Experience with Delayed- and Prompt-Gamma Neutron Activation Analysis using Accelerator-based neutrons at KFUPM: An overview

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Abstract – *Neutron Activation Analysis (NAA) is one of the major nuclear applications using neutrons. These neutrons can originate from a variety of sources, such as nuclear reactors, isotopic neutron sources, or neutron generators. The nuclear group at KFUPM has gained extensive experience in the use of fast and thermal neutrons for NAA on a variety of solid and liquid samples, with delayed and prompt gamma rays, over the past 30+ years, using both open-end and portable neutron generators. The measured concentrations ranged from a low of 6 ppm for vanadium to a high of 2.3 wt% for strontium using delayed gamma rays from activated soil samples; and from a low of 24 ppm for boron in water samples to a high of 15.8 wt% for oxygen in bulk samples for prompt gamma rays. The present paper briefly describes the salient features of the NAA setups and some of the measurements and results obtained using them. Brief information is provided while details can be found in the relevant references listed.*

Keywords: TNC, NIS, NAA, PGNAA, Neutron Generators

I. Introduction

Neutron Activation Analysis (NAA) with neutron generators is a well-established and widely used nuclear analytical technique **[1]**. It is a non-destructive, multi-elemental, and bulk analysis method that requires minimum sample preparation. The process of neutron activation can be achieved with a variety of neutron sources. The sources that achieve the best sensitivities for most elements are high-flux nuclear reactors. These sensitivities can reach down to lower than ppb levels. This is due to the availability of very high thermal neutron fluxes inside the nuclear reactor for use in Instrumental Neutron Activation Analysis (INAA) with delayed gamma rays. Moreover, even the extracted neutron beams have very high fluxes for use in Prompt Gamma Neutron Activation Analysis (PGNAA), albeit many orders of magnitude lower than those inside the nuclear reactor.

The neutrons that are produced at the KFUPM 350 keV ion accelerator **[2]** are fast neutrons with approximately 2.5 MeV and 14 MeV energies. These neutrons were thermalized using a thick moderator as illustrated in **Fig. 1**. Commercial, portable neutron generators at KFUPM produce neutrons of 2.5 MeV energy **[3]**, while a 16 Ci Am-Be isotopic ion source produces neutrons in a wide energy range, with an average of about 4.2 MeV **[4]**. The experience gained in the many aspects of nuclear instrumentation including optimization as well as data acquisition and analysis using neutron generators is directly relevant to the case when the neutrons originate from nuclear reactors instead.

During the process of neutron irradiation of a target nucleus of mass number A and atomic number Z, $X(A,Z)$, the following processes are likely to take place **[5,6]**:

1. For fast neutrons used in Neutron Inelastic Scattering (NIS), the following process takes place

$$n + \mathrm{X}(A,Z) \rightarrow n' + \mathrm{X}(A,Z)^*$$
$$\downarrow \mathrm{X}(A,Z) + \gamma_1$$

where n' is a neutron of lower energy than that of the incident neutron n, $\mathrm{X}(A,Z)^*$ is an excited state of the target nucleus, and γ_1 are prompt gamma rays emitted during the transition of $\mathrm{X}(A,Z)^*$ to its ground state $\mathrm{X}(A,Z)$.

2. For thermalized neutrons used in Thermal Neutron Capture (TNC) reactions, the following two process may take place:

a. $$n + \mathrm{X}(A,Z) \rightarrow \mathrm{Y}(A+1,Z)^*$$
$$\downarrow \mathrm{Y}(A+1,Z) + \gamma_2$$

where $\mathrm{Y}(A+1,Z)^*$ is an excited state of the compound nucleus, and γ_2 are prompt gamma rays emitted during the transition of $\mathrm{Y}(A+1,Z)^*$ to its ground state $\mathrm{Y}(A+1,Z)$. If the ground state of the compound nucleus is stable, the process ends here.

b. However, if the ground state of the compound nucleus is unstable, a beta decay occurs with a specific half-life $T_{1/2}$:

$$\mathrm{Y}(A+1,Z) \rightarrow \mathrm{C}(A+1,Z+1)^* + e^- + \bar{\nu}_e$$
$$\downarrow \mathrm{C}(A+1,Z+1) + \gamma_d$$

where $\mathrm{C}(A+1,Z+1)^*$ is the daughter nucleus in an excited state, e^- is an electron, $\bar{\nu}_e$ is an anti-neutrino of the electron type, and γ_d are called delayed gamma rays. The beta decay process happens through weak interactions, and the associated half-lives span a very wide range, from seconds to years. The number of measured delayed gamma rays is governed by the half-life $T_{1/2}$ of the parent nucleus $\mathrm{Y}(A+1,Z)$.

