peak/trough in profiles I–L.

5 Diffusivity calculation

To calculate diffusion coefficients in isothermal experimental studies, the user can input the **experimental duration** on the diffusivity calculation panel (Figure 21) and get an estimated D with its uncertainty (Figure 22). Then, the user can import all experimental data by the '**Select file**' button (D at different temperatures; Figure 21). Similar to data import of a diffusion profile, the measured D at different temperatures can be fed into the software through a delimited text (e.g., txt or csv format), spreadsheet file (Microsoft® Excel), or clipboard (by the '**clipboard**' button). The temperature and ln[D] data should be in two columns. If ln[D] uncertainties are assigned in another column, Diffuser will ask the user to input the column names of temperature, ln[D], and uncertainties. Otherwise, all ln[D] data will be treated with equal weights.
After data import, the user can calculate the parameters of ln[D_0] and E_a in the diffusion coefficient and their covariance by ordinary linear least-squares regression (Figure 23). The curve fitting result also can be saved in a file (Figure 24).
To play with Diffuser, the user can click the '**Load example**' button and Diffuser will

generate an example data of ln[D] at different temperatures. Then the user can deal with the example data in the same way. For clearing the table and plot, just click the '**Clear table**' button.

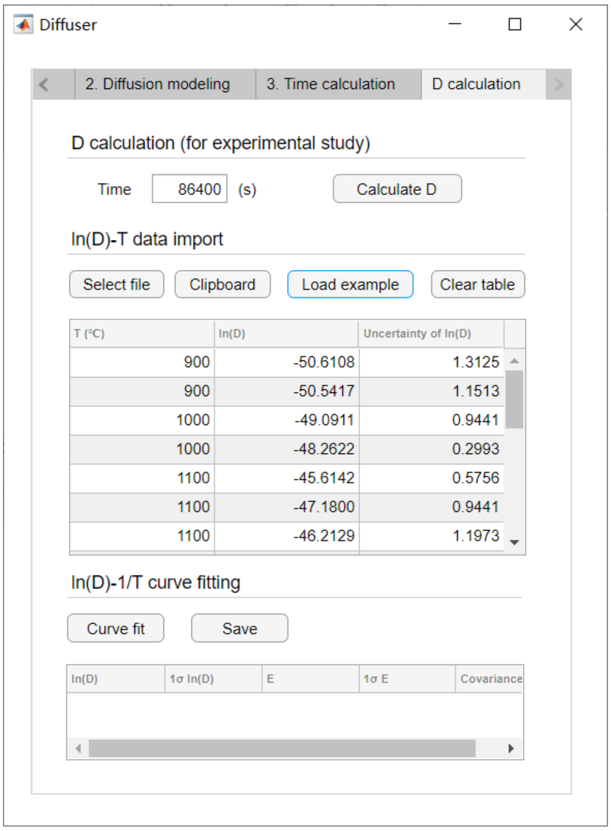

Figure 21. Diffusivity calculation panel of Diffuser

	A	B	C	D	E	F	G	H	I	J	K	L	M	N
1	Trials	R2	C1	C2	x0 (m)	half band width (m)	log10[Dt]	2s	log10[D]	2s	ln[D]	2s	D	2s
2	1	0.973650013	0	120	4.17501E-05	2.07095E-06	-12.10230331	0.10019668	-20.90213991	0.10019668	-48.12895576	0.230711379	1.25274E-21	2.89021E-22
3														

Figure 22. D calculation results in Diffuser.

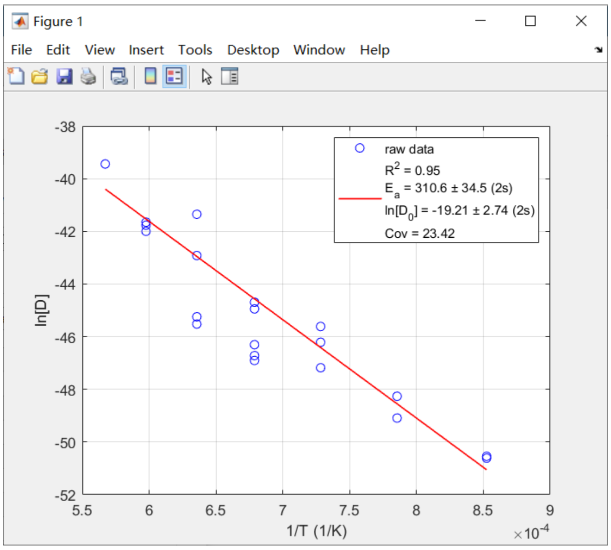

Figure 23. Curve fitting of ln[D] and 1/T.

	A	B	C	D	E
1	ln[D0] (m2/s)	2s ln[D0]	Ea (kJ/mol)	2s Ea	Covariance (ln[D0],Ea)
2	-19.2071108	2.74027404	310.6069714	34.48358696	23.42240413
3					

Figure 24. Curve fitting result of ln[D] and 1/T.

6 Web version

A web version of Diffuser is available at http://www.geoapp.cn/. It has nearly the same functions as the offline version. However, it cannot import data from the clipboard (thus the clipboard button is removed; Figure 25). It also cannot show pop-up windows, so instead of reminding the user to select the sheet and input column names of x, C, and σ (Figure 4), it imports data directly by reading the first sheet and its first two or three columns (Figure 2 and 3).

All the figures of the web version can be downloaded as portable document formats ('**Save figures**' button) so that the user can modify it easily offline.

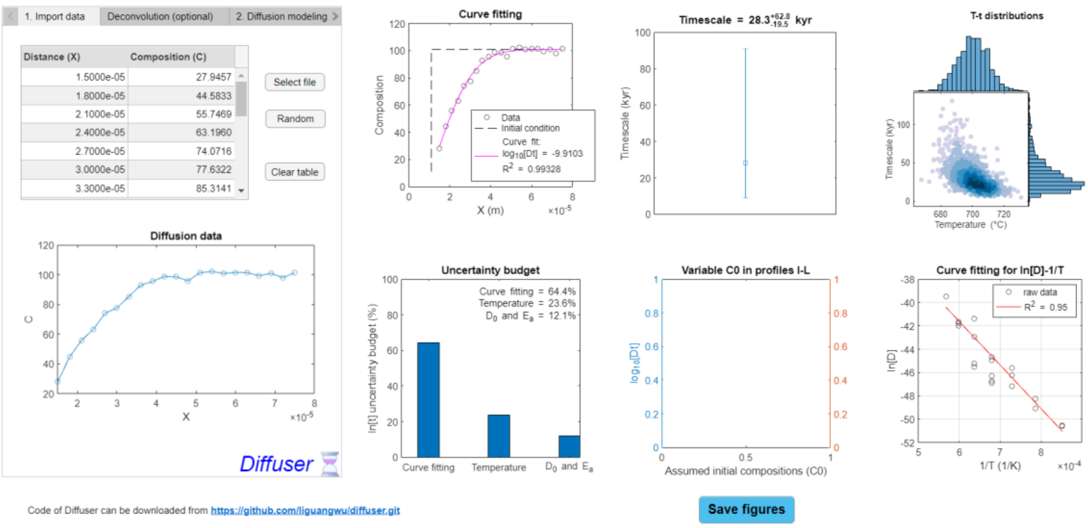

Figure 25. Web version of Diffuser.