

Reference value of the JNdi-1 isotopic material without normalization

Alexandre Quemet¹, Guillaume Lasnier¹, Sébastien Mialle², Hélène Isnard², Maud Boyet³, Marion Garçon³ and Delphine Auclair³

¹ CEA, DES, ISEC, DRMC, Univ Montpellier, Marcoule, France

² Université Paris-Saclay, CEA, Service de Physico-Chimie, 91191, Gif-sur-Yvette, France

³ Université Clermont Auvergne, CNRS, IRD, OPGC, Laboratoire Magmas et Volcans, F-63000 Clermont-Ferrand, France

E-mail address of the corresponding author: alexandre.quemet@cea.fr

Isotopic measurement

LMV protocol

For a better reproducibility of ionization conditions, only double rhenium filaments (99.999% of purity - quality Zone Refined) made by ThermoFischer Scientific were used. Filaments were outgassed for 30 minutes at 4500 mA under high vacuum using the ThermoFischer Scientific vacuum chamber. The loading procedures described below were performed under a laminar flow hood. A diluted standard solution of JNdi-1 mixed with H_3PO_4 was prepared just before the analytical sessions, so all solutions loaded on the filaments have strictly the same element/ H_3PO_4 ratio. The Nd concentration of the original JNdi-1 solution has been accurately determined by ICP-MS before loading to know the exact amount of Nd loaded on the filament. To limit the formation of oxides that would interfere during analysis, the JNdi-1 solution is loaded in free drying, *i.e.* without circulating an electric current in the filament. This technique helps to reduce (or even eliminate) the formation of oxides during measurements as described in [1]. In this study 0.5 μL of solution (*i.e.* 25 ng) is deposited in the center of the evaporation filament in only one drop, using a Hamilton syringe to have a better control on the volume loaded. With this loading method, a unique clump was obtained. This eliminates potential mixing effects than can be seen when loading several drops that can form different evaporation domains on the filament [2]. Fifteen to thirty minutes are necessary to completely dry the loading when using the “free drying” technique.

Two mass spectrometers for the analyses were used: a ThermoFischer Scientific Triton and a ThermoFischer Scientific TritonPlus. The total evaporation procedure included in the Method Editor section of the Triton’s software has been used. This method ensures a better reproducibility of the heating protocol and intensity peak shape. First, a current ramp rate of 250 mA min^{-1} was applied to the ionization filament to reach a final value of 4800 mA corresponding to a temperature between 1700°C and 1800°C. Baselines were measured during the filaments' heating (600 s) to maximize the signal acquisitions. A first focalization of lenses was made using the ^{187}Re ion beam. The evaporation filament was then heated using a rate of 50 mA min^{-1} and stopped when the ion beam of the pilot signal

reached 2 mV. A second focalization of the lenses was performed, immediately followed by a peak center, both of them based on the ion beam of the pilot signal. The current on the evaporation filament was automatically adjusted by the software to obtain and maintain the target intensity. A typical run for 25ng of Nd took approximately 35 minutes. The two isobaric interferences (^{140}Ce and ^{147}Sm) were monitored but no interference correction was necessary because the analyzed JNdi-1 solutions were pure enough to make these corrections completely negligible.

LANIE protocol

JNdi-1 solutions at around $10\text{ ng }\mu\text{L}^{-1}$ and $40\text{ ng }\mu\text{L}^{-1}$ were freshly prepared from laboratory stock solutions and ultrapur HNO_3 0.5 mol L^{-1} (Plasma PurePlus, SCP sciences). These solutions have been quantified using an ICAP RQ ICP-MS (ThermoFischer Scientific, Bremen, Germany) using the external calibration curve method. The absence of cerium has also been verified. The obtained concentrations were respectively $10.4 \pm 0.3\text{ }\mu\text{g }\mu\text{L}^{-1}$ and $41.3 \pm 1.2\text{ }\mu\text{g }\mu\text{L}^{-1}$.

A $1\text{ }\mu\text{L}$ droplet of those solutions is slowly deposited on a 99.99 % pure rhenium filaments provided by ATES (Clapiers, France) and dried with a 0.3 - 0.4 A current. Once the droplet dried, the current is progressively increased up to 2 A for few seconds. Rhenium filaments are beforehand outgassed. A double Re-filament configuration was used.

Measurements were performed on a Triton Plus TIMS (ThermoFischer Scientific, Bremen, Germany) equipped with 9 Faradays and 4 ion counters. The latest are not used in this article. The source is surrounded by a glovebox to analyze radioactive samples. Faraday cups are connected to 10^{11} Ohm amplifiers in this study. The cup configuration and the integration time is given in the table S1. Amplifier intercalibration gains were done before each turret analysis and the electronic baselines are measured at the beginning of the method.

