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ORIGINAL PAPER

Data for the Reference Man: skeleton content of chemical elements

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Abstract This study was undertaken to provide reference values of chemical element mass fractions in intact bone of Reference (European Caucasian) Man/Woman. The rib bone samples investigated were obtained from autopsies of 84 apparently healthy 15–58-year-old citizens (38 females and 46 males) of a non-industrial region in the Central European part of Russia who had suffered sudden death. The mass fractions (mg/kg given on a wet mass basis) of 69 elements in these bone samples were measured by using neutron activation analysis with high-resolution spectrometry of short-lived and long-lived radionuclides, particle-induced gamma-ray emission, inductively coupled plasma atomic emission spectrometry, and inductively coupled plasma mass spectrometry including necessary quality control measures. Using published and measured data, mass fraction values of the 79 elements for the rib bone have been derived. Based on accepted rib to skeleton mass fractions and reference values of skeleton mass for Reference Man, the elemental burdens in the skeleton were estimated. These results may provide a representative bases for establishing related reference values for the Russian Reference Man/Woman and for revising and adding current reference values for the International Commission on Radiological Protection. The data presented will also be very valuable for many other applications in radiation protection, radiotherapy radiation dosimetry, and other scientific fields.

Keywords Chemical elements · Human rib bone · Age-gender-related differences · Reference Man/Woman · Nuclear analytical methods · ICP analytical methods

Introduction

The interaction of external ionizing radiation with tissues of the human body depends in part on the elemental composition of the tissues. The dose from internal sources of ionizing radiation depends on the radioactivity and metabolism of these sources in tissue, the characteristics of radiation emitted by them, and the elemental composition of the tissue. Bone is the most mineralized tissue in the human body. According to studies conducted with radionuclides (Moskalev 1985), many chemical elements including the alkaline earth elements [barium (Ba), beryllium (Be), calcium (Ca), magnesium (Mg), radium (Ra), strontium (Sr)], rare earth elements [scandium (Sc), yttrium (Y), lanthanum (La) and lanthanides, actinium (Ac) and actinides, including thorium (Th) and uranium (U)], lead (Pb), and some others are bone-seeking elements. Thus, data on the elemental composition of bone are very important for radioecology, radiation protection, medical radiology, radiobiology, and biophysics of the bone tissue.

In 1975, the International Commission on Radiological Protection (ICRP) published comprehensive elemental data in order to define ‘Reference Man’ as a typical individual in its Publication 23 (ICRP-23 1975). Current reference values of the ICRP for element mass fractions in bone and skeleton were issued in 2002 (ICRP-89 2002). Thus, the most comprehensive data are available from people living

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Dedicated to the blessed memory of my good friend Prof. Dr. Friedrich Grass (Atominstitut der Österreichischen Universitäten, Wien), who was the leading scientist in the field of modern nuclear analytical methods in the second part of the twentieth century.

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in both West Europe and North America, the latter regions being the main basis for the calculation of the ICRP reference values (ICRP-89 2002). For skeleton and bone (bone mineral), the report 89 of ICRP presents data on no more than 11 [hydrogen (H), carbon (C), nitrogen (N), oxygen (O), sodium (Na), Mg, phosphorus (P), sulfur (S), chlorine (Cl), Ca, and iron (Fe)] and nine (H, C, N, O, Na, Mg, P, S, and Ca) elemental mass fractions, respectively. As was pointed in (ICRP-89 2002), these values may not be representative for the populations of all countries.

State-of-the-art nuclear analytical methods and inductively coupled plasma (ICP) techniques allow detection of many trace and ultra-trace element mass fractions in bone. A number of investigations based on both nuclear and non-nuclear methods have contributed to the growth of data of elemental mass fractions in human bone as documented in numerous published papers. However, it is difficult to improve and to enrich current knowledge using accumulated data because, as follows from some reviews on the subject (Iyengar et al. 1978; Zwanziger 1989; Iyengar and Tandon 1999), there is a very widespread of results for almost all elements. Several sources of potential difficulties in dealing with bone samples were identified (Grynpas et al. 1987; Zaichick 1997, 2004a). The most important difficulties are associated with the issues of sample treatment. The majority of data published are based upon non-intact bones. In most cases, bone samples were ashed or were first treated with solvents in order to remove collagen, fat, and marrow before they were ashed. There is some evidence that certain elements of the bone mineral matrix are lost by this process and that the relationship between elemental mass fractions is also affected (Grynpas et al. 1987; Zaichick 1997, 2004a).

The first aim of the present study was to use the five most powerful instrumental analytical methods available [neutron activation analysis with high-resolution spectrometry of short-lived radionuclides (NAA-SLR), neutron activation analysis with high-resolution spectrometry of long-lived radionuclides (NAA-LLR), particle-induced gamma-ray emission (PIGE), inductively coupled plasma atomic emission spectrometry (ICP-AES), and inductively coupled plasma mass spectrometry (ICP-MS)] for a systematic investigation of elemental mass fractions in intact rib bone samples of residents from a non-industrial region in the Central European part of Russia. The second aim was to investigate any potential age and gender variations in elemental mass fractions. The third aim was to estimate the median of means and the uncertainty of results published on elemental mass fractions in human bones and rib bone separately, while the fourth aim was to calculate elemental mass fractions in the skeleton of the Russian Reference Man (males/females) and to compare the results with data for the ICRP-89 Reference Man and the Chinese Reference Man (Zhu et al. 2010).

Materials and methods

Samples of the human rib bone were obtained at postmortems from intact cadavers (38 females and 46 males, 15–55 years old, Caucasian, European living habits) within 24 h after death. Each death had resulted from automobile accidents, falls, shootings, stabbing, hanging, acute alcohol poisoning, or hypothermia. All the deceased were citizens of Obninsk, a small city in a non-industrial region 105 km southwest of Moscow. None of those who died a sudden death had suffered previously from any systematic or chronic disorders. All studies were approved by the Forensic Medicine Department of Obninsk City Hospital and the Ethical Committee of the Medical Radiological Research Center.

Sample preparation

The majority of samples were taken from the 3rd, 4th, 5th, or 6th rib on the right side. A tool made of titanium and plastic was used to clean soft tissues and blood from the samples. Then, the samples were weighed and the weights were recorded and used in the data analysis. Then, the samples were freeze-dried until a constant mass was obtained. A titanium scalpel was used to cut thin cross-sections of the rib (subsamples), each weighing about 50–100 mg. Four subsamples were made from each rib bone sample: one subsample for NAA-SLR, the second for NAA-LLR, the third for PIGE, and the last one for ICP-AES and ICP-MS. The subsamples for NAA-SLR were sealed separately in thin polyethylene films prewashed with acetone and rectified alcohol. The sealed samples were placed in labeled polyethylene ampoules. The subsamples for NAA-LLR were wrapped separately in a high-purity aluminum foil washed with rectified alcohol beforehand and placed in a nitric acid-washed quartz ampoule. The subsamples for PIGE were dried for an additional 92 h in an oven at 110 °C. The subsamples for ISP-AES and ISP-MS were decomposed in autoclaves. 1.5 mL of concentrated HNO_3 (*Nitric acid 65 %, max. 0.0000005 % Hg, GR, ISO, Merck*) and 0.3 mL of H_2O_2 (*pure for analysis*) were added to rib bone samples, placed in one-chamber autoclaves (Ancon-AT2, Ltd., Russia), and then heated for 3 h at 160–200 °C. After autoclaving, they were cooled to room temperature and solutions from the decomposed samples were diluted with deionized water (up to 20 mL) and transferred to plastic measuring bottles. Simultaneously, the same procedure was performed in autoclaves without bone samples (only $HNO_3 + H_2O_2$ + deionized water), and the resultant solutions were used as control samples.

Table 1 Arithmetic means ± standard deviations or possible upper limits of the means of elemental mass fractions (mg/kg on a wet mass basis) in intact human rib bone obtained by means of five analytical methods

Element	NAA-SLR	NAA-LLR	PIGE	ICP-AES	ICP-MS	Derived value
Ag	–	<0.01 (DL)	–	–	≤0.0065	≤0.0065
Al	–	–	–	≤4.7	≤4.7	≤4.7
As	–	<0.05 (DL)	–	–	<0.006 (DL)	<0.006 (DL)
Au	–	<0.005 (DL)	–	–	<0.0004 (DL)	<0.0004 (DL)
B	–	–	–	≤0.42	≤0.42	≤0.42
Ba	–	<50 (DL)	–	1.61 ± 0.98	1.61 ± 0.98	1.62 ± 0.98
Be	–	–	–	–	≤0.0020	≤0.0020
Bi	–	–	–	–	0.0087 ± 0.0070	0.0087 ± 0.0070
Br	–	<5 (DL)	–	–	≤2.5	≤2.5
Ca (g/kg)	123 ± 36	–	–	110 ± 31	–	116 ± 30
Cd	–	<1 (DL)	–	–	0.028 ± 0.025	0.028 ± 0.025
Ce	–	≤0.016	–	–	0.018 ± 0.013	0.018 ± 0.013
Cl	621 ± 274	–	–	–	–	621 ± 274
Co	–	0.0016 ± 0.0014	–	–	<0.2 (DL)	0.0016 ± 0.0014
Cr	–	<0.5 (DL)	–	–	≤0.14	≤0.14
Cs	–	≤0.0063	–	–	≤0.0048	≤0.0049
Cu	–	–	–	0.62 ± 0.23	0.62 ± 0.23	0.62 ± 0.23
Dy	–	–	–	–	0.0013 ± 0.0009	0.0013 ± 0.0009
Er	–	–	–	–	0.00068 ± 0.00062	0.00068 ± 0.00062
Eu	–	≤0.00060	–	–	<0.0006 (DL)	≤0.00060
F	–	–	978 ± 441	–	–	978 ± 441
Fe	–	82 ± 58	–	87 ± 54	–	82.9 ± 49.9
Ga	–	–	–	–	<0.1 (DL)	<0.1 (DL)
Gd	–	<0.15 (DL)	–	–	0.0010 ± 0.0007	0.0097 ± 0.00071
Hf	–	<0.01 (DL)	–	–	–	<0.01 (DL)
Hg	–	≤0.005	–	–	≤0.011	≤0.0096
Ho	–	–	–	–	≤0.00033	≤0.00033
Ir	–	–	–	–	<0.00002 (DL)	<0.00002 (DL)
K	544 ± 271	–	–	1,171 ± 361	–	863 ± 244
La	–	<0.03 (DL)	–	–	0.0123 ± 0.0102	0.0123 ± 0.0102
Li	–	–	–	0.02 ± 0.01	0.024 ± 0.009	0.0236 ± 0.0095
Lu	–	<0.002 (DL)	–	–	≤0.00015	≤0.00015
Mg	1,331 ± 485	–	–	1,367 ± 309	–	1,345 ± 333
Mn	0.19 ± 0.12	–	–	≤0.21	≤0.21	0.165 ± 0.081
Mo	–	–	–	–	0.033 ± 0.026	0.033 ± 0.026
Na	3,025 ± 927	–	–	3,433 ± 746	–	3,222 ± 738
Nb	–	–	–	–	<0.006 (DL)	<0.006 (DL)
Nd	–	<0.06 (DL)	–	–	0.0067 ± 0.0054	0.0067 ± 0.0054
Ni	–	–	–	–	≤0.64	≤0.64
P (g/kg)	53.1 ± 15.5	–	–	48.2 ± 12.9	–	50.7 ± 12.6
Pa	–	<0.04 (DL)	–	–	–	<0.04 (DL)
Pb	–	–	–	–	1.42 ± 0.80	1.42 ± 0.80
Pd	–	–	–	–	<0.006 (DL)	<0.006 (DL)
Pr	–	–	–	–	0.0021 ± 0.0021	0.0021 ± 0.0021
Pt	–	–	–	–	<0.001 (DL)	<0.001 (DL)
Rb	–	≤1.0	–	–	0.96 ± 0.35	0.94 ± 0.38
Re	–	–	–	–	<0.0003 (DL)	<0.0003 (DL)
Rh	–	–	–	–	<0.006 (DL)	<0.006 (DL)

