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JUN 04 1991**THORIUM ISOTOPES IN HUMAN TISSUES***A. F. Stehney[†] and H. F. Lucas[‡]

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Abstract--Concentrations of ^{232}Th and activity ratios of ^{228}Th to ^{232}Th and ^{230}Th to ^{232}Th were determined in autopsy samples from five former employees of a thorium refinery. The ranges of ^{232}Th activity concentrations (mBq g^{-1}) were 0.17-94 in lungs, 3.9-1210 in pulmonary lymph nodes, 0.14-1.19 in bones, 0.015-0.68 in liver, 0.97-5.8 in spleen, and 0.009-0.068 in kidneys. These concentrations are 10 to 1000 times greater than have been reported for persons not occupationally exposed to Th. In most of the samples, the ratios of ^{230}Th to ^{232}Th and ^{228}Th to ^{232}Th activity at death of the subject were 0.1-0.2 and 0.2-0.4, respectively. Thorium-228 to ^{228}Ra activity ratios ($\pm$ standard errors) of 0.86 ± 0.11 in lungs and 1.18 ± 0.13 in lymph nodes of one subject were obtained by calculation from ratios of ^{228}Th to ^{232}Th .

Key words: ^{232}Th , ^{228}Th , ^{230}Th , occupational exposure**DISCLAIMER**

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INTRODUCTION

As part of a study of the health effects of industrial exposure to Th, our laboratory has measured the amounts of ^{232}Th and its radioactive descendants in former employees of a thorium refinery. The plant produced Th and rare-earth chemicals from ores and monazite sands from 1932 to 1973 and manufactured Th mantles from 1936 to 1947 (Stehney et al. 1980). An industrial hygiene survey of 84 plant workers in 1952 estimated daily weighted average exposures to airborne ^{232}Th of about 0.05 to 5 Bq m $^{-3}$ (Albert et al. 1955), and air filter measurements in the early 1960s showed similar concentrations at various locations in the plant (Mausner 1982). In vivo radioactivity measurements on about 300 former employees of the plant showed body contents of more than 4 Bq ^{212}Pb in 134 of the subjects and $^{224}\text{Ra}/^{228}\text{Ac}$ ratios ranging from 0.5 to 2 (Toohey et al. 1986, 1988). Radiochemical analyses of autopsy tissues from some of the former employees (Mausner 1982; Lucas and Stehney 1988) showed that the concentrations of ^{232}Th in lungs and other tissues were much greater than had been reported for non-occupationally exposed subjects (Lucas et al. 1970; Clifton et al. 1971; Ibrahim et al. 1983; Sunka et al. 1987). These findings and the apparently low uptake from the gastrointestinal (GI) tract (Maletskos et al. 1969) indicate that the high levels of Th in the former refinery employees were probably caused by inhalation of aerosols at the plant. The present work reports the results of measurements of ^{232}Th , ^{230}Th , and ^{228}Th in autopsy samples from five former thorium workers.

MATERIALS AND METHODS

The subjects' histories of employment at the thorium plant (Table 1) show a wide variety of work dates and job assignments. "Laborer" was a general category often assigned to new or temporary employees and included tasks such as shoveling monazite ores, plant maintenance, hauling process residues, and general clean-up. The work of subjects L0770 and L2932 and most of the early work of L2107 was only peripherally related to Th processing, and

all three of these men had lengthy service interruptions. Subject L2107 was rehired in 1954 as supervisor of Th production, which included preparation of Th nitrate and Th oxide, and L1521 became supervisor of rare-earth production after a long period of work involving the handling of monazite. The latter specifically mentioned working on production of "glass polishes", which are rare-earth compounds. Both L2107 and L1521 continued working at the plant until it closed in 1973. All subjects except L1521 smoked cigarettes.

Although systematic efforts were made to obtain autopsy specimens, we usually had little or no notice of an impending death, and the completeness of sampling depended on whether we were notified before the autopsy was performed. We were able to obtain specified tissues from subjects L0770 and L1521 by contacting the pathologists prior to autopsy, and our staff pathologist performed the autopsy on subject L2932. For subjects L1768 and L2107, the only samples available were those taken in the course of conventional autopsies. The samples were identified from autopsy reports and examination of the tissues and were prepared for radiochemical analysis by staff members who had performed numerous dissections of autopsy materials.