Two different methods have been used (one for the 10 ng samples and one for the 40 ng) but only few parameters changed. In both method, ionization filaments are firstly heated

up at 4.5 A in 2 min. This current is slowly increased in order to reach a ^{187}Re signal of 20 mV. After ion beam optimization (peak center and focus) on ^{187}Re , the signal is increased up to reach an ^{187}Re signal of 80 mV. A second signal optimization is then performed. Then the evaporation filament is heated to reach a cumulated signal on the isotopes ^{143}Nd , ^{144}Nd , ^{145}Nd , ^{146}Nd , ^{148}Nd and ^{150}Nd of 10 mV. A signal optimization is performed and the evaporation filament current is increased up to reach a neodymium cumulated signal of 50 mV. After a brief optimization, the TE analysis starts.

For the 10 ng and 40 ng samples, a maximum pilot signal (signal plateau) of 3 and 5 V respectively is with a heat slope of 50 mA/ cycle. This pilot signal is the cumulated signal of the ^{143}Nd , ^{144}Nd , ^{145}Nd , ^{146}Nd , ^{148}Nd and ^{150}Nd isotopes. The measurement is stopped when the signal is below 50 mA or if the evaporation filament reaches 4.5 A. A measurement is considered as valid if the signal is above 100 V on the ^{144}Nd for the 40 ng samples and 50 V for on the ^{144}Nd for the 10 ng samples.

Measurements were realized in different analytical sessions between April 2023 and July 2023.

Cups	L3	L2	L1	C	H1	H2	H3	integration time (s)	idle time (s)
Amplifiers (Ω)	10^{11}	10^{11}	10^{11}	10^{11}	10^{11}	10^{11}	10^{11}	4 s	0 s
Isotope	^{142}Nd	^{143}Nd	^{144}Nd	^{145}Nd	^{146}Nd	^{148}Nd	^{150}Nd		

Table S1: Cup configuration and the integration time used by the LANIE laboratory

LAAT protocol

The TIMS and the measurement method were previously described in detail [3]. Briefly, the measurements were performed on a ThermoFischer Scientific Triton TIMS equipped with 9 Faraday cups coupled to $10^{11} \Omega$ current amplifiers. Intercalibration gains of the Faraday cup detectors were measured daily by an automated process of the TIMS software. The baselines were measured every half a day during 20 min.

A part of the JNdi-1 powder (split 1 vial 134) was dissolved in 0.5 mol L⁻¹ HNO₃ and diluted to obtain solutions with [Nd] ≈ 10, 50 and 100 ng μL⁻¹.

A double Re-filament configuration was used to control independently the evaporation and the ionization filament temperature. These filaments (Re metal, purity 99.99% or 99.999%) are provided by ATES and were outgassed 20 min at 4.5 A in a ThermoFischer Scientific vacuum chamber (<5 × 10⁻⁶ mbar). 1 μL JNdi-1 solution was deposited on the filament and dried with a 0.4 A current. After drying, the current was increased progressively to 2 A in 10 s.

The TE method used by the LAAT laboratory was previously described [3]. Briefly, in the first step, the ionization and evaporation filaments current is increased. After ion beam optimization the data acquisition is started. The evaporation filament current is increased until the ion beam intensity of the sum of all measured isotopes reaches the target intensity: about 38 V, 21 V and 8 V for 100 ng, 50 ng and 10 ng, respectively. When the evaporation filament current reaches a maximum value of 6.5 A and the ion beam decreases down to a 25 mV signal, the data acquisition is finished.

During Nd measurements, masses 140 (¹⁴⁰Ce) and 147 (¹⁴⁷Sm) were measured to monitor any possible contamination. A maximum ¹⁴⁰Ce/¹⁴⁴Nd ratio of 4×10⁻⁴ and a maximum ¹⁴⁷Sm/¹⁴⁴Nd ratio of 9×10⁻⁶ were measured indicating possible Ce and Sm contamination are negligible. No mathematical correction for the Ce or for the Sm isobaric interferences was performed for ¹⁴²Nd, ¹⁴⁴Nd, ¹⁴⁸Nd and ¹⁵⁰Nd isotope measurements.

The measurement were performed in different sections over a 3-year period by two analysts.

Nd isotope ratio of the JNdi-1 without normalization

Laboratory	Date	TIMS	Quantity (ng)	Integration time (s)	$^{142}\text{Nd}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}$	$^{145}\text{Nd}/^{144}\text{Nd}$	$^{146}\text{Nd}/^{144}\text{Nd}$	$^{148}\text{Nd}/^{144}\text{Nd}$	$^{150}\text{Nd}/^{144}\text{Nd}$
LMV	21/11/01	Triton +	25	2	1.13962	0.511611	0.348720	0.72331	0.24252	0.23783
LMV	21/11/01	Triton +	25	2	1.13963	0.511615	0.348716	0.72330	0.24251	0.23782
LMV	21/11/01	Triton +	25	2	1.13954	0.511593	0.348738	0.72336	0.24255	0.23787
LMV	21/11/01	Triton +	25	2	1.13956	0.511596	0.348735	0.72334	0.24254	0.23785
LMV	21/11/01	Triton +	25	2	1.13970	0.511626	0.348710	0.72326	0.24249	0.23778
LMV	21/11/01	Triton +	25	2	-	0.511646	0.348706	0.72317	0.24243	0.23770
LMV	21/11/01	Triton +	25	2	-	0.511571	0.348758	0.72338	0.24257	0.23790
LMV	21/11/01	Triton +	25	2	-	0.511611	0.348732	0.72327	0.24250	0.23779
LMV	21/11/01	Triton +	25	2	1.13991	0.511667	0.348696	0.72312	0.24239	0.23764
LMV	21/11/01	Triton +	25	2	1.13983	0.511650	0.348706	0.72316	0.24242	0.23768
LMV	22/07/01	Triton	25	2	1.13982	0.511648	0.348691	0.72318	0.24243	0.23769
LMV	22/07/01	Triton	25	2	1.14002	0.511695	0.348677	0.72305	0.24234	0.23757
LMV	22/07/01	Triton	25	2	1.13969	0.511628	0.348704	0.72328	0.24249	0.23778

