

**Uranium isotope fractionation in anoxic settings and the global uranium isotope mass balance**

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## Introduction

This supplementary information contains a description of the uranium isotope mass balance model used in this study. Additional files include contextual data for Paleozoic samples, as well as all geochemical data generated for this study.

## S1. Uranium Isotope Mass Balance

As noted in the main text, the aim of the U isotope mass balance model used in this study was not to provide solution space for a specific U dataset, but instead to quantitatively demonstrate the potential solution space consistent with implementation of a ferruginous sink. The U isotope mass balance is described by the following two ordinary differential equations (ODEs; Eq. 1, 2) based on the mass balance of Dunk et al. (2002) and the isotope mass balance of Tissot and Dauphas (2015). The equations are solved using a basic ODE solver in Python from the SciPy.integrate package to achieve steady state. Steady state solutions for the parameter ranges explored (Table S1) were then represented in the heatmaps of main text Figures 8 and 9.

$$\frac{d[U]_{sw}}{dt} = J_{rv} - J_h - J_c - J_{an} - J_{so} - J_{ferr} - J_{oth} - J_{ox} \quad (1)$$

$$\begin{aligned} \frac{d [U]_{sw} \delta^{238}U_{sw}}{dt} &= (J_{rv} * \delta^{238}U_{rv}) - J_h(\delta^{238}U_{sw} + \Delta_h) - J_c(\delta^{238}U_{sw} + \Delta_c) \\ &- J_{an}(\delta^{238}U_{sw} + \Delta_{an}) - J_{so}(\delta^{238}U_{sw} + \Delta_{so}) - J_{ferr}(\delta^{238}U_{sw} + \Delta_{ferr}) \\ &- J_{oth}(\delta^{238}U_{sw} + \Delta_{oth}) - J_{ox}(\delta^{238}U_{sw} + \Delta_{ox}) \end{aligned} \quad (2)$$

Parameters for all variables excluding the ferruginous sink are from Dunk et al. (2002) and Tissot and Dauphas (2015) in order to represent the modern steady state U isotope mass balance. In this case, the ferruginous environments were set to zero. We then explored the effects of a range of ferruginous areas, fractionations, and efficiencies while keeping all other parameters constant (Fig. 8-9, main text).

Table S1. Parameters used in global U isotope mass balance model

<b>Variable</b>	<b>Value unit</b>	<b>Reference</b>
Riverine & groundwater flux ( $J_{rv}$ )	$5.13 \times 10^{16}$ <i>nmol/yr</i>	Dunk et al. (2002)
Riverine input ( $\delta^{238}U_{rv}$ )	-0.24 ‰	Tissot & Dauphas (2015)
Basalt alteration sink ( $J_h$ )	$5.7 \times 10^{15}$ <i>nmol/yr</i>	Dunk et al. (2002)
Carbonate sink ( $J_c$ )	$1.3 \times 10^{16}$ <i>nmol/yr</i>	Dunk et al. (2002)
Flux to anoxic (euxinic) environments ( $F_{an}$ )	0.15 <i>nmol/cm<sup>2</sup>yr</i>	Dunk et al. (2002)
Flux to suboxic environments ( $F_{so}$ )	0.1 <i>nmol/cm<sup>2</sup>yr</i>	Dunk et al. (2002)
Flux to ferruginous environments ( $F_{ferr}$ )	0.01 – 0.33 <i>nmol/cm<sup>2</sup>yr</i>	<i>This study, see main text for justification</i>
Flux to ( $F_{ox}$ )	0.02 <i>nmol/cm<sup>2</sup>yr</i>	Dunk et al. (2002)
Effective fractionation – Basalt alt. ( $\Delta_h$ )	0.1 ‰	Tissot & Dauphas (2015)
Effective fractionation – carbonates ( $\Delta_c$ )	0.2 ‰	Tissot & Dauphas (2015)
Effective fractionation – anoxic (euxinic) ( $\Delta_{an}$ )	0.6 ‰	Andersen et al. (2014)
Effective fractionation – suboxic ( $\Delta_{so}$ )	0.1 ‰	Tissot & Dauphas (2015)
Effective fractionation – ferruginous ( $\Delta_{ferr}$ )	-0.3 – 0.3 ‰	<i>This study, see main text for justification</i>
Effective fractionation – oxic/coastal retention ( $\Delta_{ox}$ )	-0.2 ‰	Tissot & Dauphas (2015)