Table 1 continued

Element	NAA-SLR	NAA-LLR	PIGE	ICP-AES	ICP-MS	Derived value
S	–	–	–	1,169 ± 237	–	1,169 ± 237
Sb	–	<0.006 (DL)	–	–	≤0.0056	≤0.0056
Sc	–	<0.0006 (DL)	–	–	<0.06 (DL)	<0.0006 (DL)
Se	–	≤0.18	–	–	≤0.1	≤0.11
Sm	–	<0.006 (DL)	–	–	0.00089 ± 0.00077	0.00089 ± 0.00077
Sn	–	–	–	–	<0.1 (DL)	<0.1 (DL)
Sr	162 ± 102	174 ± 125	–	186 ± 118	186 ± 118	177 ± 101
Ta	–	–	–	–	<0.003 (DL)	<0.003 (DL)
Tb	–	<0.02 (DL)	–	–	0.00024 ± 0.00015	0.00024 ± 0.00015
Te	–	–	–	–	≤0.0029	≤0.0029
Th	–	<0.03 (DL)	–	–	≤0.0019	≤0.0019
Ti[a]	–	–	–	–	≤1.7	≤1.7
Tl	–	–	–	–	0.00031 ± 0.00015	0.00031 ± 0.00015
Tm	–	–	–	–	≤0.000040	≤0.000040
U	–	<0.04 (DL)		–	0.00082 ± 0.00059	0.00082 ± 0.00059
V	–	–	–	<0.02 (DL)	<0.02 (DL)	<0.02 (DL)
W	–	–	–	–	<0.06 (DL)	<0.06 (DL)
Y	–	–	–	–	≤0.0029	≤0.0029
Yb	–	<0.02 (DL)	–	–	0.00046 ± 0.00029	0.00046 ± 0.00029
Zn	–	46.3 ± 12.4	–	59.2 ± 12.9	59.2 ± 12.9	56.9 ± 13.1
Zr	–	<0.6 (DL)	–	–	<0.02 (DL)	<0.02 (DL)

[a] Titanium tools were used for sampling and sample preparation

DL detection limit, *NAA-SLR* neutron activation analysis with high-resolution spectrometry of short-lived radionuclides, *NAA-LLR* neutron activation analysis with high-resolution spectrometry of long-lived radionuclides, *PIGE* particle-induced gamma-ray emission, *ICP-AES* inductively coupled plasma atomic emission spectrometry, *ICP-MS* inductively coupled plasma mass spectrometry, Derived value—for elements investigated by two or more methods, the mean of all results was used

Analytical procedures

The bone content of 69 elements was measured with the methods mentioned above (Table 1).

Details of the analytical methods and procedures used here such as nuclear reactions, radionuclides, gamma-energies, wavelength, isotopes, spectrometers, spectrometer parameters, and operating conditions were presented in earlier publications (Zaichick et al. 2000, 2009, 2011a; Sastri et al. 2001; Zaichick and Zaichick 2009, 2010).

Standard and certified reference material analysis

For quality control, ten subsamples of the certified reference material CRM IAEA H-5 Animal Bone from the International Atomic Energy Agency (IAEA), nine subsamples of the standard reference material SRM NIST 1486 Bone Meal from the National Institute Standards and Technologies (NIST, USA), and three subsamples of certified reference materials CRM INCT-TL-1 Tea Leaves and CRM INCT-MPH-2 Mixed Polish Herbs from the Institute of Nuclear Chemistry and Technology (INCT, Warsaw, Poland) were analyzed simultaneously with the investigated rib samples. To check the accuracy of the analytical results for fluorine (F), four subsamples of the reference materials SRM NIST 1486 Bone Meal, SRM NIST-1632a Coa, and USGS ScO-l Cody Shale 1 from NIST were also analyzed. All samples of standard and certified reference materials were treated in the same way as the rib samples. Detailed results of this quality assurance program were presented in earlier publications (Zaichick et al. 2000, 2009, 2011a; Sastri et al. 2001; Zaichick and Zaichick 2009, 2010).

Computer programs and statistics

Using Microsoft Office Excel, a summary of statistical features (arithmetic mean, standard deviation, standard error of mean, minimum and maximum values, median, percentiles with 0.025 and 0.975 levels) or upper limits of the means and detection limits for elemental mass fraction of 69 elements were determined for intact human rib bone. For elements investigated by two or more methods, the mean of all results was used. Using individual values of

Table 2 Some statistical parameters of elemental mass fractions (mg/kg on a wet mass basis) in rib bone of both healthy females and males aged 15–55 years, taken together ($n = 84$)

Element	M	SD	SEM	Min	Max	Med	P0.025	P0.975
Ba	1.62	0.98	0.11	0.392	5.40	1.39	0.554	5.14
Bi	0.0087	0.0070	0.0008	0.00017	0.0446	0.0095	0.00034	0.0192
Ca (g/kg)	116	30.2	3.3	64.6	222	116	66.2	175
Cd	0.028	0.025	0.003	0.0036	0.158	0.021	0.0042	0.0898
Ce	0.018	0.013	0.001	0.0021	0.0713	0.014	0.0040	0.0466
Cl	621	274	30	87.6	1815	628	104	1098
Co	0.0016	0.0014	0.0003	0.00005	0.00408	0.0012	0.00007	0.00402
Cu	0.62	0.23	0.03	0.195	1.23	0.60	0.25	1.11
Dy	0.0013	0.0010	0.0002	0.00011	0.00404	0.0010	0.00014	0.0033
Er	0.00068	0.00062	0.00013	0.00006	0.00224	0.00044	0.00006	0.00208
F	978	441	101	179	1,776	959	231	1,736
Fe	82.9	49.9	5.5	18.6	233	72.6	21.4	209
Gd	0.00097	0.00071	0.00008	0.00006	0.00407	0.00070	0.00008	0.00303
K	863	244	27	420	1,605	847	490	1,312
La	0.0123	0.0102	0.0011	0.0010	0.0541	0.0087	0.0027	0.0392
Li	0.0236	0.0095	0.0011	0.0081	0.0460	0.0222	0.0102	0.0436
Mg	1,345	333	37	670	2,053	1,326	695	1,992
Mn	0.165	0.081	0.009	0.0373	0.389	0.154	0.0472	0.360
Mo	0.033	0.026	0.003	0.009	0.148	0.027	0.010	0.104
Na	3,222	738	81	1,947	5,504	3,210	2,047	5,004
Nd	0.0067	0.0054	0.0006	0.00047	0.0297	0.0048	0.0014	0.0234
P (g/kg)	50.7	12.6	1.3	28.1	90.1	50.2	30.6	71.9
Pb	1.42	0.80	0.09	0.401	4.22	1.22	0.499	3.61
Pr	0.0021	0.0021	0.0002	0.00025	0.0124	0.0014	0.00044	0.00913
Rb	0.94	0.38	0.04	0.067	2.39	0.88	0.42	1.69
S	1,169	237	27	618	1,950	1,136	723	1,619
Sm	0.00089	0.00077	0.00009	0.00011	0.00460	0.00058	0.00012	0.00306
Sr	177	101	11	18.5	633	159	40.9	424
Tb	0.00024	0.00015	0.00002	0.00003	0.00071	0.00024	0.00004	0.00058
Tl	0.00031	0.00015	0.00002	0.00005	0.00113	0.00029	0.00016	0.00060
U	0.00082	0.00059	0.00007	0.00026	0.00329	0.00061	0.00029	0.00247
Yb	0.00046	0.00029	0.00004	0.00008	0.00124	0.00042	0.00011	0.00123
Zn	56.9	13.1	1.4	32.4	96.5	56.4	34.9	82.3

M arithmetic mean, *SD* standard deviation, *SEM* standard error of the mean, *Min* minimum value, *Max* maximum value, *Med* median value, *P0.025* percentile at 0.025 level, *P0.975* percentile at 0.975 level

water content in the investigated bone samples, all elemental mass fractions were calculated on a wet mass basis. The significance of any apparent difference in the results obtained for the female and male subcohorts as well as that in the results obtained for two age groups was evaluated by Student's *t* test.

Results

Table 1 depicts the results obtained for 69 elemental mass fractions (arithmetic mean ± standard deviation, upper limit of the mean, detection limit) in intact human rib bone (both females and males taken together) measured by means of the five analytical methods described above.

Table 2 represents certain statistical features (arithmetic mean, standard deviation, standard error of mean, minimum and maximum values, median, percentiles at 0.025 and 0.975 levels) of 33 elements in rib bone of both males and females, taken together.

To estimate the effect of age on the results obtained, two age groups were examined separately: one comprised a younger group with ages ranging from 15 to 35 years while the other comprised older people with ages ranging from

Table 3 Effect of age on mean values (M ± SEM) of elemental mass fractions in intact human rib bone (mg/kg on a wet mass basis)