The prompt and delayed gamma rays are characteristic of the activated elements and are used to uniquely identify them, while their intensities are used to estimate the elemental concentrations in the activated samples.

II. Experimental Setups

II.A. Delayed-Gamma NAA (NAA)

II.A. 1. Irradiation Station

The Delayed-Gamma NAA Facility at KFUPM uses neutrons from the 350-keV ion accelerator or neutrons from the 16 Ci Am-Be isotopic source. Fast, monoenergetic neutrons are produced with about 2.5 MeV and 14 MeV energies in the D(d,n)^{3}He and T(d,n)^{4}He reactions, respectively. The neutrons from the Am-Be source are produced when an alpha particle emitted by the 241-Am radioisotope reacts with a ^{9}Be nucleus to produce a neutron and a ^{12}C nucleus in an excited state. The nominal output of such sources is 2.2 x 10^6 n/s for each 1 Ci of activity. Its half-life is 432 years and the average neutron energy is 4.2 MeV with a maximum of about 11 MeV **[4]**.

II.A.2. Thermalization of fast neutrons

Fast neutrons from the 350-keV ion accelerator first pass through a 1 cm-thick water layer, then through a 0.5 cm-thick stainless-steel water jacket casing, and are finally slowed down in a cylindrical moderator. The maximum thermal-neutron flux achieved with the D(d,n)^{3}He reaction was 2.5 x 10^6 n/cm^2-s and that achieved with the T(d,n)^{4}He reaction was 3 x 10^7 n/cm^2-s **[7]**. A drawing of a typical setup for Neutron Activation using Thermal Neutron Capture (TNC) with neutrons from the 350 keV ion accelerator. On the other hand, a paraffin sphere is used to moderate fast neutrons from the Am-Be source. The maximum thermal neutron flux achieved with the Am-Be neutron source was 2.7 x 10^4 n/cm^2-s **[4]**.

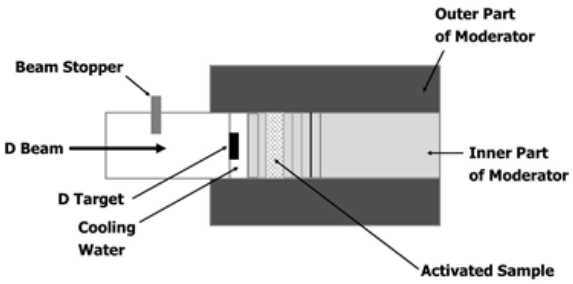

Fig. 1. Drawing of typical setup for Neutron Activation with delayed gamma rays in Thermal Neutron Capture using the 350 keV ion accelerator

II.A.3. Counting station

The counting station was housed in a separate room that is about 60 m away from the irradiation

station. It was based on a heavily shielded HPGe detector with its associated electronics and a PC-based data acquisition and analysis system with 4096 channels. The resolution of the detector is about 2 keV at the 1.33 MeV line of ^{60}Co. The system is calibrated using standard γ-ray sources such as ^{22}Na, ^{137}Cs, and ^{60}Co. The data acquisition and analysis were carried out using the MAESTRO software package provided by EG&G Ortec. More details on the counting station can be found in **[4]**.

II.A.4. Applications

Vanadium determination in phosphate samples

Economically viable phosphate deposits have been found in northwestern Saudi Arabia **[8,9]**. Phosphate deposits usually contain important trace elements such as vanadium and rare earth elements **[10]**. Such elements can be exploited, with existing technology, as by-products from the commercially viable phosphate deposits, and can help in the interpretation of the genesis and diagenesis of these phosphate deposits **[11]**. The accelerator-based thermal neutrons at the KFUPM NAA facility were used to analyze ten phosphorite samples collected from the phosphate deposits in northwestern Saudi Arabia. The nuclear data of vanadium relevant to the present analysis are listed in **Table 1**, along with the data of other studied elements, all of which were measured using delayed gamma rays.

Table 1. Nuclear data of elements determined in accelerator-based thermal-neutron activation analysis.