Laboratory	Date	TIMS	Quantity (ng)	Integration time (s)	$^{142}\text{Nd}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}$	$^{145}\text{Nd}/^{144}\text{Nd}$	$^{146}\text{Nd}/^{144}\text{Nd}$	$^{148}\text{Nd}/^{144}\text{Nd}$	$^{150}\text{Nd}/^{144}\text{Nd}$
LMV	22/07/01	Triton	25	2	1.13963	0.511613	0.348728	0.72330	0.24251	0.23781
LMV	22/07/01	Triton	25	2	1.13970	0.511626	0.348703	0.72327	0.24249	0.23778
LANIE	23/04/17	Triton +	10	4	1.13948	0.511581	0.348752	0.72336	0.24255	0.23787
LANIE	23/04/17	Triton +	10	4	1.13960	0.511607	0.348749	0.72329	0.24252	0.23781
LANIE	23/04/17	Triton +	41	4	1.13956	0.511590	0.348747	0.72331	0.24253	0.23784
LANIE	23/04/18	Triton +	10	4	1.13911	0.511464	0.348808	0.72357	0.24272	0.23810
LANIE	23/04/18	Triton +	10	4	1.14000	0.511713	0.348671	0.72304	0.24235	0.23755
LANIE	23/04/18	Triton +	41	4	1.13962	0.511609	0.348749	0.72330	0.24251	0.23781
LANIE	23/04/27	Triton +	10	4	1.13978	0.511649	0.348730	0.72319	0.24243	0.23771
LANIE	23/04/27	Triton +	10	4	1.14003	0.511695	0.348680	0.72304	0.24234	0.23756
LANIE	23/04/27	Triton +	41	4	1.13995	0.511684	0.348692	0.72308	0.24236	0.23760
LANIE	23/04/27	Triton +	41	4	1.13992	0.511684	0.348701	0.72312	0.24240	0.23763
LANIE	23/05/04	Triton +	10	4	1.13987	0.511658	0.348694	0.72312	0.24240	0.23765
LANIE	23/05/04	Triton +	10	4	1.13976	0.511644	0.348710	0.72320	0.24244	0.23772
LANIE	23/05/04	Triton +	41	4	1.13990	0.511678	0.348690	0.72311	0.24238	0.23763

Laboratory	Date	TIMS	Quantity (ng)	Integration time (s)	$^{142}\text{Nd}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}$	$^{145}\text{Nd}/^{144}\text{Nd}$	$^{146}\text{Nd}/^{144}\text{Nd}$	$^{148}\text{Nd}/^{144}\text{Nd}$	$^{150}\text{Nd}/^{144}\text{Nd}$
LANIE	23/05/04	Triton +	41	4	1.13997	0.511692	0.348691	0.72306	0.24234	0.23758
LANIE	23/06/10	Triton +	10	4	1.13959	0.511598	0.348758	0.72334	0.24252	0.23785
LANIE	23/06/10	Triton +	10	4	1.13944	0.511561	0.348750	0.72339	0.24257	0.23790
LANIE	23/06/10	Triton +	41	4	1.14006	0.511712	0.348668	0.72301	0.24232	0.23754
LANIE	23/06/10	Triton +	41	4	1.13972	0.511629	0.348719	0.72322	0.24245	0.23774
LAAT	20/02/28	Triton	100	0.1	1.13922	0.511507	0.348800	0.72355	0.24268	0.23806
LAAT	20/02/28	Triton	100	0.1	1.13972	0.511623	0.348724	0.72324	0.24247	0.23775
LAAT	20/02/28	Triton	100	0.1	1.13977	0.511632	0.348716	0.72320	0.24245	0.23772
LAAT	20/02/28	Triton	100	0.1	1.13953	0.511580	0.348751	0.72334	0.24254	0.23786
LAAT	20/02/28	Triton	100	0.1	1.13961	0.511602	0.348750	0.72332	0.24253	0.23783
LAAT	20/02/28	Triton	10	0.1	1.13953	0.511582	0.348760	0.72335	0.24255	0.23787
LAAT	20/02/28	Triton	10	0.1	1.13947	0.511563	0.348789	0.72345	0.24260	0.23795
LAAT	20/02/28	Triton	10	0.1	1.13958	0.511582	0.348745	0.72335	0.24254	0.23786
LAAT	20/03/02	Triton	10	0.1	1.13950	0.511573	0.348766	0.72339	0.24258	0.23791
LAAT	20/03/02	Triton	10	0.1	1.13982	0.511644	0.348723	0.72323	0.24247	0.23774