Element	Males			Females			Females and males		
	15–35 years *n* = 22	36–55 years *n* = 24	*p* *t* test	15–35 years *n* = 18	36–55 years *n* = 20	*p* *t* test	15–35 years *n* = 40	36–55 years *n* = 44	*p* *t* test
Ba	1.26 ± 0.11	1.44 ± 0.13	N.S.	1.83 ± 0.25	1.94 ± 0.31	N.S.	1.56 ± 0.15	1.67 ± 0.16	N.S.
Bi	0.0094 ± 0.0013	0.0062 ± 0.0011	N.S.	0.0096 ± 0.0015	0.0102 ± 0.0022	N.S.	0.0095 ± 0.0010	0.0080 ± 0.0012	N.S.
Ca (g/kg)	114 ± 5	103 ± 5	N.S.	134 ± 7	119 ± 7	N.S.	123 ± 5	110 ± 4	≤0.05
Cd	0.026 ± 0.007	0.027 ± 0.004	N.S.	0.021 ± 0.004	0.035 ± 0.007	N.S.	0.023 ± 0.004	0.031 ± 0.004	N.S.
Ce	0.0138 ± 0.0024	0.0245 ± 0.0025	≤0.01	0.0112 ± 0.0022	0.0202 ± 0.0033	≤0.05	0.0126 ± 0.0017	0.0226 ± 0.0020	≤0.001
Cl	641 ± 35	709 ± 39	N.S.	659 ± 91	455 ± 65	N.S.	650 ± 45	594 ± 41	N.S.
Co	0.0013 ± 0.0004	0.0019 ± 0.0004	N.S.	–	–		0.0013 ± 0.0004	0.0019 ± 0.0004	N.S.
Cu	0.66 ± 0.08	0.64 ± 0.04	N.S.	0.67 ± 0.06	0.53 ± 0.03	N.S.	0.66 ± 0.05	0.59 ± 0.03	N.S.
Dy	0.00059 ± 0.00006	0.00169 ± 0.00026	≤0.001	≤0.00063	≤0.00087	–	0.00061 ± 0.00019	0.00159 ± 0.00024	≤0.01
Er	0.00022 ± 0.00003	0.00092 ± 0.00018	≤0.001	≤0.00046	≤0.00031	–	0.00032 ± 0.00015	0.00084 ± 0.00016	≤0.05
F	971 ± 226	1250 ± 251	N.S.	830 ± 161	929 ± 37	N.S.	900 ± 134	1112 ± 150	N.S.
Fe	79.5 ± 8.9	119 ± 12	≤0.05	59.6 ± 8.5	64.3 ± 6.8	N.S.	70.3 ± 6.3	94.0 ± 8.4	≤0.05
Gd	0.00060 ± 0.00003	0.00137 ± 0.00017	≤0.001	0.00057 ± 0.00009	0.00115 ± 0.00015	≤0.001	0.00059 ± 0.00005	0.00127 ± 0.00012	≤0.001
K	827 ± 50	950 ± 45	N.S.	795 ± 67	860 ± 52	N.S.	812 ± 40	909 ± 34	N.S.
La	0.0085 ± 0.0012	0.0176 ± 0.0024	≤0.01	0.0077 ± 0.0015	0.0132 ± 0.0024	N.S.	0.0081 ± 0.0010	0.0156 ± 0.0018	≤0.001
Li	0.022 ± 0.002	0.025 ± 0.002	N.S.	0.023 ± 0.003	0.025 ± 0.002	N.S.	0.022 ± 0.002	0.025 ± 0.002	N.S.
Mg	1,405 ± 69	1,236 ± 56	N.S.	1,498 ± 78	1,273 ± 83	≤0.05	1,448 ± 51	1,253 ± 48	≤0.01
Mn	0.133 ± 0.011	0.184 ± 0.017	≤0.05	0.175 ± 0.018	0.169 ± 0.025	N.S.	0.152 ± 0.010	0.177 ± 0.014	N.S.
Mo	0.028 ± 0.005	0.024 ± 0.002	N.S.	0.048 ± 0.010	0.035 ± 0.005	N.S.	0.039 ± 0.006	0.029 ± 0.003	N.S.
Na	3,160 ± 143	2986 ± 123	N.S.	3,514 ± 197	3,310 ± 181	N.S.	3,320 ± 120	3,133 ± 108	N.S.
Nd	0.0043 ± 0.0007	0.0093 ± 0.0012	≤0.001	0.0040 ± 0.0007	0.0080 ± 0.0014	≤0.05	0.0042 ± 0.0005	0.0087 ± 0.0009	≤0.001
P (g/kg)	49.8 ± 2.4	45.1 ± 2.0	N.S.	57.4 ± 2.9	52.6 ± 3.2	N.S.	53.2 ± 1.9	48.5 ± 1.9	N.S.
Pb	1.27 ± 0.21	1.57 ± 0.17	N.S.	1.41 ± 0.23	1.34 ± 0.13	N.S.	1.35 ± 0.16	1.47 ± 0.11	N.S.
Pr	0.00112 ± 0.00020	0.00310 ± 0.00053	≤0.01	0.00159 ± 0.00052	0.00224 ± 0.00042	N.S.	0.00136 ± 0.00028	0.00271 ± 0.00035	≤0.01
Rb	0.96 ± 0.10	0.98 ± 0.07	N.S.	0.92 ± 0.09	0.90 ± 0.07	N.S.	0.94 ± 0.07	0.94 ± 0.05	N.S.
S	1179 ± 56	1175 ± 49	N.S.	1,178 ± 70	1,142 ± 43	N.S.	1,178 ± 44	1,161 ± 33	N.S.
Sm	0.00052 ± 0.00005	0.00132 ± 0.00016	≤0.001	0.00041 ± 0.00003	0.00111 ± 0.00023	≤0.01	0.00047 ± 0.00003	0.00123 ± 0.00013	≤0.001
Sr	153 ± 17	153 ± 11	N.S.	238 ± 37	176 ± 18	N.S.	191 ± 20	163 ± 10	N.S.
Tb	0.000178 ± 0.000027	0.000307 ± 0.000035	≤0.01	≤0.00019	≤0.00023	–	0.000182 ± 0.000021	0.000288 ± 0.000030	≤0.01
Tl	0.00027 ± 0.00003	0.00033 ± 0.00005	N.S.	0.00032 ± 0.00002	0.00032 ± 0.00003	N.S.	0.00030 ± 0.00002	0.00032 ± 0.00003	N.S.
U	0.00075 ± 0.00011	0.00099 ± 0.00015	N.S.	0.00071 ± 0.00016	0.00075 ± 0.00009	N.S.	0.00073 ± 0.00010	0.00088 ± 0.00009	N.S.
Yb	0.00032 ± 0.00007	0.00061 ± 0.00010	≤0.05	≤0.00039	≤0.00040	–	0.00037 ± 0.00004	0.00051 ± 0.00005	≤0.05
Zn	53.1 ± 2.2	53.6 ± 2.8	N.S.	59.6 ± 2.6	62.5 ± 3.3	N.S.	56.1 ± 1.8	57.7 ± 2.2	N.S.

M arithmetic mean, *SEM* standard error of the mean, *N.S.* not significant, *n* number of individuals

Table 4 Effect of gender on mean values (M ± SEM) of elemental mass fractions in intact human rib bone (mg/kg on a wet mass basis)

Element	15–35 years			36–55 years			15–55 years		
	Females $n = 18$	Males $n = 22$	p *t* test	Females $n = 20$	Males $n = 24$	p *t* test	Females $n = 38$	Males $n = 46$	p *t* test
Ba	1.83 ± 0.25	1.26 ± 0.11	N.S.	1.94 ± 0.31	1.44 ± 0.13	N.S.	1.89 ± 0.20	1.37 ± 0.09	≤0.05
Bi	0.0096 ± 0.0015	0.0094 ± 0.0013	N.S.	0.0102 ± 0.0022	0.0062 ± 0.0011	N.S.	0.0099 ± 0.0014	0.0075 ± 0.0009	N.S.
Ca (g/kg)	134 ± 7	114 ± 5	≤0.05	119 ± 7	103 ± 5	N.S.	126 ± 5	108 ± 4	≤0.01
Cd	0.021 ± 0.004	0.026 ± 0.007	N.S.	0.035 ± 0.007	0.027 ± 0.004	N.S.	0.028 ± 0.005	0.027 ± 0.004	N.S.
Ce	0.0112 ± 0.0022	0.0138 ± 0.0024	N.S.	0.0202 ± 0.0033	0.0245 ± 0.0025	N.S.	0.016 ± 0.002	0.020 ± 0.002	N.S.
Cl	659 ± 91	641 ± 35	N.S.	455 ± 65	709 ± 39	≤0.01	554 ± 57	677 ± 27	N.S.
Co	–	0.0013 ± 0.0004	–	–	0.0019 ± 0.0004	–	<0.06	0.0016 ± 0.0003	–
Cu	0.67 ± 0.06	0.66 ± 0.08	N.S.	0.53 ± 0.03	0.64 ± 0.04	≤0.05	0.59 ± 0.04	0.65 ± 0.04	N.S.
Dy	≤0.00063	0.00059 ± 0.00006	–	≤0.00087	0.00169 ± 0.00026	–	≤0.00073	0.0015 ± 0.0002	–
Er	≤0.00046	0.00022 ± 0.00003	–	≤0.00031	0.00092 ± 0.00018	–	≤0.00040	0.00076 ± 0.00015	–
F	830 ± 161	971 ± 226	N.S.	929 ± 37	1250 ± 251	N.S.	863 ± 106	1082 ± 166	N.S.
Fe	59.6 ± 8.5	79.5 ± 8.9	N.S.	64.3 ± 6.8	119 ± 12	≤0.001	62.1 ± 5.3	100 ± 8	≤0.001
Gd	0.00057 ± 0.00009	0.00060 ± 0.00003	N.S.	0.00115 ± 0.00015	0.00137 ± 0.00017	N.S.	0.00089 ± 0.00011	0.00105 ± 0.00012	N.S.
K	795 ± 67	827 ± 50	N.S.	860 ± 52	950 ± 45	N.S.	829 ± 41	891 ± 34	N.S.
La	0.0077 ± 0.0015	0.0085 ± 0.0012	N.S.	0.0132 ± 0.0024	0.0176 ± 0.0024	N.S.	0.0106 ± 0.0015	0.0138 ± 0.0017	N.S.
Li	0.023 ± 0.003	0.022 ± 0.002	N.S.	0.025 ± 0.002	0.025 ± 0.002	N.S.	0.024 ± 0.002	0.023 ± 0.001	N.S.
Mg	1,498 ± 78	1,405 ± 69	N.S.	1,273 ± 83	1,236 ± 56	N.S.	1,379 ± 59	1,315 ± 45	N.S.
Mn	0.175 ± 0.018	0.133 ± 0.011	N.S.	0.169 ± 0.025	0.184 ± 0.017	N.S.	0.17 ± 0.02	0.16 ± 0.01	N.S.
Mo	0.048 ± 0.010	0.028 ± 0.005	N.S.	0.035 ± 0.005	0.024 ± 0.002	≤0.05	0.041 ± 0.006	0.025 ± 0.002	≤0.05
Na	3,514 ± 197	3,160 ± 143	N.S.	3,310 ± 181	2,986 ± 123	N.S.	3,407 ± 132	3,069 ± 94	≤0.05
Nd	0.0040 ± 0.0007	0.0043 ± 0.0007	N.S.	0.0080 ± 0.0014	0.0093 ± 0.0012	N.S.	0.0061 ± 0.0009	0.0072 ± 0.0009	N.S.
P (g/kg)	57.4 ± 2.9	49.8 ± 2.4	≤0.05	52.6 ± 3.2	45.1 ± 2.0	≤0.05	54.8 ± 2.2	47.3 ± 1.6	≤0.01
Pb	1.41 ± 0.23	1.27 ± 0.21	N.S.	1.34 ± 0.13	1.57 ± 0.17	N.S.	1.37 ± 0.13	1.46 ± 0.14	N.S.
Pr	0.00159 ± 0.00052	0.00112 ± 0.00020	N.S.	0.00224 ± 0.00042	0.00310 ± 0.00053	N.S.	0.0019 ± 0.0003	0.0023 ± 0.0004	N.S.
Rb	0.92 ± 0.09	0.96 ± 0.10	N.S.	0.90 ± 0.07	0.98 ± 0.07	N.S.	0.91 ± 0.06	0.97 ± 0.06	N.S.
S	1178 ± 70	1179 ± 56	N.S.	1142 ± 43	1,175 ± 49	N.S.	1,159 ± 40	1,177 ± 36	N.S.
Sm	0.00041 ± 0.00003	0.00052 ± 0.00005	N.S.	0.00111 ± 0.00023	0.00132 ± 0.00016	N.S.	0.00078 ± 0.00013	0.00099 ± 0.00011	N.S.
Sr	238 ± 37	153 ± 17	≤0.05	176 ± 18	153 ± 11	N.S.	205 ± 20	153 ± 10	≤0.05
Tb	≤0.00019	0.000178 ± 0.000027	–	≤0.00023	0.000307 ± 0.000035	–	≤0.00021	0.00025 ± 0.00002	–
Tl	0.00032 ± 0.00002	0.00027 ± 0.00003	N.S.	0.00032 ± 0.00003	0.00033 ± 0.00005	N.S.	0.00032 ± 0.00002	0.00031 ± 0.00003	N.S.
U	0.00071 ± 0.00016	0.00075 ± 0.00011	N.S.	0.00075 ± 0.00009	0.00099 ± 0.00015	N.S.	0.00073 ± 0.00009	0.00089 ± 0.00010	N.S.
Yb	≤0.00039	0.00032 ± 0.00007	–	≤0.00040	0.00061 ± 0.00010	–	≤0.00040	0.00053 ± 0.00008	–
Zn	59.6 ± 2.6	53.1 ± 2.2	N.S.	62.5 ± 3.3	53.6 ± 2.8	≤0.05	61.1 ± 2.1	53.4 ± 1.8	≤0.01

M arithmetical mean, *SEM* standard error of the mean, ≤ *M* possible upper limit of the mean value, *N.S.* not significant, *n* number of individuals

36 to 55 years. The results for females and males, taken separately and together, are shown in Table 3.

The entire dataset for both females and males taken separately was also used to quantify any gender-related differences (see Table 4). However, significant differences were found to accrue from differences in age only, as judged by *p* values (see Table 3). This led us to provide the additional information in Table 4.