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Sample Name	Height/Depth (m)	Locality	Interpreted Age	Period	Age	Lithology	Formation/Strat	Country	Reference
DL12		Loc.107	428	Silurian	Homerian	dark grey shale	Tamaghrou Formation	Morocco	LÜNING et al., 2000
DL11	96.2	El Pintado section 1	430	Silurian	Homerian	dark grey shale	Lower Graptolitic Shales	Spain	Robardet et al., 1998
DL20		Banwy River section	431	Silurian	Sheinwoodian	medium dark grey siltstone	Nant-ysgollon Shales Formation	Wales	Loydell and Cave, 1996
DL22		Banwy River section	432	Silurian	Sheinwoodian	dark grey shale	Nant-ysgollon Shales Formation	Wales	Loydell and Cave, 1996
DL25	1.23	Buttington	432	Silurian	Sheinwoodian	medium dark grey silty shale	Trewern Brook Mudstone Formation	Wales	Loydell and Large, 2019
DL26	8.53	Buttington	432	Silurian	Sheinwoodian	medium dark grey shale	Trewern Brook Mudstone Formation	Wales	Loydell and Large, 2019
DL28	911.52	Aizpute-41	432	Silurian	Sheinwoodian	medium grey silty shale	JÅ«rmala Formation	Latvia	Loydell et al., 2003
DL29	920	Aizpute-41	433	Silurian	Sheinwoodian	medium light grey silty shale	JÅ«rmala Formation	Latvia	Loydell et al., 2003
DL13		Loc.109	435	Silurian	Telychian	medium dark grey shale	Tamaghrou Formation	Morocco	LÜNING et al., 2000
DL31	942.45	Aizpute-41	436	Silurian	Telychian	medium grey shale	JÅ«rmala Formation	Latvia	Loydell et al., 2003
DL33	959.35	Aizpute-41	437	Silurian	Telychian	medium dark grey silty shale	JÅ«rmala Formation	Latvia	Loydell et al., 2003
DL47		Litohlavý	437	Silurian	Telychian	dark grey shale	Litohlavý Formation	Czech Republic	Kříž, 1992
DL14		Loc.100	439	Silurian	Aeronian	dark grey siltstone	Argillaceous shales with graptolites	Morocco	LÜNING et al., 2000
DL35	969.66	Aizpute-41	439	Silurian	Aeronian	medium dark grey silty shale	Dobeles Formation	Latvia	Loydell et al., 2003
DL44	46.62	BG-14 core	443	Silurian	Rhuddanian	light olive grey shale	Batra Formation	Jordan	Loydell, 2007
DL45	41.85	BG-14 core	443	Silurian	Rhuddanian	light olive grey shale	Batra Formation	Jordan	Loydell, 2007
DL19		Hartfell Score	450	Ordovician	Katian	dark grey shale	Lower Hartfell Shale Formation	Scotland	Zalasiewicz et al., 1995
DL17		Llanfawr Quarry, Llandrindod Wells	458	Ordovician	Sandbian	dark grey shale	Llanfawr Mudstones Formation	Wales	Hughes, 1989
DL40		Ogof Ddŷ»	483	Ordovician	Tremadocian	medium dark grey silty shale	Dol-cyn-afon Formation	Wales	Howells and Smith, 1997
LenzCP-35		Bathurst Island	420	Silurian	Pridoli	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-36		Bathurst Island	420	Silurian	Pridoli	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-37		Bathurst Island	420	Silurian	Pridoli	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-38		Bathurst Island	417	Devonian	Lochkovian	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-39		Bathurst Island	412.5	Devonian	Lochkovian	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-40		Bathurst Island	412.5	Devonian	Lochkovian	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-41		Bathurst Island	411.5	Devonian	Lochkovian	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-42		Bathurst Island	411.5	Devonian	Lochkovian	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-43		Bathurst Island	411.5	Devonian	Lochkovian	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-45		Bathurst Island	408	Devonian	Pragian	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-46		Bathurst Island	408	Devonian	Pragian	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013
LenzCP-48		Bathurst Island	408	Devonian	Pragian	calcareous shale	Cape Phillips Formation	Canada	Lenz, 2013