Laboratory	Date	TIMS	Quantity (ng)	Integration time (s)	$^{142}\text{Nd}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}$	$^{145}\text{Nd}/^{144}\text{Nd}$	$^{146}\text{Nd}/^{144}\text{Nd}$	$^{148}\text{Nd}/^{144}\text{Nd}$	$^{150}\text{Nd}/^{144}\text{Nd}$
LAAT	20/06/02	Triton	10	0.1	1.13919	0.511506	0.348789	0.72357	0.24269	0.23809
LAAT	20/07/01	Triton	10	0.1	1.13908	0.511481	0.348828	0.72366	0.24275	0.23818
LAAT	21/12/09	Triton	100	1	1.13962	0.511609	0.348741	0.72329	0.24251	0.23781
LAAT	21/12/09	Triton	100	2	1.13934	0.511547	0.348786	0.72346	0.24262	0.23798
LAAT	21/12/10	Triton	100	4	1.13928	0.511528	0.348789	0.72350	0.24264	0.23801
LAAT	22/10/14	Triton	100	2	1.13940	0.511561	0.348774	0.72343	0.24260	0.23795
LAAT	22/10/19	Triton	100	2	1.13987	0.511662	0.348712	0.72316	0.24243	0.23769
LAAT	22/10/19	Triton	100	2	1.13978	0.511643	0.348722	0.72321	0.24246	0.23773
LAAT	22/10/19	Triton	100	2	1.13954	0.511592	0.348762	0.72337	0.24256	0.23788
LAAT	22/10/19	Triton	100	2	1.13943	0.511564	0.348784	0.72344	0.24261	0.23795
LAAT	23/02/27	Triton	50	1	1.13951	0.511590	0.348772	0.72340	0.24258	0.23790
LAAT	23/03/01	Triton	50	1	1.13956	0.511589	0.348761	0.72336	0.24254	0.23787
LAAT	23/03/08	Triton	50	1	1.13952	0.511584	0.348776	0.72339	0.24256	0.23790
LAAT	23/03/24	Triton	50	1	1.13951	0.511589	0.348761	0.72339	0.24257	0.23789
LAAT	23/03/27	Triton	50	1	1.13912	0.511493	0.348806	0.72359	0.24271	0.23811

Laboratory	Date	TIMS	Quantity (ng)	Integration time (s)	$^{142}\text{Nd}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}$	$^{145}\text{Nd}/^{144}\text{Nd}$	$^{146}\text{Nd}/^{144}\text{Nd}$	$^{148}\text{Nd}/^{144}\text{Nd}$	$^{150}\text{Nd}/^{144}\text{Nd}$
LAAT	23/03/28	Triton	50	1	1.13931	0.511544	0.348799	0.72352	0.24265	0.23803
LAAT	23/03/30	Triton	50	1	1.13894	0.511462	0.348843	0.72372	0.24280	0.23823
LAAT	23/03/31	Triton	50	1	1.13925	0.511524	0.348797	0.72352	0.24265	0.23802

Table S2: Individual Nd isotope ratio measurement for the JNdi-1 obtained by the LMV, LANIE and LAAT laboratories