Range of means (medians) and max/min ratio of elemental mass fractions in human bone (various bones) and human rib bone (given separately) according to data from the literature are shown in Tables 5, 6, respectively. In the literature, sometimes, elemental mass fractions in human rib bone were not expressed on a wet mass basis. In this case, for Table 6, we calculated these values using published data for water and ash contents in the rib bone (Zaichick et al. 2000; Tzaphlidou and Zaichick 2003).

Variations due to analytical uncertainty, analytical uncertainty and individual element variability combined, and due to differences between the results obtained in the present work and those of a previous publication with good quality control (Zhu et al. 2010) are given in Table 7, while in Table 8, median values of means (medians) of 79 elemental mass fractions calculated using published data are compared with results obtained here for the human rib bone.

Skeleton burdens of 79 elements were finally estimated for the Russian Reference Man/Woman using the results obtained in the present work and the accepted values of mass fractions for some bulk (C, H, N, O), trace (Ge, I, Si), and ultra-trace (In, Po, Ra) elements (Table 9). The results are compared with the published data (also shown in Table 9).

Discussion

The use of five analytical methods allowed to estimate the mass fractions of 69 elements in human rib bones. Good agreement was found between the results obtained with NAA and ICP for major (Ca, P), minor (K, Mg, Na), and trace elements (Fe, Mn, Sr, Zn) indicating complete digestion of the rib bone samples (for ICP techniques) and correctness of all results obtained by the various methods (Table 1). The fact that the elemental mass fractions (mean ± SD) of the standard and certified reference materials obtained in the present work were in good agreement with the certified values and within the corresponding 95 % confidence intervals (Zaichick 1995; Zaichick et al. 2000, 2009; Sastri et al. 2001; Zaichick and Zaichick 2009, 2010; Zaichick et al. 2011a, b) suggests an acceptable accuracy of the measurements performed on intact rib bone samples.

Elemental mass fractions in rib bone of Russian adult men/women

The mass fractions for 33 chemical elements listed in Table 1 (Ba, Bi, Ca, Cd, Ce, Cl, Co, Cu, Dy, Er, F, Fe, Gd, K, La, Li, Mg, Mn, Mo, Na, Nd, P, Pb, Pr, Rb, S, Sm, Sr, Tb, Tl, U, Yb, and Zn) were measured in the total or in a major fraction of the investigated rib bone samples. This allowed calculation of the mean values and selected statistical features for these elements for both males and females, taken separately and together (Table 2).

The mass fractions of Ag, Al, B, Be, Br, Cr, Cs, Eu, Hg, Ho, Lu, Ni, Sb, Se, Te, Th, Ti, Tm, and Y (19 elements) were determined only in a few samples collected. The possible upper limit of the mean (≤M) for these elements (Table 1) was calculated as the average mass fraction for each element, using the value of the detection limit instead of the individual value when these latter were found below the detection limit:

$$\leq M = \left(\sum_{i}^{n_i} C_i + DL \cdot n_j \right) \Big/ n$$

where C_i is the individual value of chemical element mass fraction in the *ith* sample, n_i is the number of samples with a measured mass fraction above the detection limit (DL), n_j is the number of samples with a measured mass fraction below the DL, and $n = n_i + n_j$ is the number of investigated samples.

Generally, the mass fractions of 17 elements were lower than the corresponding detection limits (DL, mg/kg on wet mass basis): As(<0.006), Au(<0.0004), Ga(<0.1), Hf(<0.01), Ir(<0.00002), Nb(<0.006), Pa(<0.04), Pd(<0.006), Pt(<0.001), Re(<0.0003), Rh(<0.006), Sc(<0.0006), Sn(<0.1), Ta(<0.003), V(<0.02), W(<0.06), and Zr(<0.02) (Table 1).

Age-related changes in elemental mass fractions of rib bone

An age-related decrease in Ca, Mg, and P mass fractions was observed in separate cohorts of females and males (see Table 3). Note that the most pronounced and statistically significant age-related differences in Ca and Mg mass fractions were detected when the age groups of females and males were analyzed together. Information from other sources on the impact of age on Ca, Mg, and P mass fractions is contradictory. Tipton et al. (1968) reported that Americans of Caucasian origin lose some 20 % of Ca by the age of 55 years. The rib content of Mg also drops with age in American adults. For Germans, the Ca loss was reported to be much lower in comparison with Americans (Schneider and Anke 1971; Anke et al. 1978). However, for Japanese (Yoshinaga et al. 1989; Yoshinaga et al.

Table 5 Range (minimum–maximum value) of means/medians of elemental mass fractions (mg/kg on a dry mass basis) and max/min ratio of the elemental mass fractions in various human bones according to the data from the literature

Element	Published data					Ratio max/min combined data
	(Bowen 1966; 1979)	(Zwanziger 1989)	(Iyengar et al. 1978; Iyengar and Tandon 1999)	Data published in Russia (own data)	Combined data	
Ag	0.01–0.44	–	0.0028–0.72	<0.1–130	0.0028–130	4.6×10^4
Al	4–27	19.5–54	0.24–5,430	0.1–380	0.1–5,430	5.4×10^4
As	0.08–1.6	0.01	<0.002–2.7	0.01–0.09	<0.002–2.7	$>1.4 \times 10^3$
Au	1.6×10^{-5}	$(<0.01–4) \times 10^3$	3×10^{-6}–0.6	<0.003	3×10^{-6}–0.6	2.0×10^5
B	1.1–3.3	<2–14	0.89–30	<3.0–166	0.89–166	1.9×10^2
Ba	3–70	10.2–314	0.41–314	1.96	0.41–314	7.7×10^2
Be	0.003	–	<0.0002–13.0	–	<0.0002–13.0	$>6.5 \times 10^1$
Bi	<0.2	–	<0.13–<0.2	–	<0.13–<0.2	–
Br	6.7–7	0.02–23.4	0.6–410	0.36	0.02–410	2.1×10^4
C (g/kg)	8–360	–	–	93–278	8–360	4.5×10^1
Ca (g/kg)	170–260	0.24–405	60–730	2.0–285	0.24–730	3.0×10^3
Cd	1.8	0.28–24.7	<0.001–8.0	–	<0.001–24.7	2.5×10^4
Ce	2.7	–	–	–	2.7	–
Cl	900–2,700	170–1,200	228–4,300	920–2,000	170–4,300	2.5×10^1
Co	0.01–5	<0.01–3.8	0.01–43.5	0.12–0.7	<0.01–43.5	$>4.4 \times 10^3$
Cr	0.1–33	<2	0.1–33	<0.5–6.0	<0.1–33	$>3.3 \times 10^2$
Cs	0.013–0.14	–	0.01–0.17	7.2	0.01–7.2	7.2×10^2
Cu	1–26	0.09–7.9	<0.1–39	0.007–117	0.007–117	1.7×10^4
Dy	<0.36	–	<0.01–0.06	–	<0.01–0.06	>6
Er	<0.28	–	<0.01–0.04	–	<0.01–0.28	2.8×10^1
Eu	0.032	0.004–0.069	<0.001–0.073	–	<0.001–0.073	$>7.3 \times 10^1$
F	1,500–12,000	120–4,320	214–18,600	120–1,060	120–18,600	1.55×10^2
Fe	3–380	4.5–6,300	2.3–2,458	2.3–6,242	2.3–6,300	2.7×10^3
Ga	–	–	<0.03	1.7	<0.03–1.7	$>5.7 \times 10^1$
Gd	<0.4	–	<0.008–0.2	–	<0.008–<0.4	–
Ge	–	–	<0.07	–	<0.07	–
H (g/kg)	52	–	–	34–66	34–66	1.94
Hf	0.12	–	<0.2–0.6	–	0.12–0.6	5.0
Hg	0.1–0.45	0.011–4.7	<0.01–1.2	–	<0.01–4.7	$>4.7 \times 10^2$
Ho	<0.08	–	<0.004	–	<0.004–0.08	$>2.0 \times 10^1$
I	21	–	18	0.36	0.36–21	5.8×10^1
In	–	–	<0.01–0.01	–	<0.01–0.01	–
Ir	–	–	–	1.2	1.2	–
K	900–2,100	4,190	17.5–7,360	1,060–4,300	17.5–7,360	4.2×10^2
La	<0.08–10	0.06–0.92	<0.13–<8.0	–	0.06–10	1.7×10^2
Li	–	0.23–1.1	<0.05–0.23	0.0036–3.6	0.0036–3.6	1.0×10^3
Lu	<0.08	–	<0.004–0.017	–	<0.004–0.017	>4.3
Mg	700–4,000	1,000–5,500	100–6,100	120–2,700	100–6,100	6.1×10^1
Mn	0.2–100	0.11–10.1	0.06–140	0.01–179	0.01–179	1.8×10^4
Mo	<0.7	–	<0.01–67	0.8–115	<0.01–115	$>1.2 \times 10^4$
N (g/kg)	43–45	114–130	39–130	27–43	27–130	4.8
Na	5,000–10,000	3,300–34,000	3,000–14,800	600–8,300	600–34,000	5.7×10^1
Nb	<0.07	–	<0.07–<0.1	–	<0.07–<0.1	–
Nd	<0.28	–	<0.02–1.0	–	<0.02–1.0	$>5.0 \times 10^1$
Ni	<0.7–2.5	1.3–2.1	0.14–71.5	2.4–<6	0.14–71.5	5.1×10^2
O (g/kg)	285	138–381	72–405	394–446	72–446	6.2

Table 5 continued

Element	Published data					Ratio max/min combined data
	(Bowen 1966; 1979)	(Zwanziger 1989)	(Iyengar et al. 1978; Iyengar and Tandon 1999)	Data published in Russia (own data)	Combined data	
P (g/kg)	62–120	0.1–206	49–540	0.2–117	0.1–540	$5.4 \cdot 10^{3}$
Pb	3.6–30	4–410	0.23–840	8.0–491	0.23–840	3.7×10^{3}
Po	–	–	$(3–260) \times 10^{-12}$	–	$(3–260) \times 10^{-12}$	8.7×10^{1}
Pr	–	–	<0.01–0.3	–	<0.01–0.3	$>3.0 \times 10^{1}$
Pt	–	–	<0.06	–	<0.06	–
Ra	4×10^{-9}	–	$(1.1–7.5) \times 10^{-9}$	–	$(1.1–7.5) \times 10^{-9}$	6.8
Rb	0.9–6.8	<0.04–62	0.1–48	12	<0.04–62	$>1.6 \times 10^{3}$
Re	–	–	<0.004	–	<0.004	–
S	500–2,400	–	500–800	2,000–5,200	500–5,200	1.04×10^{1}
Sb	0.01–0.6	0.004–0.081	<0.0018–1.0	0.24	<0.0018–1.0	$>5.6 \times 10^{2}$
Sc	0.001–6.0	<0.001–0.30	0.00005–5.5	–	0.00005–6	1.2×10^{5}
Se	109	0.097–0.93	<0.065–8.95	–	<0.065–109	$>1.7 \times 10^{3}$
Si	17	–	17	0.2–1,052	0.2–1,052	5.3×10^{3}
Sm	0.23	–	<0.004–0.2	–	<0.004–0.23	$>5.8 \times 10^{1}$
Sn	1.4–6	–	<0.21–7.2	1.0–<1.5	<0.21–7.2	$>3.4 \times 10^{1}$
Sr	30–140	<0.1–270	32–714	4.8–3,183	<0.1–3,183	$>3.2 \times 10^{4}$
Ta	0.03	–	<0.008–0.07	–	<0.008–0.07	>8.8
Tb	0.05	–	<0.004–0.04	–	<0.004–0.05	$>1.25 \times 10^{1}$
Te	–	–	<0.3	–	<0.3	–
Th	0.002–0.012	–	<0.003–0.55	–	0.002–0.55	2.75×10^{2}
Ti	–	0.48–0.52	0.3–4.6	0.024–59	0.024–59	2.5×10^{3}
Tl	0.002	0.0014–0.003	<0.0024–0.008	–	0.0014–0.008	5.7
Tm	<0.08	–	<0.004	–	<0.004–0.08	$>2.0 \times 10^{1}$
U	$(1.6–700) \times 10^{-7}$	–	0.0002–0.26	<0.0012–0.002	1.6×10^{-7}–0.26	1.6×10^{6}
V	0.0035–1	<0.01–1.74	0.7–21.2	0.06–<1.8	0.0035–21.2	6.1×10^{3}
W	0.00025	$(<0.1–0.7) \times 10^{-3}$	0.00025–1.2	0.0023	<0.0001–1.2	$>1.2 \times 10^{4}$
Y	0.04–0.07	–	<0.03–0.3	–	<0.03–0.3	$>1.0 \times 10^{1}$
Yb	<0.28	–	<0.004–0.02	–	<0.004–0.28	7.0×10^{1}
Zn	75–170	1.22–900	<31–1,060	4.2–299	1.22–1,060	8.7×10^{2}
Zr	<0.1	–	<0.065–89	–	<0.065–89	$>1.4 \times 10^{3}$

1995), no change in Ca and P content was observed for subjects up to the age of 90 years.