Locality	Ocean Depth (m)	Setting	Sample Code	U238peru	2SE (‰)	TOC wt%	Th (ppm)	U(ppm)	U/Th
Peru	86	Top edge OM	2MC-35	<b>-0.26</b>	0.04	6.36	6.74499498	3.42325503	0.508
Peru	96	Top edge OM	5MC-015	<b>-0.24</b>	0.05	7.03	5.28643202	6.43762155	1.218
Peru	102	Top edge OM	29MC-25	<b>-0.14</b>	0.06	7.89	6.31475307	4.21107944	0.667
Peru	115	Top edge OM	120MC-05	<b>-0.21</b>	0.04	9.78	4.56837363	14.2501264	3.119
Peru	321	OMZ	1MC-1375	<b>-0.27</b>	0.06	21.3	2.05614275	9.32790428	4.537
Peru	282	OMZ	8MC-075	<b>-0.21</b>	0.06	14.4	2.60420164	6.88774264	2.645
Peru	153	OMZ	45MC-075	<b>-0.26</b>	0.05	20.7	4.48023844	10.9791268	2.451
Peru	255	OMZ	18MC-05	<b>-0.22</b>	0.05	20.6	3.81591208	11.9493092	3.131
Peru	239	OMZ	71MC-075	<b>-0.17</b>	0.05	19.5	2.04754409	8.6285115	4.214
Peru	185	OMZ	104MC-05B	<b>-0.15</b>	0.05	16.8	3.06529586	6.53365816	2.131
Peru	364	OMZ	122MC-25	<b>-0.08</b>	0.06	20.6	2.31673625	12.9704055	5.599
Peru	654	below OMZ	14MC-05	<b>-0.34</b>	0.04	5.6	2.02507825	4.17069006	2.060
Peru	1357	below OMZ	33MC-05	<b>-0.30</b>	0.04	6.4	7.18340708	4.21999757	0.587
Peru	598	below OMZ	35MC-05	<b>-0.39</b>	0.06	6.2	5.41766049	17.6678409	3.261
Peru	1278	below OMZ	81MC-05	<b>-0.25</b>	0.05	3.1	13.7011574	11.1063179	0.811