Target Isotope	Abund. [%]	σ_{th} [barns]	Product Isotope ($T_{1/2}$)	E_γ [keV]	Intensity [%]
^{23}Na	100	0.513	^{24}Na (15.0 h)	1369	100
^{26}Mg	11.0	0.037	^{27}Mg (9.46 m)	1014	28.6
^{27}Al	100	0.226	^{28}Al (2.24 m)	1779	100
^{37}Cl	24.2	0.423	^{38}Cl (37.2 m)	1643	31.0
^{41}K	6.73	1.45	^{42}K (12.4 h)	1525	18.8
^{51}V	99.8	4.79	^{52}V (3.75 m)	1434	100
^{55}Mn	100	13.2	^{56}Mn (2.58 h)	847	98.9
^{86}Sr	9.86	0.770	^{87}Sr (2.81 h)	388	82.3
^{138}Ba	71.7	0.405	^{139}Ba (84.6 m)	166	22.1
^{164}Dy	28.2	2730	^{165}Dy (2.33 h)	94.7	3.58
^{185}Re	37.4	106	^{186}Re (90.6 h)	137	8.50
^{187}Re	62.6	73.2	^{188}Re (17.0 h) ^{188m}Re (18.6 m)	155 106	14.9 10.8

The results of the analysis are shown in **Table 2**. The errors were mainly due to counting statistics. The determination of vanadium concentration in the various phosphate samples was taken as the average of three independent measurements **[12]**.

Table 2. Vanadium concentrations in the phosphate samples as determined by accelerator-based thermal-neutron activation analysis.

Sample No.	Mass [g]	Vanadium [ppm]
1	21.8	457 ± 13
2	56.7	295 ± 6
3	89.6	197 ± 4
4	91.1	87 ± 3
5	109	69 ± 3
6	93.4	29 ± 2
7	42.1	28 ± 2
8	39.4	18 ± 2
9	8.69	77 ± 16
10	109	23 ± 1

Determination of rhenium in Pt-Re/Alumina reforming catalysts

In addition to platinum, rhenium is commonly used in reforming catalysts in the petroleum refining industry because of its high activity, selectivity, and long lifetime. Both calcined and uncalcined Pt-Re/Al catalyst samples should be analyzed and characterized using multiple analytical techniques to understand their physical characteristics. The accelerator-based thermal neutron flux at the KFUPM NAA facility was used to irradiate several calcined and uncalcined Pt-Re/Al catalyst samples of (0.5% Pt + 0.5% Re)/Al, 0.2% and 0.8% Re/Al. **Table 1** shows the relevant nuclear data for the determination of Re. The minimum detection limit for Re was 35 μg **[13]**.

Elemental analysis of local soil samples

Due to the importance of soil composition in agriculture, it was deemed useful to carry out an elemental analysis of a selected number of local agricultural soil samples in order to assess the capability of the KFUPM NAA facility for the elemental analysis of such samples. The soil samples used in this investigation were collected from Al-Hofuf area in eastern Saudi Arabia. The elements

determined in the soil samples were: K, Mn, V, Al, Na, Cl, Ba, Sr, and Mg. **Table 1** lists the relevant nuclear data for the elements determined in this analysis. The minimum, maximum, average, and standard deviation of the absolute concentrations of the elements determined in the local agricultural soil samples are listed in **Table 3 [14]**.

Table 3. Minimum and standard deviation of absolute concentrations of the elements determined in soil samples using delayed gamma rays from accelerator-based thermal-neutron activation analysis.

The table is added below

II.B. Prompt-Gamma NAA (PGNAA)

The Prompt-Gamma Neutron Activation Analysis (PGNAA) setups at KFUPM use either 2.5 MeV or 14 MeV neutrons from the 350 keV ion accelerator, or 2.5 MeV neutrons from the portable neutron generators for Neutron Inelastic Scattering (NIS). They also use polyethylene moderators to thermalize 2.5 MeV neutrons from the portable neutron generators for TNC studies. At one time or another, the scintillation detectors used in PGNAA measurements consisted of Sodium Iodide (NaI), Bismuth Germanate (BGO), Lanthanum Tri-Bromide (LaBr3), Lanthanum Tri-Chloride (LaCl3), or Cerium Tri-Bromide (CeBr3) cylindrical detectors. In all experiments the scintillation detectors were shielded against direct neutrons and scattered gamma rays. Nonetheless, they underwent some radiation damage. A typical arrangement of the activation setup is shown in **Fig 2**. An essential ingredient in the use of PGNAA is an up-to-date and accurate knowledge of the data tables of the multiple gamma ray lines of the various elements of interest **[15]**. Another important, if not essential, component for the efficient use of PGNAA are the Monte Carlo simulation codes that are used to optimize the setups, most notably MCNP **[16-19]**.