The mass fractions of Fe in the male rib bone increase significantly with age (Table 3). A positive Fe-age correlation in the rib bone was also observed by Yoshinaga et al. (1995). In contrast, age variations for the studied trace elements Cu, Sr, and Zn were found to be not significant. This is in agreement with the data by Sowden and Stitch (1957), Nusbaum et al. (1965), Anke et al. (1978), Tanaka et al. (1981), Patti et al. (1984), and Schuhmacher et al. (1992). Nevertheless, a positive Zn-age correlation in the rib bone was observed by Yoshinaga et al. (1995), and a negative Sr-age correlation was reported by Schneider and Anke (1971).

Comparison of mass fractions of rare earth elements (REEs) in the ribs of the two age groups revealed a statistically significant increase with age of Ce, Dy, Er, Gd, La, Nd, Pr, Sm, Tb, and Yb (Table 3). Thus, the present results indicate an age-dependent accumulation for those REEs, for which the determination of average mass fractions and other statistical features was possible. The observed age-dependent increase of the REE mass fractions is better fitted by an exponential relation than by a linear, polynomial, logarithmic, or power relation (Zaichick et al. 2011b). However, the regression parameters (R^2) obtained for most of the REEs are very low, and differences between the different relations are not significant. If this exponential increase of the mass

Table 6 Median and minimum and maximum values of means/medians of elemental mass fractions (mg/kg on a wet mass basis) and max/min ratios of the elemental mass fractions in human rib bone according to the data from the literature

Element	Median (*n*)	Minimum (references)	Maximum (references)	Max/Min
Ag	0.10 (4)	0.00254 (Zhu et al. 2010)	0.33 (Hamilton 1979)	1.3×10^{2}
Al	8.5 (5)	2.4 (Kehoe et al. 1940)	26 (Yoshinaga et al. 1989)	1.1×10^{1}
As	0.038 (6)	0.013 (Nusbaum et al. 1965)	0.15 (Bacso et al. 1993)	1.15×10^{1}
Au	<0.009 (4)	0.000225 (Zhu et al. 2010)	<0.06 (Yoshinaga et al. 1995)	$<2.7 \times 10^{2}$
B	1.8 (2)	0.187 (Zhu et al. 2010)	3.0 (Hamilton 1979)	1.6×10^{1}
Ba	5.75 (12)	0.6 (Schroeder et al. 1972)	262 (Jaritz et al. 1998)	4.4×10^{2}
Be	0.00081 (1)	0.00081 (Zhu et al. 2010)	0.00081 (Zhu et al. 2010)	–
Bi	<0.006 (3)	0.000555 (Zhu et al. 2010)	<0.13 (Yoshinaga et al. 1995)	2.3×10^{2}
Br	2.6 (7)	0.472 (Zhu et al. 2010)	≤15 (Forssen 1972)	3.2×10^{1}
Ca (g/kg)	116 (34)	48 (Schroeder et al. 1972)	253 (Suzuki 1979)	5.3
Cd	0.6 (14)	0.057 (Yoshinaga et al. 1989)	1.7 (Samudralwar and Robertson 1993)	3.0×10^{1}
Ce	0.037 (3)	<0.02 (Zaichick et al. 2009)	1.7 (Brättter et al. 1977)	$>8.5 \times 10^{1}$
Cl	590 (9)	289 (Takata et al. 2005)	1,626 (Bacso et al. 1993)	5.6
Co	0.075 (8)	0.0012 (Zaichick et al. 2009)	13.5 (Nusbaum et al. 1965)	1.1×10^{4}
Cr	1.2 (9)	0.062 (Sumino et al. 1975)	10.0 (Nusbaum et al. 1965)	1.6×10^{2}
Cs	0.024 (5)	<0.0025 (Yoshinaga et al. 1995)	0.030 (Hamilton 1979)	$>1.2 \times 10^{1}$
Cu	3.6 (21)	0.12 (Yoshinaga et al. 1995)	8.2 (Nusbaum et al. 1965)	6.8×10^{1}
Dy	<0.006 (3)	0.00152 (Zhu et al. 2010)	<0.27 (Hamilton 1979)	$<1.8 \times 10^{2}$
Er	<0.006 (3)	0.000421 (Zhu et al. 2010)	<0.21 (Hamilton 1979)	$<5.0 \times 10^{2}$
Eu	0.0015 (5)	<0.0006 (Zaichick et al. 2009)	<0.12 (Hamilton 1979)	$\sim 2.0 \times 10^{2}$
F	626 (13)	75 (Suzuki 1979)	2,544 (Zipkin et al. 1985)	3.4×10^{1}
Fe	127 (14)	11.8 (Saiki et al. 1999)	532(Takata et al. 2005)	4.5×10^{1}
Ga	≤0.08 (2)	<0.02 (Yoshinaga et al. 1995)	0.137 (Zhu et al. 2010)	>6.9
Gd	<0.005 (4)	0.00222 (Zhu et al. 2010)	<0.30 (Hamilton 1979)	$<1.4 \times 10^{2}$
Hf	<0.07 (2)	<0.013 (Zaichick et al.2009)	<0.13 (Yoshinaga et al. 1995)	$\sim 1.0 \times 10^{1}$
Hg	0.01 (5)	<0.005 (Zaichick et al. 2009)	2.1 (Brättter et al. 1977)	$>4.2 \times 10^{2}$
Ho	<0.0025 (3)	0.00010 (Zhu et al. 2010)	<0.06 (Hamilton 1979)	$<6.0 \times 10^{2}$
K	841 (12)	30.2 (Yoshinaga et al. 1995)	2,857 (Forssen 1972)	9.5×10^{1}
La	<0.06 (5)	0.027 (Zhu et al. 2010)	0.54 (Brättter et al. 1977)	2.0×10^{1}
Li	<0.03 (1)	<0.03 (Yoshinaga et al. 1995)	<0.03 (Yoshinaga et al. 1995)	–
Lu	<0.0025 (4)	0.000539 (Zhu et al. 2010)	<0.06 (Hamilton 1979)	$<1.1 \times 10^{2}$
Mg	1,454 (18)	392 (Crawford and Crawford 1969)	2,644 (Takata et al. 2005)	6.7
Mn	2.2 (18)	0.074 (Sumino et al. 1975)	6.1 (Anke et al. 1978)	8.2×10^{1}
Mo	<0.05 (6)	0.0208 (Zhu et al. 2010)	32.6 (Nusbaum et al. 1965)	1.6×10^{3}
Na	3,305 (15)	196 (Utsumi et al. 1999)	5,589 (Takata et al. 2005)	2.9×10^{1}
Nb	<0.04 (2)	<0.02 (Hamilton 1979)	<0.06 (Yoshinaga et al. 1995)	~ 3
Nd	<0.012 (4)	0.0106 (Zhu et al. 2010)	<0.21 (Hamilton 1979)	$<2.0 \times 10^{1}$
Ni	1.6 (11)	0.23 (Sumino et al. 1975)	34.8 (Nusbaum et al. 1965)	1.5×10^{2}
P (g/kg)	57.8 (22)	25 (Bacso et al. 1993)	113 (Takata et al. 2005)	4.5
Pb	7.5 (38)	0.35 (Sumino et al. 1975)	59 (Crawford and Crawford 1969)	1.7×10^{2}
Pr	<0.006 (3)	0.0034 (Zhu et al. 2010)	<0.06 (Hamilton 1979)	$<1.8 \times 10^{1}$
Pt	<0.02 (2)	0.00071 (Zhu et al. 2010)	<0.064(Yoshinaga et al. 1995)	$<9.0 \times 10^{1}$
Rb	1.3 (7)	<0.05 (Yoshinaga et al. 1995)	3.2 (Zhu et al. 2010)	$>6.4 \times 10^{1}$
Re	<0.0025 (1)	<0.0025 (Yoshinaga et al. 1995)	<0.0025 (Yoshinaga et al. 1995)	–
S	1,492 (4)	967 (Utsumi et al. 1999)	5,500 (Zhu et al. 2010)	5.7
Sb	<0.06 (4)	0.00886 (Zhu et al. 2010)	0.50 (Hamilton, 1979)	5.6×10^{1}
Sc	<0.06 (3)	0.000779 (Zhu et al. 2010)	0.117 (Brättter et al. 1977)	1.5×10^{2}
Se	0.12 (3)	0.11 (Bacso et al. 1993)	<1.3 (Yoshinaga et al. 1995)	$<1.2 \times 10^{1}$

Table 6 continued

Element	Median (*n*)	Minimum (references)	Maximum (references)	Max/Min
Sm	<0.0025 (4)	0.00168 (Zhu et al. 2010)	<0.3 (Hamilton 1979)	$<1.8 \times 10^2$
Sn	0.5 (5)	0.0128 (Zhu et al. 2010)	4.5 (Koch et al. 1956)	3.5×10^2
Sr	45.8 (15)	21 (Bacso et al. 1993)	252 (Brättter et al. 1977)	1.2×10^1
Ta	<0.005 (1)	<0.005 (Yoshinaga et al. 1995)	<0.005 (Yoshinaga et al. 1995)	–
Te	<0.2 (1)	<0.2 (Yoshinaga et al. 1995)	<0.2 (Yoshinaga et al. 1995)	–
Th	<0.012 (4)	0.00182 (Zhu et al. 2010)	<0.04 (Yoshinaga et al. 1995)	$<2.2 \times 10^1$
Ti	<2 (3)	0.983 (Zhu et al. 2010)	<3 (Yoshinaga et al. 1989)	<3.1
Tl	<0.005 (3)	0.000792 (Zhu et al. 2010)	<0.18 (Hamilton 1979)	$<2.3 \times 10^2$
Tm	<0.0025 (3)	0.000140 (Zhu et al. 2010)	<0.059 (Hamilton 1979)	$<4.2 \times 10^2$
U	0.0028 (8)	0.00158 (Zhu et al. 2010)	<0.0059 (Hamilton 1979)	<3.7
V	≤0.05 (6)	0.0021 (Byrne and Kosta, 1978)	0.2 (Sumino et al. 1975)	9.5×10^1
W	<0.05 (1)	<0.05 (Yoshinaga et al. 1995)	<0.05 (Yoshinaga et al. 1995)	–
Y	0.021 (4)	0.0027 (Zhu et al. 2010)	≤25 (Forssen 1972)	9.3×10^3
Yb	<0.02 (4)	0.000623 (Zhu et al. 2010)	<0.21 (Hamilton 1979)	$<3.4 \times 10^2$
Zn	61 (26)	16 (Koch et al. 1957)	139 (Takata et al. 2005)	8.7
Zr	0.0567 (4)	<0.03 (Hamilton 1979)	<0.2 (Yoshinaga et al. 1995)	~6.7

n number of references, *M* arithmetic mean, *SEM* standard error of the mean, ≤*M* possible upper limit of the mean value, <*M* all the individual values are below the detection limit

fractions of REEs in the ribs of healthy people living in a non-industrial, ecologically safe region will be confirmed by additional studies, this could be interpreted as the result of a global increase of the concentrations of La and lanthanides in the environment.