Location	Setting	Samp Name	Depth of core (m)	Sed. Col. depth (cm)	$\delta^{238}\text{U}$ Bulk	2SE (‰)	Th (ppm)	U (ppm)	U/Th	(use min U/Th)	(use crust U/Th)	(use max U/Th)
										(Cole et al., 2017)	(Rudnick and Gao, 2014)	(Cole et al., 2017)
										$\delta^{238}\text{U}$ auth. Max	$\delta^{238}\text{U}$ auth. Avg.	$\delta^{238}\text{U}$ auth. Min
Brownie Lake (seds.)	Oxic	B2-1	3	0	-0.19	0.06	4.463	4.020	0.901	-0.19	-0.19	-0.19
Brownie Lake (seds.)	Oxic	B2-2	3	0.5	-0.17	0.04	2.435	3.833	1.574	-0.17	-0.17	-0.17
Brownie Lake (seds.)	Oxic	B2-3	3	1.5	-0.22	0.04	2.806	3.840	1.369	-0.22	-0.22	-0.22
Brownie Lake (seds.)	Oxic	B2-4	3	2.5	-0.24	0.04	2.491	3.794	1.523	-0.24	-0.24	-0.24
Brownie Lake (seds.)	Oxic	B2-5	3	4	-0.19	0.03	2.370	3.452	1.457	-0.19	-0.19	-0.19
Brownie Lake (seds.)	Oxic	B2-6	3	6	-0.18	0.05	3.762	3.036	0.807	-0.18	-0.18	-0.18
Brownie Lake (seds.)	Oxic	B2-7	3	8	-0.14	0.04	3.090	2.062	0.667	-0.14	-0.14	-0.14
Brownie Lake (seds.)	Oxic	B2-8	3	10	-0.16	0.04	2.795	1.673	0.599	-0.16	-0.16	-0.16
Brownie Lake (seds.)	Oxic	B2-9	3	12	-0.20	0.03	3.901	1.891	0.485	-0.20	-0.20	-0.20
Brownie Lake (seds.)	Oxic	B2-10	3	14	-0.18	0.04	2.334	1.932	0.828	-0.18	-0.18	-0.18
Brownie Lake (seds.)	Oxic	B2-11	3	16	-0.21	0.04	5.195	2.039	0.392	-0.21	-0.21	-0.21
Brownie Lake (seds.)	Oxic	B2-12	3	18	-0.14	0.03	2.536	1.583	0.624	-0.14	-0.14	-0.14
Brownie Lake (seds.)	Oxic	B2-13	3	20	-0.30	0.03	2.427	1.637	0.674	-0.30	-0.30	-0.30
Brownie Lake (seds.)	Oxic	B2-14	3	22	-0.31	0.04	2.564	1.500	0.585	-0.31	-0.31	-0.31
Brownie Lake (seds.)	Anoxic	B1-1	14	0	-0.26	0.04	3.855	2.268	0.588	-0.26	-0.26	-0.26
Brownie Lake (seds.)	Anoxic	B1-2	14	0.5	-0.27	0.04	21.223	2.324	0.109	-0.27	-0.27	-0.27
Brownie Lake (seds.)	Anoxic	B1-3	14	1.5	-0.32	0.03	5.207	2.340	0.449	-0.32	-0.32	-0.32
Brownie Lake (seds.)	Anoxic	B1-4	14	2.5	-0.29	0.04	4.673	2.226	0.476	-0.29	-0.29	-0.29
Brownie Lake (seds.)	Anoxic	B1-5	14	3.5	-0.23	0.04	3.145	2.147	0.682	-0.23	-0.23	-0.23
Brownie Lake (seds.)	Anoxic	B1-6	14	4.5	-0.29	0.04	2.492	1.937	0.777	-0.29	-0.29	-0.29
Brownie Lake (seds.)	Anoxic	B1-7	14	6.5	-0.27	0.04	3.212	1.938	0.603	-0.27	-0.27	-0.27
Brownie Lake (seds.)	Anoxic	B1-8	14	7.5	-0.19	0.03	20.752	2.252	0.109	-0.19	-0.19	-0.19
Brownie Lake (seds.)	Anoxic	B1-9	14	9	-0.27	0.03	5.076	2.202	0.434	-0.27	-0.27	-0.27
Brownie Lake (seds.)	Anoxic	B1-10	14	11	-0.15	0.04	3.362	2.177	0.648	-0.15	-0.15	-0.15
Brownie Lake (seds.)	Anoxic	B1-11	14	13	-0.22	0.04	5.110	2.002	0.392	-0.22	-0.22	-0.22
Brownie Lake (seds.)	Anoxic	B1-12	14	15	-0.11	0.03	3.306	2.133	0.645	-0.11	-0.11	-0.11
Brownie Lake (seds.)	Anoxic	B1-13	14	17	0.32	0.03	2.523	2.045	0.811	0.32	0.32	0.32
Brownie Lake (seds.)	Anoxic	B1-14	14	19	0.17	0.05	3.732	1.953	0.523	0.17	0.17	0.17
Brownie Lake (seds.)	Anoxic	B1-15	14	21	0.36	0.04	1.898	1.723	0.908	0.36	0.36	0.36
Brownie Lake (seds.)	Anoxic	B1-16	14	23	-0.21	0.04	2.773	1.808	0.652	-0.21	-0.21	-0.21
Brownie Lake (seds.)	Anoxic	B1-17	14	25	-0.21	0.04	5.018	1.854	0.370	-0.21	-0.21	-0.21
Brownie Lake (seds.)	Anoxic	B1-18	14	27	-0.23	0.03	2.549	1.885	0.740	-0.23	-0.23	-0.23
Brownie Lake (seds.)	Anoxic	B1-19	14	29	-0.25	0.04	3.079	1.853	0.602	-0.25	-0.25	-0.25
Brownie Lake (seds.)	Anoxic	B1-20	14	31	-0.13	0.03	3.548	1.762	0.497	-0.13	-0.13	-0.13
Brownie Lake (seds.)	Anoxic	B1-21	14	33	-0.08	0.04	3.518	1.991	0.566	-0.08	-0.08	-0.08
Brownie Lake (seds.)	Anoxic	B1-22	14	35	-0.11	0.04	3.212	2.094	0.652	-0.11	-0.11	-0.11
Brownie Lake (seds.)	Anoxic	B1-23	14	37	-0.24	0.04	3.544	2.162	0.610	-0.24	-0.24	-0.24
Brownie Lake (seds.)	Anoxic	B1-24	14	40	-0.26	0.03	3.169	2.156	0.680	-0.26	-0.26	-0.26
Lake Pavin	oxic	C3-3-1	32	0	-0.28	0.05	1.296	0.309	0.239	-0.28	-0.28	-0.28
Lake Pavin	oxic	C3-4-1	32	3.5	-0.34	0.04	1.446	0.380	0.263	-0.34	-0.34	-0.34
Lake Pavin	oxic	C3-5-1	32	4.5	-0.40	0.05	1.666	0.390	0.234	-0.40	-0.40	-0.40
Lake Pavin	oxic	C3-6-1	32	5.5	-0.42	0.03	2.567	0.583	0.227	-0.42	-0.42	-0.42
Lake Pavin	oxic	C3-7-1	32	6.5	-0.43	0.04	2.738	0.606	0.221	-0.43	-0.43	-0.43
Lake Pavin	oxic	C3-8-1	32	7.5	-0.37	0.04	4.272	0.900	0.211	-0.37	-0.37	-0.37
Lake Pavin	oxic	C3-9-1	32	10	-0.38	0.04	4.189	0.855	0.204	-0.38	-0.38	-0.38
Lake Pavin	chemocline	D-2	60	2.75	-0.11	0.04	0.537	0.249	0.464	-0.11	-0.11	-0.11
Lake Pavin	chemocline	F-2	60	4.3	-0.20	0.04	0.638	0.281	0.440	-0.20	-0.20	-0.20
Lake Pavin	chemocline	H-2	60	6.3	-0.35	0.03	0.821	0.345	0.421	-0.35	-0.35	-0.35
Lake Pavin	chemocline	I-2	60	9	-0.52	0.04	1.822	0.513	0.281	-0.52	-0.52	-0.52
Lake Pavin	chemocline	L-2	60	12.75	-0.53	0.04	2.283	0.551	0.241	-0.53	-0.53	-0.53
Lake Pavin	chemocline	C2-2-1	65	1.5	-0.24	0.05	0.585	0.265	0.453	-0.24	-0.24	-0.24