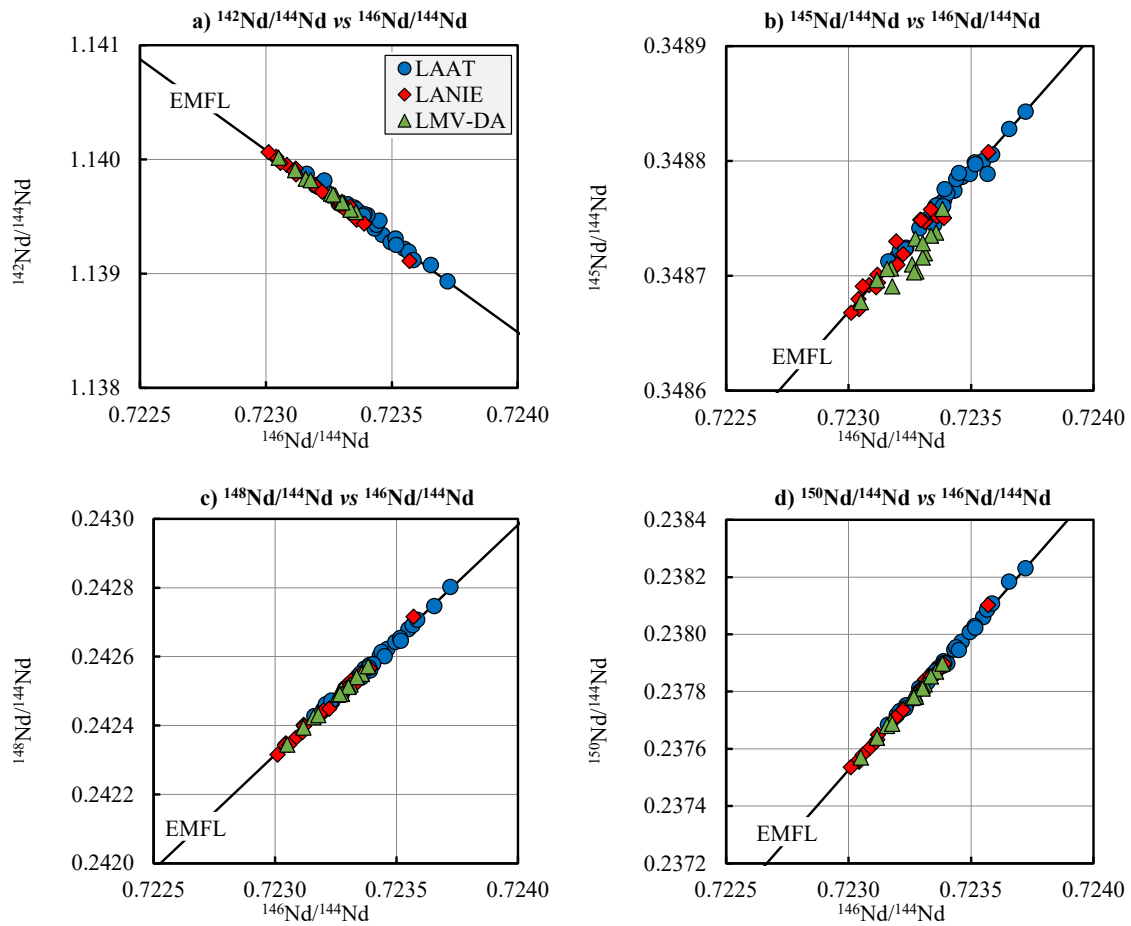


Fig. S1: $^X\text{Nd}/^{144}\text{Nd}$ ($X = 142, 145, 148, 150$) plotted against the $^{146}\text{Nd}/^{144}\text{Nd}$ ratio (figure a to d, respectively) for the LAAT (blue circle), LANIE (red diamond) and LMV (green triangle) laboratories. The solid line represents the Exponential Mass Fractionation Law calculated with the reference values given by Garçon *et al.* [14]