Prior to analysis, the samples were weighed and stored at about -10°C . While there was a 2-6% loss of weight during storage for some of the samples, the wet weight reported for each sample was adjusted to the original weight basis. Soft-tissue samples were minced, dried at 110°C , ashed with ionized oxygen until constant weight was obtained, and dissolved in 6 M HCl. Bone samples were scraped to remove soft tissue, then cut into small pieces, dried, ashed in platinum crucibles at 600°C , and dissolved in 12 M HCl. The sample solutions were filtered through Whatman #42 paper and processed for extraction of Th. The gross α -activity of undissolved residual material was determined by counting for 20 h or more in a low-background (1 to 2 c h^{-1}) 2 pi proportional counter. If the net counting rate was greater than twice the background, the residue was dissolved by repeated fuming with HNO_3 -HF and processed as a sample.

After addition of ^{229}Th or ^{234}Th to the sample solution as an isotopic diluent, thorium was chemically separated by precipitation with Ca oxalate, anion exchange from 8 M HNO_3 solution (Larsen and Urnezis 1976), and elution

with 12 M HCl (Mausner 1982). The Th was electrodeposited onto electrolytically polished stainless steel disks from a solution of $\text{H}_3\text{PO}_4\text{-H}_2\text{SO}_4$ that had been adjusted to a pH of 3 to 4 by addition of NH_4OH and HCl (Mausner 1982). During the course of the work, interference from ^{210}Po was noted in the determination of ^{228}Th in bone and other samples of internal organs, so the removal of Po by anion-exchange from 2 M HBr solution was added to the chemical procedures.

Thorium-232, ^{230}Th , and ^{228}Th were determined by α -particle spectrometry with surface-barrier detectors. Alpha-spectra were accumulated in 256 channels in the energy range of 3.5-9 MeV and entered into a computer file for assistance in the analysis and calculation of results. The zero intercept and energy per channel were checked by locating the ^{232}Th and ^{228}Th peaks, and the channels to be assigned to each isotope were determined by visual inspection of the spectra.

Thorium-228 activity in each sample at time of death of the subject was estimated by correcting for radioactive decay and for growth from ^{232}Th and ^{228}Ra . In the equations below, T_1 is the time from death to separation of Th; T_2 is the time from separation to count of α particles; $T = T_1 + T_2$; λ_2 is the radioactive decay constant of ^{228}Ra (half-life 5.75 y); and λ_3 is the radioactive decay constant of ^{228}Th (half-life 1.91 y). Since the decay constant of ^{232}Th is essentially equal to zero, the ratio of the activity of ^{228}Th at time of death to the activity of ^{232}Th may be expressed as

$$R_1 = \frac{\frac{D_3}{A_1} - F_1}{F_3 + \frac{F_2}{R_2}}, \quad (1)$$

where: D_3 is the activity of ^{228}Th at count time; A_1 is the activity of ^{232}Th ; R_2 is the ratio of ^{228}Th to ^{228}Ra at time of death;

$$F_1 = \frac{\lambda_3}{\lambda_3 - \lambda_2} \left[(1 - e^{-\lambda_2 T_2}) + e^{-\lambda_2 T_2} (1 - e^{-\lambda_1 T_1}) - \frac{\lambda_2}{\lambda_3} (1 - e^{-\lambda_3 T}) \right];$$

$$F_2 = \frac{\lambda_3}{\lambda_3 - \lambda_2} e^{-\lambda_3 T_2} (e^{-\lambda_2 T_1} - e^{-\lambda_3 T_1}) ; \quad \text{and} \quad F_3 = e^{-\lambda_3 T} .$$

In view of the lengthy exposure and post-exposure times of the subjects, the activities of ^{228}Th and ^{228}Ra in body tissues were assumed to be equal at the time of death (vide infra).