Gender-related differences in elemental mass fractions of rib bone

It was pointed out by Schneider and Anke (1971), Schroeder et al. (1972), Anke et al. (1978), and Yoshinaga et al. (1989, 1995) that in women, the rib bone mass fractions of Ca, P, and Sr are higher than those for men. Our results also indicate this (Table 4). In fact, gender-related differences were detected for Ba, Ca, Fe, Mo, Na, P, Sr, and Zn mass fractions for all the age groups studied. Note, however, that statistically significant differences became apparent only for the entire age interval (15–55 years), for both females and males. Higher mass fractions of Ba, Mo, Na, and a lower mass fraction of Fe in the female rib as compared to the male rib have not been reported previously. Elevated levels of Ca, P, and lower level of Fe in the bone tissue of women compared with men can be attributed to physiological characteristics of the female body related to menstruation and reproduction. A high mass fraction of Ba, Na, and Sr in the bones of women is likely due to differences in diet preferences. Usually, Central European region of Russia females consume more vegetable foods than males, which is the main supplier of Ba and Sr in the human body, and more marinated and salted appetizers, which contain salt (NaCl). Note that salt is one of the main sources of Na in nutrition. Elevated levels of Mo may be attributed to a much more frequent use of costume jewelry by women, because stainless steel contains Mo both as an alloying ingredient and a cheap substitute for platinum. The lower Zn mass fraction in bones of male compared with female can be explained by the specific metabolism of this element associated with the physiology of the male reproductive system. A lower content of Zn in male ribs, as compared to female ribs, has not been reported previously.

No statistically significant gender-related differences were detected for all the REEs studied. But, according to the results shown in Table 4, it appears that REEs' mass fractions (for Ce, Dy, Er, Gd, La, Nd, Pr, Tb, and Yb) for males are either equivalent to or higher than those of females, but never lower. Of course, the involved uncertainties are large, and differences between mass fractions for males and females may not be significant for each REE. However, when considering all the REEs, the present results suggest that REEs' mass fractions may be higher in males' rib bones than in females' rib bones. This observation needs, however, to be confirmed by further measurements. No published data referring to any gender dependence of REEs' mass fractions in human bone have been found.

Table 7 Comparison of the variations in the max/min ratios of the elemental mass fractions in bone arising from analytical uncertainty alone, from analytical and elemental variability combined, and between those of this work and those of Zhu et al. (2010)

Range of max/min ratio (scale)	Reference material	Human various bones	Human rib bone	
	Analytical uncertainty alone[a]	Analytical and elemental variability combined[b]	Analytical and elemental variability combined[c]	Comparison this work results and Zhu et al. (2010)[d]
1.0–<1.1	–	–		Ca, Cr, Cu, Fe, Hg, Mg, Na, P, Th, Y
1.1–<1.2	–	–		Zn
1.2–<1.3	–	–		Dy, K, Pb, Se
1.3–<1.5	–	–		Cl, Ga, Yb, Pt, Sc
1.5–<2.0	–	H		Al, Ba, Ce, Cs, Er, Mo, Nd, Pr, Sb, Sm, Ti, U, V
2.0–<10	Ba, Ca, F, Mo	Dy, Hf, Lu, N, O, Ta, Tl, Ra	Ca, Cl, Ga, Mg, Nb, P, S, Ti, U, Zn, Zr	Ag, Au, B, Be, Br, Cd, Eu, Gd, Ho, La, Lu, Mn, Ni, Rb, S, Sn, Sr, Tb, Tl, Tm, Zr
10–<102	K, Pb, Rb, S, Si, Sr	Be, C, Cl, Er, Eu, Ga, Ho, I, Mg, Na, Nd, Po, Pr, S, Sm, Sn, Tb, Tm, Y, Yb	Al, As, B, Br, Cd, Ce, Cs, Cu, F, Fe, Hf, K, La, Mn, Na, Nd, Pr, Pt, Rb, Sb, Se, Sr, Tb, Th, V	As, Bi, Co
10^2–<10^3	Al, Cd, Cl, Mg, Na, Sb, Zn	B, Ba, Cr, Cs, F, Hg, K, La, Ni, Sb, Th, Zn	Ag, Au, Ba, Bi, Cr, Dy, Er, Eu, Gd, Hg, Ho, Lu, Ni, Pb, Sc, Sm, Sn, Tl, Tm, Yb	–
10^3–<10^4	As, Br, Fe	As, Ca, Co, Fe, Li, P, Pb, Rb, Se, Si, Ti, V, Zr	Mo, Y	–
10^4–<10^5	Cr, Mn, Ni, P, Se	Ag, Al, Br, Cd, Cu, Mn, Mo, Sr, W	Co	–
10^5–<10^6	Co, Cu, Hg	Au, Sc		–
$>10^6$	V	U		–

[a] Results based on IAEA H-5 Animal Bone material (Parr 1982; Zaichick 2006b)

[b] Based on the data from the literature (see Table 5)

[c] Based on the present review of the data in the literature (see Table 6)

[d] Based on the results of this work (see Table 1) and those of Zhu et al. (2010) (see Table 8)

Analytical uncertainty and individual variability of elemental mass fractions in bones

Levels of variation (maximum/minimum ratio of means/medians) reported in the literature arising from both analytical uncertainty and individual variability of elemental mass fractions in human bones are surprisingly wide (Table 5). To eliminate the effect of bone differences on the levels of these reported variations, a review of the published data on elemental mass fractions in human rib bones, published in the period from 1940 to 2010, was prepared (Table 6). The means and the corresponding upper limits obtained in the present work for Ag, Al, B, Ba, Be, Bi, Br, Ca, Ce, Cl, Co, Cr, Cs, Cu, Er, F, Fe, Hg, Ho, K, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Se, Sr, and Zn, as shown in Table 1 (last column), agree well with ranges of the mean values cited by other researchers for the human rib bone (including samples received from persons, who died from different diseases) (Table 6). The mean values for Cd, Gd, Dy, La, Li, Nd, Pr, Sm, Tb, Tl, U, and Yb (Table 1, last column) are somewhat lower than the minimum mean value of previously reported data (Table 6). The upper limit of means for Eu, Lu, Sb, Th, Ti, Tm, and Y (Table 1, last column) is nearly equal to two orders of magnitude and for Te (Table 1) is two orders of magnitude lower, than previously reported results (Table 6). The detection limits of ICP-MS show that the content of Re in intact rib bone of healthy men (Table 1, last column) is at least one order of magnitude lower than previously reported data (Table 6). No published data referring to Ir, Pa, Pd, and Rh mass fractions of human rib bone were found. Levels of variation (max/min ratio of means/medians) arising from both analytical uncertainty and individual variability of elemental mass fractions in human rib bone separately, according to the data published in the literature (Table 6), show some values lower than those for various human bones (Table 5); however, the differences are minor.

Thus, it seems reasonable to assume that the enormous level of variation observed arises mainly from the analytical uncertainty. This assumption may well be illustrated

Table 8 Median of means/medians of 79 element mass fractions (mg/kg on a wet mass basis) in human bone (adult) from the literature in comparison with the present results

Element	Published data (for human bones)				This work result (for human rib bone only)		
	Bone mineral ICRP-89 (2002) period up to 2002 mean	Various bones (Iyengar et al. 1978) period up to 1978 Median of means/medians	Various bones (Iyengar and Tandon 1999) period 1978–1999 median of means/medians	Rib bone (Zhu et al. 2010) Median	Our review of published data Period 1940–2010 median of means/medians	Human intact rib bone (see Table 1) mean	Accepted values of mass fraction mg/kg wet tissue
Ag	–	0.59 (3)	0.033 (5)	0.00254	0.10 (4)	≤0.0065	≤0.007
Al	–	31 (7)	19.5 (6)	7.69	8.5 (5)	≤4.7	≤5
As	–	1.1 (2)	0.0059 (3)	0.102	0.038 (6)	<0.006	<0.006
Au	–	0.016 (2)	≤0.00039 (1)	0.00023	<0.009 (4)	<0.0004	<0.0004
B	–	0.9 (3)	16.4 (3)	0.19	1.8 (2)	≤0.42	≤0.4
Ba	–	4.2 (9)	15.2 (8)	3.2	5.75 (12)	1.61	1.6
Be	–	≤0.01 (3)	–	0.00081	0.00081 (1)	≤0.0020	≤0.002
Bi	–	<0.1 (1)	<0.17 (1)	0.00056	<0.006 (3)	0.0087	0.0087
Br	–	38 (1)	9.4 (5)	0.47	2.6 (7)	≤2.5	≤2.5
C	160,000	–	–	–	264,000 (1)	–	264,000
Ca	215,000	170,000 (10)	187,500 (28)	110,000	116,000 (34)	116,000	116,000
Cd	–	1.8 (2)	0.17 (13)	0.075	0.6 (14)	0.028	0.028
Ce	–	–	–	0.037	0.037 (3)	0.018	0.018
Cl	–	632 (1)	850 (7)	930	590 (9)	621	621
Co	–	0.038 (5)	0.08 (9)	0.034	0.075 (8)	0.0016	0.0016
Cr	–	6 (3)	2.0 (8)	0.13	1.2 (9)	≤0.14	≤0.14
Cs	–	0.05 (5)	0.03 (3)	0.0089	0.024 (5)	≤0.0049	≤0.005
Cu	–	4.2 (5)	1.2 (16)	0.70	3.6 (21)	0.62	0.62
Dy	–	–	<0.01 (1)	0.0015	<0.006 (3)	0.0013	0.0013
Er	–	–	<0.01 (1)	0.0004	<0.006 (3)	0.0007	0.00070
Eu	–	–	0.013 (2)	0.0015	0.0015 (5)	≤0.0006	≤0.0006
F	–	2,820 (9)	539 (6)	–	626 (13)	978	978
Fe	–	198 (6)	210 (18)	81.2	127 (14)	83	83
Ga	–	–	<0.025 (1)	0.14	≤0.08 (2)	<0.1	≤0.08
Gd	–	–	<0.007 (1)	0.0022	<0.005 (4)	0.0010	0.0010
Ge	–	–	<0.06 (1)	0.089	≤0.07 (2)	–	≤0.07
H	35,000	–	–	–	63,500 (1)	–	63,500
Hf	–	–	<0.2 (1)	–	<0.07 (2)	<0.01	<0.01
Hg	–	≤0.45 (2)	0.015 (3)	0.010	0.01 (5)	≤0.0096	≤0.01
Ho	–	–	<0.003 (1)	0.00010	<0.0025 (3)	≤0.00033	≤0.0003
I	–	15 (1)	–	0.23	0.23 (1)	–	0.23
In	–	–	<0.008 (1)	0.00015	0.00015 (1)	–	0.00015
Ir	–	–	–	–	–	<0.00002	<0.00002
K	–	1,470 (1)	356 (5)	1,140	841 (12)	863	863
La	–	≤3.9 (2)	<0.25 (1)	0.027	<0.06 (5)	0.0123	0.012
Li	–	–	≤0.12 (2)	–	<0.03 (1)	0.024	0.024
Lu	–	–	0.005 (2)	0.00054	<0.0025 (4)	≤0.00015	≤0.00015
Mg	2,000	1,800 (5)	2,160 (19)	1,450	1,454 (18)	1,345	1,450
Mn	–	1.9 (6)	2.0 (12)	0.448	2.2 (18)	0.165	0.165
Mo	–	55.6 (1)	5.5 (3)	0.021	<0.05 (6)	0.033	0.033
N	4,2000	–	40,800 (5)	–	48,000 (1)	–	48,000
Na	3,000	8,535 (4)	4,780 (15)	3,480	3,305 (15)	3,222	3,260
Nb	–	<0.07 (1)	<0.08 (1)	–	<0.04 (2)	<0.006	<0.006