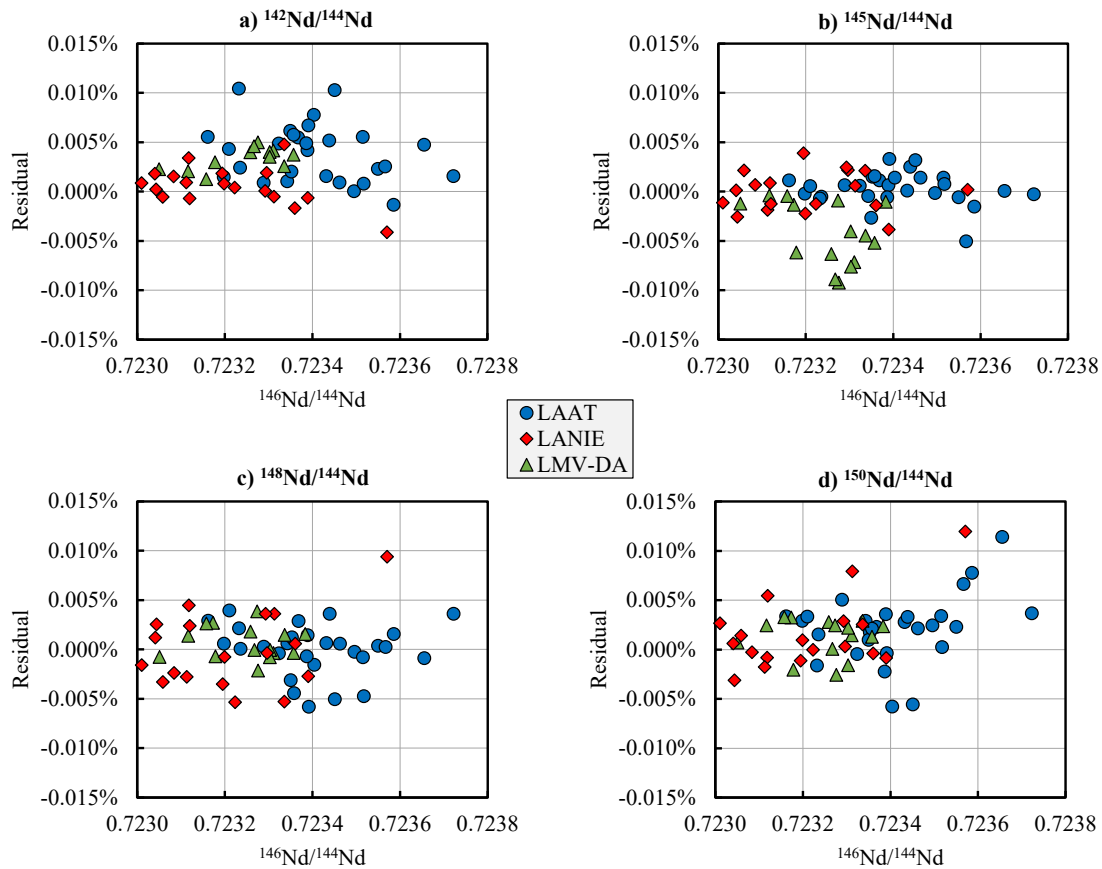


Fig. S2: Vertical deviation (residual) of the $^X\text{Nd}/^{144}\text{Nd}$ ($X = 142, 145, 148, 150$) isotope ratios from the EMFL curve as function of the $^{146}\text{Nd}/^{144}\text{Nd}$ isotope ratio (figure a to d, respectively) for the LAAT (blue circle), LANIE (red diamond) and LMV (green triangle) laboratories.

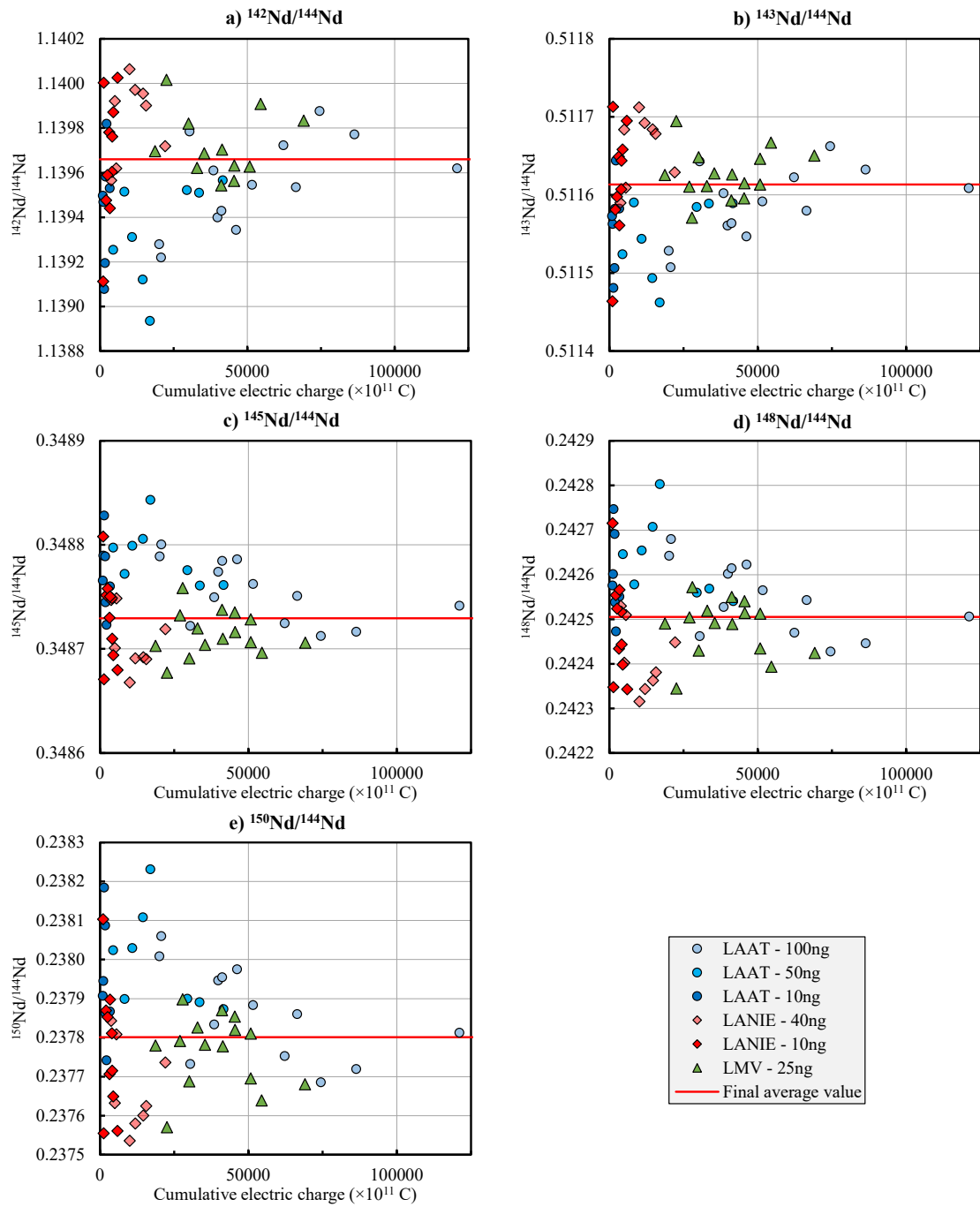


Fig. S3: $^X\text{Nd}/^{144}\text{Nd}$ ($X = 142, 143, 145, 148, 150$) isotope ratio as function of the cumulative electric charge on all Nd isotopes (figure a to e, respectively) by the LAAT (blue circle), LANIE (red diamond) and LMV (green triangle) laboratories and for different quantities