RESULTS

The ^{232}Th concentrations and ratios of ^{228}Th and ^{230}Th to ^{232}Th in autopsy samples from the former Th workers, corrected to the time of death, are shown in Table 2, along with the standard errors (SEs). Each line represents a separate sample; if two or more aliquots of a sample solution were analyzed, the values shown are the means of the values obtained for each aliquot weighted by the reciprocals of the variances, and the SEs are the errors calculated from the deviations from the mean or, if larger, the errors calculated from the counting data. The SEs of the $^{228}\text{Th}/^{232}\text{Th}$ ratios were calculated by applying to Eqn (1) the usual formulas for the propagation of errors (Bevington 1969), and they included an uncertainty of 20% in the assumed $^{228}\text{Th}/^{228}\text{Ra}$ ratio of 1.00 at time of death in order to impose a larger estimate of error on ratios obtained from Th separations made long after the death of the subject. Because ^{210}Po contamination was not observed in samples of lung, ratios of ^{228}Th to ^{232}Th are shown in Table 2 for all lung samples, but $^{228}\text{Th}/^{232}\text{Th}$ ratios for the other types of sample are shown only if anion exchange from HBr solution was used to remove ^{210}Po . Interference of ^{229}Th in the alpha-particle spectrum of ^{230}Th was observed in samples for which ^{229}Th had been used as the isotopic diluent, and the degree of interference depended on the relative amounts of ^{230}Th and ^{229}Th and the spectral resolution of the sample mount. No objective criteria could be established for accepting ^{230}Th values obtained in the presence of ^{229}Th , so ratios of ^{230}Th to ^{232}Th are shown only for samples analyzed with ^{234}Th as the diluent.

Table 2 includes results obtained by reanalysis of alpha-particle spectra previously obtained by Mausner (1982) from tissue samples of subjects L2107 and L2932. The results for bone and those shown as replicates for these two subjects and all results for the other subjects are new values obtained in the present work.

The ^{232}Th concentrations shown for the less easily dissolved material ("residues") have been normalized to the wet weights of the samples, so the total concentrations in the samples are the sum of the two values shown. The residues of the first two lung samples of subject L1521 were analyzed by alpha-particle spectrometry, but the ^{232}Th value for the third residue was obtained only from the gross alpha count. Ratios of 5.15 and 5.65 mBq ^{232}Th per count per minute (cpm) of gross alpha-particle activity were obtained for the residues of the first two samples (2.3 and 3.7 mg weights), and application of the mean ratio of 5.40 ± 0.35 mBq/cpm led to the value shown for the third residue (2.0 mg). The higher $^{228}\text{Th}/^{232}\text{Th}$ ratios in the residue than in the rest of the sample imply differences in aerosols and/or less translocation of ^{228}Ra out of the matrix of the residue than out of more easily dissolved material in the lung. The residue from the spleen sample of subject L1768 also contained measurable ^{232}Th activity, but less than one percent of the ^{232}Th activity of the sample was found in residues from one sample of lung from L0770, from five lung samples and one lymph node sample of L2107, and from one lung sample of L2932.

For some samples, it was possible to calculate the ratio of ^{228}Th to ^{228}Ra at death from the activities in two separations of Th from the sample that had been done at an interval of a year or more. Each separation provided a set of T_1 and T_2 values and the activities of ^{228}Th and ^{232}Th at T for Eqn (1). Normalizing the ^{228}Th activity of the second separation to the ^{232}Th activity of the first then provided two equations that could be solved simultaneously for R_2 , i.e., the ratio of ^{228}Th to ^{228}Ra at the death of the subject. Aliquots of lung samples from subject L1521 that were analyzed 5 y apart provided four values of R_2 (0.78 ± 0.15 , 0.96 ± 0.19 , 0.91 ± 0.36 , and 0.94 ± 0.38) that had a weighted mean and SE of 0.86 ± 0.11 , and aliquots of pulmonary lymph node analyzed 7 y apart provided values of 1.22 ± 0.19 and

1.14 $\pm$ 0.18 for R_2 in lymph nodes of the same subject (weighted mean of 1.18 $\pm$ 0.13). Other sample solutions could not be reanalyzed, because the entire solutions had been used for the initial analyses. However, if one makes the reasonable assumption that all samples of a given type of tissue from an individual had the same ratio of ^{228}Th to ^{228}Ra at death, it is possible to obtain estimates of the ratio by pairing samples instead of aliquots. Unfortunately, large relative errors were propagated into the calculated values of R_2 for most of the paired samples, because the calculation involved small differences between relatively large quantities. Calculations for all combinations of samples with at least 1 y between separations yielded only the following estimates of R_2 with fractional SEs less than or equal to 0.5: two values from bone samples of L1521 with a weighted mean of 0.51 $\pm$ 0.51, 15 values from lung samples of L2107 with a weighted mean of 1.05 $\pm$ 0.19, and one value (1.46 $\pm$ 0.74) from lymph-node samples of L2107.