Table 8 continued

Element	Published data (for human bones)				This work result (for human rib bone only)		
	Bone mineral ICRP-89 (2002) period up to 2002 mean	Various bones (Iyengar et al. 1978) period up to 1978 Median of means/medians	Various bones (Iyengar and Tandon 1999) period 1978–1999 median of means/medians	Rib bone (Zhu et al. 2010) Median	Our review of published data Period 1940–2010 median of means/medians	Human intact rib bone (see Table 1) mean	Accepted values of mass fraction mg/kg wet tissue
Nd	–	–	<0.017 (1)	0.011	<0.012 (4)	0.0067	0.0067
Ni	–	59.4 (1)	1.2 (10)	1.95	1.6 (11)	≤0.64	≤0.64
O	445,000	–	499,000 (5)	–	442,000 (1)	–	442,000
P	95,000	93,350 (4)	86,320 (17)	55,100	57,800 (22)	50,700	57,800
Pa	–	–	–	–	–	<0.04	<0.04
Pb	–	23.2 (13)	7.0 (33)	1.12	7.5 (38)	1.42	1.42
Pd	–	–	–	–	–	<0.006	<0.006
Po	–	5.9×10^{-12} (3)	8.2×10^{-12} (2)	–	–	–	8×10^{-12}
Pr	–	–	<0.008 (1)	0.0034	<0.006 (3)	0.0021	0.0021
Pt	–	–	<0.05 (1)	0.0007	<0.02 (2)	<0.001	<0.001
Ra	–	6.8×10^{-9} (5)	1.09×10^{-9} (1)	–	–	–	1×10^{-9}
Rb	–	1.92 (3)	2.87 (10)	3.2	1.3 (7)	0.94	1.0
Re	–	–	<0.003 (1)	–	<0.0025 (1)	<0.0003	<0.0003
Rh	–	–	–	–	–	<0.006	<0.006
S	3,000	500 (1)	664 (1)	5,500	1,492 (4)	1,169	1,170
Sb	–	0.3 (3)	≤0.010 (4)	0.0089	<0.06 (4)	≤0.0056	≤0.006
Sc		2.3 (2)	0.064 (4)	0.0008	<0.06 (3)	<0.0006	<0.0006
Se	–	5 (2)	0.17 (6)	0.12	0.12 (3)	≤0.11	≤0.11
Si	–	17 (1)	–	–	–	–	17
Sm	–	–	<0.003 (1)	0.0017	<0.0025 (4)	0.0009	0.0009
Sn	–	3 (3)	≤1.3 (2)	0.013	0.5 (5)	<0.1	<0.1
Sr	–	79 (15)	69 (19)	42.3	45.8 (15)	177	180
Ta	–	–	0.022 (2)	–	<0.005 (1)	<0.003	<0.003
Tb	–	–	0.0070 (3)	0.0011	<0.0025 (4)	0.0002	0.0002
Te	–	–	<0.25 (1)	–	<0.2 (1)	≤0.0029	≤0.003
Th	–	0.012 (6)	0.13 (2)	0.0018	<0.012 (4)	≤0.0019	≤0.002
Ti	–	–	1.62 (4)	0.98	<2 (3)	≤1.7[a]	≤2[a]
Tl	–	0.002 (1)	<0.007 (1)	0.00079	<0.005 (3)	0.0003	0.0003
Tm	–	–	<0.003 (1)	0.00014	<0.0025 (3)	≤0.00004	≤0.00004
U	–	0.0077 (7)	0.0059 (12)	0.0016	0.0028 (8)	0.0008	0.0008
V	–	0.87 (3)	2.0 (4)	0.034	≤0.05 (6)	<0.02	<0.02
W	–	0.00025 (1)	0.0028 (2)	–	<0.05 (1)	<0.06	<0.06
Y	–	0.07 (1)	<0.025 (1)	0.0027	0.021 (4)	≤0.0029	≤0.003
Yb	–	–	<0.003 (1)	0.0006	<0.02 (4)	0.0005	0.0005
Zn	–	100 (9)	85 (24)	50	61 (26)	56.9	57
Zr	–	<0.05 (1)	≤13 (2)	0.057	0.057 (4)	<0.02	<0.02
							Σ ≤ 999,999.5 mg/kg

Data in the last column for C, H, N, and O are from Woodard and White (1982)

[a] titanium tools were used for sampling and sample preparation

n number of references, ≤*M* possible upper limit of the median of mean value, <*M* all the mean/median values are below the detection limit

Table 9 Skeleton (sum of all skeleton tissues) burdens of 79 elements (g) in different Reference individuals

Element	ICRP-89[a] Reference Man/Woman $m_s = (10.5/7.8)$ kg ICRP 2002	Chinese Reference Man $m_s = 8.0$ kg Zhu et al. 2010	Russian Reference Man/Woman (Caucasian, European living habits)		
			Skeleton burdens (human) $m_s = 10.0$ kg	Skeleton burdens (male) $m_s = 10.5$ kg	Skeleton burdens (female) $m_s = 7.8$ kg
Ag	–	0.000020	≤0.00007	≤0.00007	≤0.00006
Al	–	0.062	≤0.05	≤0.05	≤0.04
As	–	0.00082	<0.00006	<0.00006	<0.00005
Au	–	0.0000018	<0.000004	<0.000004	<0.000003
B	–	0.0015	≤0.004	≤0.004	≤0.003
Ba	–	0.026	0.016	0.014	0.015
Be	–	0.0000065	≤0.00002	≤0.00002	≤0.00002
Bi	–	0.0000044	0.000087	0.000091	0.000068
Br	–	0.0038	≤0.025	≤0.025	≤0.020
C	3,075/2,265	–	2,640	2,800	2,050
Ca	1,180/860	880	1,160	1,130	985
Cd	–	0.00060	0.00028	0.00029	0.00022
Ce	–	0.00029	0.00018	0.00019	0.00014
Cl	3.3/2.7	7.44	6.2	6.6	4.8
Co	–	0.00027	0.000016	0.000017	0.000012
Cr	–	0.0011	≤0.0014	≤0.0015	≤0.0011
Cs	–	0.000071	≤0.00005	≤0.00005	≤0.00004
Cu	–	0.0056	0.0062	0.0065	0.0048
Dy	–	0.000012	0.000013	0.000014	0.000010
Er	–	0.0000034	0.0000070	0.0000074	0.0000055
Eu	–	0.000012	≤0.000006	≤0.000006	≤0.000005
F	–	–	9.8	10.3	7.6
Fe	1.2/0/9	0.65	0.83	1.05	0.48
Ga	–	0.0011	≤0.0008	≤0.0008	≤0.0006
Gd	–	0.000018	0.000010	0.000011	0.0000076
Ge	–	0.00071	≤0.0007	≤0.0007	≤0.0006
H	707/529	–	635	670	500
Hf	–	–	<0.0001	<0.0001	<0.00008
Hg	–	0.000080	≤0.0001	≤0.0001	≤0.00008
Ho	–	0.00000080	≤0.000003	≤0.000003	≤0.000002
I	–	0.0018	0.0023	0.0024	0.0018
In	–	0.0000012	0.0000015	0.0000016	0.0000012
Ir	–	–	<0.0000002	<0.0000002	<0.0000002
K	–	9.12	8.6	9.0	6.7
La	–	0.00022	0.00012	0.00013	0.00009
Li	–	–	0.00024	0.00025	0.00019
Lu	–	0.0000043	≤0.0000015	≤0.0000016	≤0.0000012
Mg	13.7/10.1	11.6	14.5	15.2	11.3
Mn	–	0.0036	0.0017	0.0018	0.0013
Mo	–	0.00017	0.00033	0.00027	0.00032
N	314/232	–	480	530	370
Na	25.7/19.2	27.9	32.6	32.2	26.6
Nb	–	–	<0.00006	<0.00006	<0.00005
Nd	–	0.000085	0.000067	0.000070	0.000052
Ni	–	0.016	≤0.006	≤0.006	≤0.005
O	4,373/3,277	–	4,420	4,780	3,400

Table 9 continued

Element	ICRP-89[a] Reference Man/Woman $m_s = (10.5/7.8)$ kg ICRP 2002	Chinese Reference Man $m_s = 8.0$ kg Zhu et al. 2010	Russian Reference Man/Woman (Caucasian, European living habits)		
			Skeleton burdens (human) $m_s = 10.0$ kg	Skeleton burdens (male) $m_s = 10.5$ kg	Skeleton burdens (female) $m_s = 7.8$ kg
P	558/409	441	578	500	427
Pa	–	–	<0.0004	<0.0004	<0.0003
Pb	–	0.0090	0.014	0.015	0.011
Pd	–	–	<0.00006	<0.00006	<0.00006
Po	–	–	8×10^{-14}	8×10^{-14}	8×10^{-14}
Pr	–	0.000027	0.000021	0.000022	0.000016
Pt	–	0.0000057	<0.00001	<0.00001	<0.00001
Ra	–	–	1×10^{-11}	1×10^{-11}	1×10^{-11}
Rb	–	0.026	0.010	0.011	0.0078
Re	–	–	<0.000003	<0.000003	<0.000002
Rh	–	–	<0.00006	<0.00006	<0.00005
S	31/24	44	12	13	9.0
Sb	–	0.000071	$\le$0.00006	$\le$0.00006	$\le$0.00005
Sc	–	0.0000062	<0.000006	<0.000006	<0.000005
Se	–	0.00094	$\le$0.0011	$\le$0.0012	$\le$0.0009
Si	–	–	0.17	0.18	0.13
Sm	–	0.000013	0.000009	0.0000095	0.000007
Sn	–	0.00010	<0.001	<0.001	<0.0008
Sr	–	0.338	1.8	1.6	1.6
Ta	–	–	<0.00003	<0.00003	<0.00002
Tb	–	0.0000084	0.000002	0.000002	0.0000016
Te	–	–	$\le$0.00003	$\le$0.00003	$\le$0.00002
Th	–	0.000015	$\le$0.00002	$\le$0.00002	$\le$0.00002
Ti	–	0.0079	$\le$0.02[b]	$\le$0.02[b]	$\le$0.02[b]
Tl	–	0.0000063	0.000003	0.000003	0.0000023
Tm	–	0.0000011	$\le$0.0000004	$\le$0.0000004	$\le$0.0000003
U	–	0.000013	0.000008	0.000008	0.000006
V	–	0.00028	<0.0002	<0.0002	<0.0002
W	–	–	<0.0006	<0.0006	<0.0005
Y	–	0.000022	$\le$0.00003	$\le$0.00003	$\le$0.00002
Yb	–	0.0000050	0.0000050	0.000005	0.000004
Zn	–	0.400	0.57	0.60	0.48
Zr	–	0.00045	<0.0002	<0.0002	<0.0002
			$\Sigma \le 10{,}000$ g	$\Sigma \le 10{,}500$ g	$\Sigma \le 7{,}800$ g

[a] calculated by us using the ICRP-89 (2002) data: Skeleton burden (g) = $\Sigma f_i \cdot m_j$ (g), where f_i is the value of *ith* elemental mass fraction(m_j) of *jth* skeleton tissue (bone mineral, active marrow, inactive marrow, cartilage, and teeth)

[b] titanium tools were used for sampling and sample preparation

$\le M$ possible upper limit of the mass fraction value, m_s mass of the skeleton

by the following historical example: In the late 1970s/early 1980s, the International Atomic Energy Agency (IAEA) was concerned about the poor reproducibility of chemical element analysis in biological samples. Therefore, it was decided to introduce several approaches of improving measurement quality, including the development of standard reference materials from natural biological substances. For this purpose, a large amount of certain biological materials (including animal bones) was dried, homogenized, and packaged in hermetically sealed containers. Samples of each biological material were sent to key analytical laboratories around the world for a quantitative analysis of chemical element mass fractions. The idea was that the average values of element mass fractions measured with acceptable reproducibility would provide appropriate reference values and could then be used as international standards for designated elements. The results were amazing. Analyses of the same element from the same biological sample received from various laboratories differed by many orders of magnitude (Zaichick 2006b). The ranges of variation (max/min ratios) of the element mass fractions obtained for the animal bone sample are presented in Table 7. The assumption expressed above is confirmed if one compares the IAEA results and the variations of data for the element mass fractions in human rib bones taken from the literature (Tables 5,6).