Location	Setting	Samp Name	Depth of core (m)	Sed. Col. depth (cm)	$\delta^{238}\text{U}$ Bulk	2SE (%)	Th (ppm)	U (ppm)	U/Th	(use min U/Th)	(use crust U/Th)	(use max U/Th)
										<i>(Cole et al., 2017)</i>	<i>(Rudnick and Gao, 2014)</i>	<i>(Cole et al., 2017)</i>
										$\delta^{238}\text{U}$ auth. Max	$\delta^{238}\text{U}$ auth. Avg.	$\delta^{238}\text{U}$ auth. Min
Lake Pavin	chemocline	C2-3-1	65	2.5	-0.27	0.03	0.605	0.283	0.468	-0.27	-0.27	-0.27
Lake Pavin	chemocline	C2-4-1	65	3.5	-0.29	0.06	0.590	0.259	0.439	-0.29	-0.29	-0.29
Lake Pavin	chemocline	C2-6-1	65	5.5	-0.26	0.04	0.712	0.312	0.438	-0.26	-0.26	-0.26
Lake Pavin	chemocline	C2-7-1	65	6.5	-0.04	0.04	0.891	0.404	0.453	-0.04	-0.04	-0.04
Lake Pavin	chemocline	C2-8-1	65	7.5	-0.28	0.05	0.700	0.307	0.439	-0.28	-0.28	-0.28
Lake Pavin	chemocline	C2-9-1	65	8.5	-0.52	0.04	0.482	0.218	0.454	-0.52	-0.52	-0.52
Lake Pavin	anoxic	LPC1-1	92	3.5	-0.28	0.04	1.983	0.629	0.317	-0.28	-0.28	-0.28
Lake Pavin	anoxic	LPC1-3	92	8.5	-0.45	0.04	1.150	0.596	0.519	-0.45	-0.45	-0.45
Lake Pavin	anoxic	LPC1-4	92	11.5	-0.11	0.04	1.518	0.830	0.547	-0.11	-0.11	-0.11
Lake Pavin	anoxic	LPC1-6	92	16.5	-0.16	0.05	2.575	1.026	0.399	-0.16	-0.16	-0.16
Lake Pavin	anoxic	LPC1-7	92	18.5	-0.07	0.04	1.640	0.950	0.579	-0.07	-0.07	-0.07
Lake Pavin	anoxic	LPC1-8	92	21.5	0.01	0.05	1.297	0.858	0.661	0.01	0.01	0.01
Lake Pavin	anoxic	LPC1-11	92	28.5	-0.11	0.04	1.930	1.212	0.628	-0.11	-0.11	-0.11