SUMMARY AND DISCUSSION

A summary of the ^{232}Th measurements is set out in Table 3 as the geometric means of the concentrations, the standard errors of the geometric means (GSEs), and the number of samples analyzed for each subject and type of tissue. The geometric means are shown because the distribution of Th within some organs is probably non-uniform. (In particular, Table 2 indicates that the Th concentrations in the lower portions of the lungs were higher than in the upper portions [subjects L1521 and L2107] and that the concentrations in vertebrae were higher than in other samples of bone [subjects L0770, L1521, and L2932].) The last two sets of values, median concentrations for two general populations (Ibrahim et al. 1983), have been added to Table 3 to illustrate that the levels of Th in the former thorium workers are orders of magnitude greater than the levels in persons not occupationally exposed to Th. Comparison with Table 1 shows that the concentrations of ^{232}Th in the lungs of the thorium workers are strongly related to the lengths of employment and work assignments of the subjects, but the concentrations in bone and other internal organs show less of a correlation. An analysis of the metabolic behavior of

inhaled thorium that is implied by these findings and estimates of doses to the workers from internal radiation are given in a companion paper (Stehney 1991).

Table 4 contains summaries of the activity ratios of ^{228}Th and ^{230}Th to ^{232}Th . The weighted means of the samples of each organ are shown instead of the geometric means because it was assumed that the ratios (but not the concentrations) were the same in all parts of the organ. Except for the lymph node of subject L0770, the ratios of ^{228}Th to ^{232}Th range from 0.24 to 0.47. These ratios obviously were affected by differences in the metabolic behavior of ^{232}Th and ^{228}Ra and by time factors related to the exposure histories of the individual subjects. At low levels of Th from occupational exposure, uptake of environmental ^{228}Ra from the GI tract might greatly increase the ratio of ^{228}Th to ^{232}Th in the internal organs (Wrenn et al. 1981). The activity ratios of ^{230}Th to ^{232}Th are mostly 0.1-0.2. Here, the isotopic composition of the aerosols to which a worker was exposed should have been the principal determinant of the ratio for that individual. Monazite sands imported into the thorium refinery at various times were said to contain about 6% by weight of ThO_2 , 45% rare-earth oxides, and 0.1% uranium. The activity ratio of ^{230}Th to ^{232}Th in these sands should then be about 0.05 if the ^{230}Th is in radioactive equilibrium with its precursor, ^{238}U . However, the 0.1% U content that was reported for the monazite is probably only a nominal figure; Albert (1966) cites compositions of monazite sands from various countries that would have $^{230}\text{Th}/^{232}\text{Th}$ ratios as low as 0.08 and as high as 0.4.

The residues in the three samples of lung from subject L1521 had ashed weights of 2.3 mg, 3.7, and 2.0 mg, respectively. On the basis of these weights, the ^{232}Th activities in the residues were equivalent to 0.92 ± 0.03 , 0.50 ± 0.02 , and $2.7 \pm 0.2\%$ by weight of ThO_2 , so the residues were closer in Th content to monazite than to purified Th. It is likely that the residues were composed of monazite, environmental silica, and highly insoluble rare-earth compounds that the plant had to purify to less than 0.25% Th in order to avoid classification of such compounds as radioactive materials.

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TABLE CAPTIONS

Table 1. Case data for former thorium workers.

Table 2. Th isotopes in tissue samples (^{228}Th corrected to time of death).

Table 2 (continued).

Table 3. Concentrations of ^{232}Th in autopsy samples (mBq g^{-1}): geometric means and geometric standard errors (GSE) of the measured values in N samples of each type of tissue.^a

Table 4. Activity ratios of ^{228}Th and ^{230}Th to ^{232}Th corrected to time of death: weighted means and standard errors (SE) of the values determined in N samples of each type of tissue.