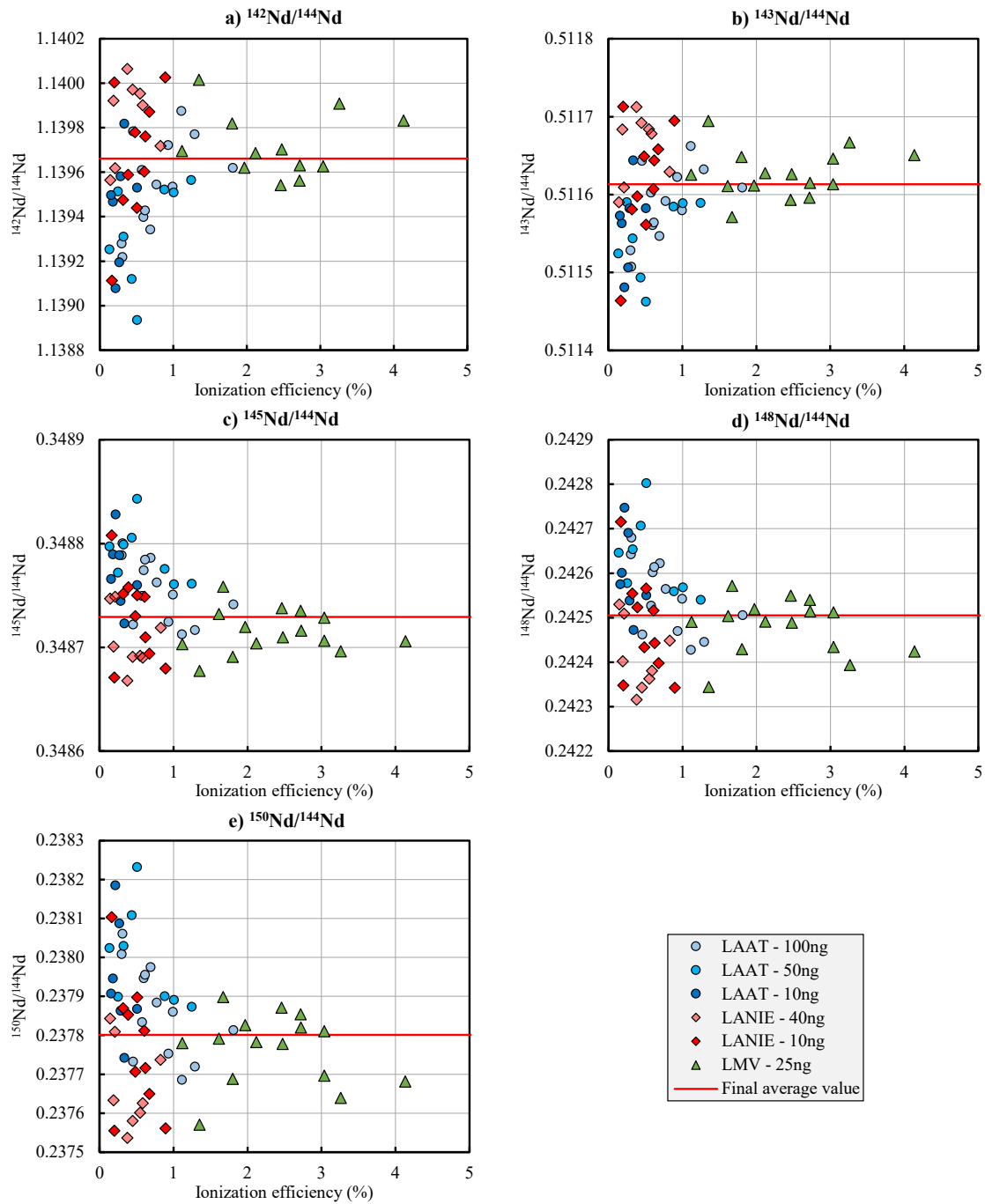


Fig. S4: $^X\text{Nd}/^{144}\text{Nd}$ ($X = 142, 143, 145, 148, 150$) isotope ratio as function of the cumulative electric charge on all Nd isotopes (figure a to e, respectively) by the LAAT (blue circle), LANIE (red diamond) and LMV (green triangle) laboratories and for different quantities