Location	Sample Name	Water Depth (m)	Temp (°C)*	pH	Dissolved O2(mg/L)*	Sulfate (μmol)*	DIC (mM)*	$\delta^{238}\text{U}$ Bulk	2SE (%)	U (ppb)	Dissolved Fe (μmol)
Brownie Lake (waters)		0	17.3	8.81	10.78						
Brownie Lake (waters)	009BW-1	1	17.39	8.85	10.84		1.6	-0.11	0.06	0.71	0.979
Brownie Lake (waters)	011BW-2	2	16.24	8.04	8.12		2.2	-0.15	0.08	1.08	1.351
Brownie Lake (waters)	012BW-3	3	14.05	7.67	3.75	49.4	2.4	-0.25	0.06	1.15	1.459
Brownie Lake (waters)	013BW-3.5	3.5	13.2	7.54	1.37	50.6	2.5	-0.16	0.07	1.17	1.504
Brownie Lake (waters)	015BW-4	4	12.4	7.49	0.6	55.7	2.8	-0.11	0.06	1.26	3.02
Brownie Lake (waters)	016BW-4.5	4.5	10.97	7.5	0.4	58.8	3.1	-0.09	0.06	1.37	1.975
Brownie Lake (waters)	017BW-5	5	9.03	7.51	0.15	67.1	3.2	-0.17	0.04	1.34	1.317
Brownie Lake (waters)	019BW-5.5	5.5	7.61	7.38	0.14	61.6	3.7	-0.24	0.05	1.31	5.691
Brownie Lake (waters)	020BW-6	6	6.84	7.2	0.13	40.7	4.4	-0.22	0.04	1.24	8.073
Brownie Lake (waters)		6.5	6.71	7.08	0.13						
Brownie Lake (waters)	021BW-7	7	6.71	7.02	0.13	40.8	7.1	-0.52	0.05	0.36	363.762
Brownie Lake (waters)		8	6.9	6.96	0.12		8.8				542.052
Brownie Lake (waters)	023BW-9	9	7.05	6.91	0.12		9.7	-0.57	0.05	0.45	721.417
Brownie Lake (waters)	024BW-10	10	7.14	6.87	0.11		10.6	-0.43	0.05	0.44	1045.052
Brownie Lake (waters)	025BW-11	11	7.19	6.86	0.11		12.5	-0.47	0.04	0.53	1226.852
Brownie Lake (waters)	027BW-12	12	7.22	6.85	0.11		12.3	-0.26	0.04	0.46	1266.066
Brownie Lake (waters)	028BW-13	13	7.26	6.85	0.11		13.2	-0.38	0.05	0.43	
Brownie Lake (waters)		14	7.3	6.86	0.11						

\* published in Lambrecht et al., 2018