Subject	Sex	Job	Job dates at plant	Years on job	Year of death (age)	Cause of death
L0770	M	Millwright Electrician	1948 1959-1966	0.2 6.6	1981 (63)	Auto accident
L1521	M	Laborer Rare-Earth Prod'n	1950-1958 1958-1973	7.7 15.2	1983 (63)	Cancer of pancreas
L1768	M	Laborer	1956-1959	2.8	1978 (42)	Gunshot
L2107	M	Laborer Fireman Th Production	1938-1942 1945-1947 1954-1973	3.0 1.9 18.9	1979 (58)	Cancer of esophagus
L2932	M	Laborer Truck Driver	1941 1943-1945	0.6 2.5	1976 (72)	Heart failure

Case	Sample ^a	Wet weight (g)	²³² Th		²²⁸ Th/ ²³² Th		²³⁰ Th/ ²³² Th			
			(mBq g ⁻¹ ± SE ^b)		(Ratio ± SE)		(Ratio ± SE)			
L0770	Lung ^c	93.0	1.92	± 0.06	0.40 ± 0.03		0.15 ± 0.02			
	Lung ^c	51.8	1.60	0.05	0.26 0.03					
	PLN ^{c,d}	2.39	5.61	± 0.26	0.85 ± 0.14					
	Vertebra ^c	25.1	1.19	± 0.03	0.15 ± 0.02		0.18 ± 0.01			
	Vertebra ^c	22.2	0.88	0.03						
	Rib ^c	23.4	0.60	0.04						
	Sternum ^c	34.8	0.39	0.02	0.19	0.03	0.18	0.01		
	Liver ^c	108.5	0.17	± 0.01						
	Spleen	85.9	0.97	± 0.03						
	Bronchus ^c	10.4	0.010	± 0.021						
L1521	Lung R Mid T ^c	12.0	49.9	± 3.3	0.31 ± 0.02		0.13 ± 0.01			
	Residue ^c	(12.0)	6.36	0.21	0.89 0.04					
	Lung R Mid M ^c	10.7	50.7	1.4	0.28 0.01					
	Residue ^c	(10.7)	6.12	0.22	0.90 0.03					
	Lung R Mid B ^c	12.9	79.2	2.1	0.26 0.01					
	Residue	(12.9)	15.1	1.0						
	PLN ^c	0.79	1210.	± 40.	0.43 ± 0.02					
	MLN ^e	1.12	0.48	0.09	1.06 0.62					
	MLN	0.89	2.09	0.19	0.48 0.22					
	Vertebra ^c	25.3	0.80	± 0.03	0.28 ± 0.02				0.19 ± 0.07	
	Vertebra	30.1	0.62	0.03	0.32 0.02					
	Rib ^c	21.7	0.59	0.02	0.44 0.04					
	Rib	18.1	0.38	0.02	0.45 0.04				0.19 0.01	
	Liver	29.3	0.13	± 0.01	0.30 ± 0.06					
	Liver	33.8	0.17	0.01	0.41 0.06					
	Spleen	32.5	1.13	± 0.05						
	Spleen	31.3	1.02	0.05						
L1768	Spleen ^c	11.1	1.13	± 0.14	0.45 ± 0.10		0.16 ± 0.07			
	Residue	(11.1)	0.082	0.045						

Case	Sample ^a	Wet weight (g)	²³² Th		²²⁸ Th/ ²³² Th		²³⁰ Th/ ²³² Th	
			(mBq g ⁻¹ ± SE ^b)		(Ratio ± SE)		(Ratio ± SE)	
L2107	Lung L Inf	8.35	13.1	± 0.6	0.26	± 0.03	0.13	± 0.01
	Lung L Inf	7.88	16.9	0.8	0.25	0.03	0.12	0.01
	Lung L Inf	8.45	30.0	1.2	0.26	0.03	0.12	0.01
	Lung L Inf	11.35	21.9	0.8	0.28	0.03	0.12	0.01
	Lung ^c	7.07	29.4	1.7	0.17	0.03	0.14	0.01
	Lung R Sup ^c	22.6	4.77	0.16	0.26	0.04	0.16	0.02
	Lung R Sup ^c	8.77	4.56	0.60	0.31	0.09	0.13	0.06
	Lung R Sup ^c	17.3	4.97	0.51	0.26	0.04	0.14	0.01
	Lung R Mid B ^c	13.8	11.8	0.4	0.26	0.04	0.13	0.01
	PLN	16.7	61.5	± 2.3	0.31	± 0.04	0.12	± 0.01
	PLN ^c	11.5	15.4	11.6	0.25	0.05	0.13	0.01
	Liver	15.6	0.68	± 0.03			0.12	± 0.01
	Spleen	8.57	5.80	± 0.23			0.12	± 0.01
	Spleen	7.48	4.75	0.20			0.12	0.01
	Kidney	31.1	0.068	± 0.004			0.14	± 0.02
L2932	Lung	34.8	0.35	± 0.02	0.53	± 0.08	0.16	± 0.01
	Lung ^c	31.7	0.17	0.02	0.25	0.16	0.31	0.05
	PLN	1.88	3.86	± 0.20			0.13	± 0.02
	Vertebra	14.2	0.17	± 0.01			0.17	± 0.03
	Femur	23.0	0.14	0.02			0.11	0.05
	Liver	27.1	0.015	± 0.003			0.16	± 0.10
	Kidney	37.6	0.009	± 0.002			0.39	± 0.15