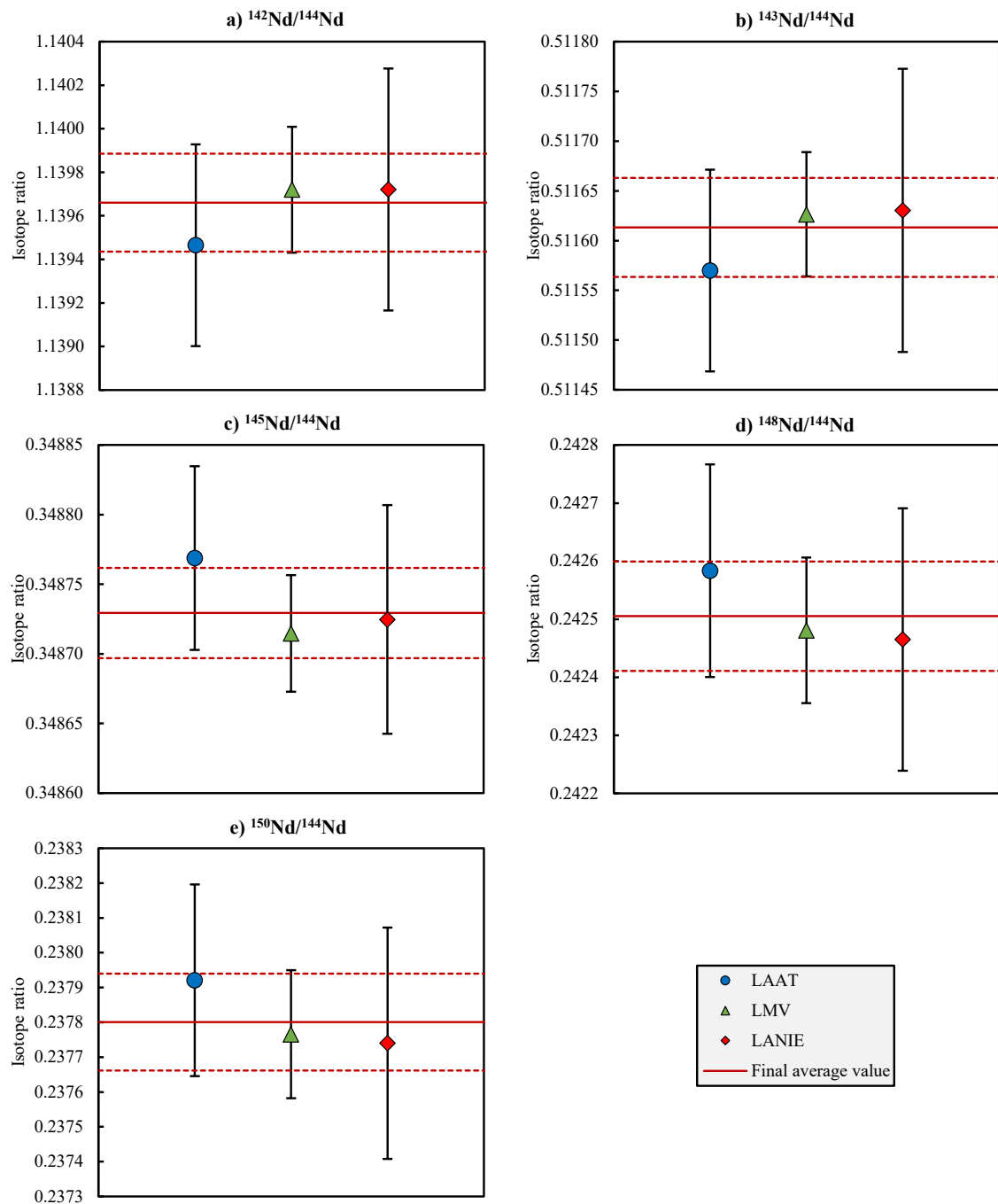


Fig. S5: $^X\text{Nd}/^{144}\text{Nd}$ ($X = 142, 143, 145, 148, 150$) isotope ratios and their expanded uncertainties measured by the LAAT (blue circle), LANIE (red diamond) and LMV (green triangle) laboratories (figure a to e, respectively). The red line corresponds to the final average value with its expanded uncertainty (red dotted line)

References

1. Mialle S (2011) Développements analytiques en spectrométrie de masse à thermo-ionisation pour l'analyse isotopique de faibles quantités. Université Toulouse III - Paul Sabatier
2. Upadhyay D, Scherer EE, Mezger K (2008) Fractionation and mixing of Nd isotopes during thermal ionization mass spectrometry: implications for high precision $^{142}\text{Nd}/^{144}\text{Nd}$ analyses. *J Anal At Spectrom* 23:561. <https://doi.org/10.1039/b715585a>
3. Quemet A, Angenieux M, Ruas A (2021) Nd, Am and Cm isotopic measurements after simultaneous separation in transmutation irradiated samples. *J Anal At Spectrom* 36:1758–1767. <https://doi.org/10.1039/D1JA00165E>
4. Tanaka T, Togashi S, Kamioka H, Amakawa H, Kagami H, Hamamoto T, Yuhara M, Orihashi Y, Yoneda S, Shimizu H, Kunimaru T, Takahashi K, Yanagi T, Nakano T, Fujimaki H, Shinjo R, Asahara Y, Tanimizu M, Dragusanu C (2000) JNdi-1: A neodymium isotopic reference in consistency with LaJolla neodymium. *Chem Geol* 168:279–281. [https://doi.org/10.1016/S0009-2541\(00\)00198-4](https://doi.org/10.1016/S0009-2541(00)00198-4)
5. Garçon M, Boyet M, Carlson RW, Horan MF, Auclair D, Mock TD (2018) Factors influencing the precision and accuracy of Nd isotope measurements by thermal ionization mass spectrometry. *Chem Geol* 476:493–514. <https://doi.org/10.1016/j.chemgeo.2017.12.003>