At the end of the twentieth century, a special system of measures of analytical quality control was developed to guarantee good reproducibility of results. This complex system includes regular intra-laboratory quality monitoring and inter-laboratory or external control, as well as the obligatory use of national or international standard materials for comparison. Comparison of the results obtained in the present work with those of Zhu et al. (2010), the most representative study on the subject with modern analytical quality control, shows that the level of variation (max/min ratios) arising from individual variability of element mass fraction in intact human rib bone is now much lower (Table 7): Although the comparison includes different populations (of various races and on different continents), the variations observed for means for all main bone elements (Ca, P, Mg, Na) are in a range of a few percent, and those for almost all of the trace elements do not exceed one order of magnitude.

Comparison with published data and accepted values of mass fractions

Data presented in Table 8 summarize current knowledge of elemental mass fractions in human bone including 79 elements, that is, the major portion of the 91 naturally occurring (not artificially synthesized) elements in the periodic table with only 12 elements, that is, noble gases and ultra-trace rare elements (Ru, Os, Pm, At, Fr, and Ac) being excluded. Table 8 demonstrates that the ICRP-89 Reference Man includes only nine reference values for bulk (C, H, N, O, S), major (Ca and P), and minor (Mg and Na) elements for human bone (ICRP-89 2002). In contrast, the studies by Iyengar et al. (1978) and by Iyengar and Tandon (1999) provide mass fractions for 48 and 69 elements, respectively. For rib bone of the Chinese Reference Man, mass fractions for 60 elements were recently determined by Zhu et al. (2010). In the present work, data published over a period of 70 years (1940–2010) were reviewed and mass fractions of 72 elements in rib bone were found (see Table 8). As follows from the data collected in Table 8, the values of many trace and ultra-trace element mass fractions obtained in the twentieth century differ greatly from those obtained in the last decade, which reflects the impact of improved analytical techniques and quality control. Therefore, the more recent findings may provide a more systematic and representative dataset than those published earlier, for either establishment of parameters relevant for an improved ICRP Reference Man or for further revision of the ICRP Reference Man.

The mass fraction of bulk elements C, H, N, and O in human rib bone published by Woodard and White (1982) was accepted for the bone tissue of the Russian Reference Man (Table 8, last column). According to our review, the median of means/medians of the Ge, I, and In mass fractions and the median values of Po and Ra mass fractions Iyengar and Tandon (1999) were also accepted for the Russian Reference Man. The value of the Ca mass fraction chosen in the present study (116,000 mg/kg on a wet mass basis) is equal to the median mass fraction obtained from 34 references. The data obtained in the present study for Mg and P were nearly equal to the medians calculated based on 18 and 22 references, respectively, and values of 1,450 and 57,800 mg/kg (on a wet mass basis) were accepted for the Russian Reference Man. For Na, the result of our review (about 3,300 mg/kg) and of our measurements (about 3,220 mg/kg) was quite similar and the mean value (3,260 mg/kg) was accepted for the Russian Reference Man. For all other elements, mass fractions obtained in the present study were accepted for the bone tissue of the Russian Reference Man (Table 8, last column).

It is shown in Table 8 that among 79 elements in the bone tissue, the Po mass fraction (8×10^{-12} mg/kg on a wet mass basis) is the lowest while that for O is the highest (442,000 mg/kg on a wet mass basis); thus, the range of elemental mass fractions in human rib bone covers 17 orders of magnitude from 10^{-1} to 10^{-18} kg/kg. The sum of accepted values of 79 element mass fractions in the bone tissue of the Russian Reference Man (0.9999995 kg/kg on a wet mass basis) is very close to 1 as it should be.

The mass fractions of N, Na, and O in the bone of the Russian Reference Man are close to, those of Ca, Mg, P, and S are 1.5–3.0 times lower, and those of C and H are 1.5–2.0 times higher than the corresponding mass fractions used for ICRP-89 Reference Man (2002).

Note that the mass fractions obtained in the present work differ somewhat from those used in the Russian State Standard (GOST18622-79 1981) for bone-tissue equivalent materials, for H, C, N, O, P, and Ca (in g/kg: H–40, C–156, N–44, O–443, P–105, and Ca–211).

Skeleton burdens of elements in the Russian Reference Man/Woman

The bone samples selected for the present study were taken from ribs, which are easy to sample at autopsy. In our previous studies, it was found, however, that there are some differences in elemental mass fractions of various bones (Zaichick 2004b, 2006a, 2007; Zaichick et al. 2000, 2009). A typical adult human skeleton consists of 206 bones. Thus, for the precise measuring of elemental mass fractions in the whole skeleton, it would be necessary to investigate 206 bones, which is practically not possible. However, for an approximate estimation of elemental ratios, there is an alternative option: The biggest differences in elemental composition are between compact (cortical) and trabecular (cancellous, spongy) bone (Zaichick 2006a). The densities of compact and trabecular fresh bone are typically of the order of 2.0 (1.8–2.3) g/cm^3 and 1.1 (0.8–1.4) g/cm^3, respectively. The rib bone samples contain both compact and trabecular bone and, therefore, the densities of the whole fresh adult skeleton (1.30 g/cm^3—MCRP-89, 2002) and fresh rib bone (1.41 g/cm^3—Woodard and White 1982) are quite similar. Thus, it was assumed here that the ribs' elemental mass fractions are representative to those of the skeleton in general. For example, this was also assumed in the estimation of elemental burdens in the skeleton of Chinese Reference Man (Zhu et al. 2010).

Using data on the accepted mass fractions (bulk elements C, H, N, and O and trace elements Ge, I, In, Po, and Ra) and on the skeleton's mass (ICRP-89 2002), total skeleton burdens of 79 elements in the bone tissue of the Russian Reference Man/Woman were calculated. For this purpose, the elemental mass fractions of Ba, Ca, Fe, Mo, Na, P, Sr, and Zn, that is, elements with statistically significant gender-related differences in mass fractions, for rib bone of males and females (see Table 4) were used (Table 9). For comparison, Table 9 also includes information on skeleton burdens of elements in the ICRP-89 Reference Man (2002), Iyengar's reevaluation (1998), and the Chinese Reference Man (Zhu et al. 2010).

The comparison shows that—for almost all elements—the values obtained in the present study for the Russian Reference Man more or less agree with those for the Chinese Reference Man (Zhu et al. 2010), with the skeleton burdens of As, Bi, and Co being the only exceptions (Table 9): The skeleton burden of As and Co in the Russian Reference Man/Woman is one order of magnitude lower than in the Chinese Reference Man. This may be associated with different level of these elements in the environment. For example, it was shown in studies with biomonitors that atmospheric levels of As and Co are at least one order of magnitude lower in Central Russia than in China (Zhang et al. 2003; Ermakova et al. 2004). In contrast, in the Russian Reference Man/Woman, the skeleton burden of Bi is one order of magnitude higher than in the Chinese Reference Man.

Note that in the skeleton of the Russian Reference Woman/Man, the burden of C, Ca, Fe (for men), H, Mg, Na, O, and P is nearly equal to that of Fe (women) and S is two times lower, and that of N and Cl is 1.5–2.0 times higher than those in the ICRP-89 Reference Man (2002).

The data shown in Tables 8, 9 suggest that elemental mass fractions in bone and those in the skeleton of the ICRP Reference Man need revision.

Conclusions

The combination of various analytical methods allowed measurement of the contents of no less than 69 elements in human rib bone samples of healthy adults. Mean values (M ± SD) for the elemental mass fractions (mg/kg on a wet mass basis) of 33 elements could be measured: Ba (1.62 ± 0.98), Bi (0.0087 ± 0.0070), Ca (116,000 ± 30200), Cd (0.028 ± 0.025), Ce (0.018 ± 0.013), Cl (621 ± 274), Co (0.0016 ± 0.0014), Cu (0.62 ± 0.23), Dy (0.0013 ± 0.0010), Er (0.00068 ± 0.00062), F (978 ± 441), Fe (83 ± 50), Gd (0.00097 ± 0.00071), K (863 ± 244), La (0.012 ± 0.010), Li (0.0236 ± 0.0095), Mg (1,345 ± 333), Mn (0.165 ± 0.081), Mo (0.033 ± 0.026), Na (3,222 ± 738), Nd (0.0067 ± 0.0054), P (50,700 ± 12,600), Pb (1.42 ± 0.80), Pr (0.0021 ± 0.0021), Rb (0.94 ± 0.38), S (1,169 ± 237), Sm (0.00089 ± 0.00077), Sr (177 ± 101), Tb (0.00024 ± 0.00015), Tl (0.00031 ± 0.00015), U (0.00082 ± 0.00059), Yb (0.00046 ± 0.00029), and Zn (57 ± 13). For 19 elements, upper limits of mean elemental mass concentrations were calculated: Ag $\leq$ 0.0065, Al $\leq$ 4.7, B $\leq$ 0.42, Be $\leq$ 0.0020, Br $\leq$ 2.5, Cr $\leq$ 0.14, Cs $\leq$ 0.0049, Eu $\leq$ 0.00060, Hg $\leq$ 0.0096, Ho $\leq$ 0.00033, Lu $\leq$ 0.00015, Ni $\leq$ 0.64, Sb $\leq$ 0.0056, Se $\leq$ 0.11, Te $\leq$ 0.0029, Th $\leq$ 0.0019, Ti $\leq$ 1.7, Tm $\leq$ 0.000040, and Y $\leq$ 0.0029. Finally, elemental mass concentrations of 17 elements were lower than their detection limits: As < 0.006, Au < 0.0004, Ga < 0.1, Hf < 0.01, Ir < 0.00002, Nb < 0.006, Pa < 0.04, Pd < 0.006, Pt < 0.001, Re < 0.0003, Rh < 0.006, Sc < 0.0006, Sn < 0.1, Ta < 0.003, V < 0.02, W < 0.06, and Zr < 0.02.

It was shown that Ca, Mg, and P mass fractions decrease while mass fractions of rare earth elements in the human rib increase with age. Gender-related differences were detected for Ba, Ca, Fe, Mo, Na, P, Sr, and Zn.

Since all the deceased were citizens of a small city in a non-industrial region, and none of them had suffered previously from any acute or chronic disorders, the data of the present study on 69 elements in intact rib bone may be representative for residents of the Central European region of Russia.

Using published and measured data, the mass fractions for 79 elements for the rib bone have been derived. Based on accepted elemental mass fractions for bone and reference values for the skeleton mass of Reference Man, the elemental burdens in the total skeleton were estimated. These results may provide a representative basis for establishing related reference values of the Russian Reference Man/Woman and revising and extending current reference values of the International Commission on Radiological Protection (ICRP) Reference Man. The data obtained in the present study will also be very valuable for many other applications in radiation protection, radioecology, radiotherapy dosimetry, medical radiology, radiobiochemistry, radiobiophysics, and other scientific fields.

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