^aIf known, the sites of the lung samples are identified as follows: L = left; R = right; Sup = superior lobe; Mid = middle lobe; Inf = inferior lobe; T = upper portion of lobe; M = middle portion; B = lower portion.

^bSE = standard error.

^cReplicate analyses.

^dPLN = pulmonary lymph nodes.

^eMLN = mesenteric lymph nodes.

Subject	Lung	PLN ^b	Bone	Liver	Spleen	Kidney
L0770	1.75	5.40	0.70	0.17	0.97	na ^c
GSE (N)	1.10 (2)	(0.26)	1.27 (4)	(0.01)	(0.03)	
L1521	67.1	1210.	0.58	0.15	1.08	na
GSE (N)	1.19 (3)	(40.)	1.17 (4)	1.14 (2)	1.06 (2)	
L1768	na	na	na	na	1.21	na
GSE (N)					(0.15)	
L2107	12.1	30.7	na	0.68	5.25	0.068
GSE (N)	1.29 (9)	2.00 (2)		(0.03)	1.10 (2)	(0.004)
L2932	0.24	3.86	0.16	0.015	na	0.009
GSE (N)	1.45 (2)	(0.20)	1.07 (2)	(0.003)		(0.002)
NORMAL1 ^d	0.012	0.10	0.0037	0.0019	na	0.0011
NORMAL2 ^d	0.022	0.29	0.0059	0.0011	0.0033	0.0026

^aIf only one sample was measured, the standard error (SE) of the measurement is shown in parentheses below the measured value.

^bPLN = pulmonary lymph nodes. ^cna = not available.

^dMedian concentrations reported by Ibrahim et al. (1983) for two populations not occupationally exposed to Th.

Subject	Lung	PLN ^a	Bone	Liver	Spleen	Kidney
<u>²²⁸Th/²³²Th</u>						
L0770	0.34	0.85	0.18	na ^b	na	na
SE (N)	0.07 (2)	0.14 (1)	0.02 (3)			
L1521	0.35	0.43	0.34	0.36	na	na
SE (N)	0.01 (2)	0.02 (1)	0.05 (4)	0.06 (2)		
L1768	na	na	na	na	0.45	na
SE (N)					0.10 (1)	
L2107	0.24	0.29	na	na	na	na
SE (N)	0.01 (9)	0.03 (2)				
L2932	0.47	na	na	na	na	na
SE (N)	0.13 (2)					
<u>²³⁰Th/²³²Th</u>						
L0770	0.15	na	0.18	na	na	na
SE (N)	0.02 (1)		0.01 (3)			
L1521	na	0.13	0.19	na	na	na
SE (N)		0.01 (1)	0.01 (2)			
L1768	na	na	na	na	0.16	na
SE (N)					0.07 (1)	
L2107	0.13	0.13	na	0.12	0.12	0.14
SE (N)	0.01 (9)	0.01 (2)		0.01 (1)	0.01 (2)	0.02 (1)
L2932	0.17	0.13	0.17	0.16	na	0.39
SE (N)	0.10 (2)	0.02 (1)	0.03 (1)	0.10 (1)		0.15 (1)

^aPLN = pulmonary lymph nodes.

^bna = not available.

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