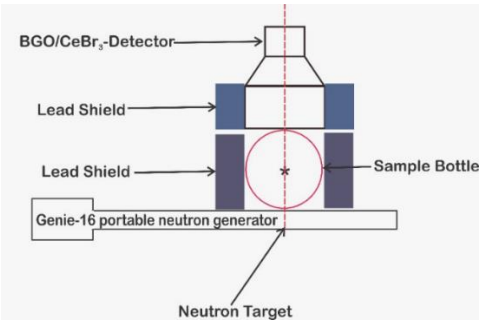

Fig. 2. Typical activation setup used in PGNAA using Neutron Inelastic Scattering with 2.5 MeV neutrons

II.B.1 Applications

PGNAA has been extensively used at KFUPM for the detection and measurement of a large number of elements, particularly for environmental applications. The technique is useful for the measurement of major concentrations of most elements, including few light elements such as carbon, and oxygen. **Table 4** lists the studied elements using TNC-NAA. **Table 5** lists the elements studied using NIS-NAA with 2.5 MeV neutrons, and **Table 6** lists the elements studied using 14 MeV NIS-NAA. The performance of the PGNAA setup is indicated by the Minimum Detectable Concentrations (MDC) and their uncertainties (σ_{MDC}) for the studied elements.

Table 4. Elements studied with PGNAA using TNC.

Element	sample	Energy [keV]	MDC ± σ_{MDC} [wt%]	Ref.
Ni	Soil	8553	0.79 ± 0.24	[20]
Ni	Soil	8998	1.27 ± 0.39	[20]
S	Soil	5420	1.3 ± 0.4	[23]
Cl	Water	517	0.08 ± 0.02	[24]
Cl	Water	786-788	0.07 ± 0.02	[24]
Cl	Water	1165	0.18 ± 0.05	[24]
Cl	Water	1951/1959	0.11 ± 0.03	[24]
Cl	Water	2863	0.17 ± 0.05	[24]
Cl	Water	6111	0.13 ± 0.04	[24]
Cl	Water	6619	0.18 ± 0.05	[24]
Cl	Water	8578	0.8 ± 0.24	[24]
B	Water	478	24.4 ± 7.43 ppm	[25]
Cd	Water	245-651	95.6 ± 29.1 ppm	[25]
Hg	Water	368	0.15 ± 0.05	[25]

Table 5. Elements studied with PGNAA using NIS of 2.5 MeV neutrons.

Element	sample	Energy [keV]	MDC ± σ_{MDC} [wt%]	Refe
Ni	Soil	1331	2.00 ± 0.70	[20]
Ni	Soil	1454	1.79 ± 0.55	[20]
Cr	Soil	1430	0.85 ± 0.26	[21]
Ti	Soil	984	0.68 ± 0.18	[21]
Zn	Soil	1005	1.53 ± 0.47	[21]
P	TSP	2230	0.55 ± 0.17	[22]
Na	TSP	438	0.79 ± 0.24	[22]
Na	NaCl	438	0.73 ± 0.22	[22]
P	TSP	2230	0.37 ± 0.11	[22]
Na	TSP	438	0.31 ± 0.09	[22]
Na	NaCl	438	0.35 ± 0.11	[22]
S	Mg-SO_4	2240	0.56 ± 0.17	[22]
S	Soil	2240	0.68 ± 0.21	[22]

Table 6. Elements studied with PGNAA using NIS of 14 MeV neutrons.

Element	sample	Energy [keV]	MDC [wt%]	σ_{MDC} [wt%]	Ref.
C	bulk	4439	12.2/3.2	3.8/1.0	[26]
O	bulk	6130	15.8/x	4.8/x	[26]

The first number for the MDC and σ_{MDC} corresponds to the $LaBr_3$ detector while the second number after the slash sign corresponds to the BGO detector. Notice that Oxygen cannot be analyzed using the BGO detector because of the large presence of Oxygen in the detector material.

III. Conclusions

The experience gained at KFUPM in NAA and PGNAA using TNC and NIS with 2.5 MeV and 14 MeV neutrons from the 350 keV ion accelerator and the portable neutron generators shows that, even though the available neutron fluxes are relatively low, they nonetheless can be advantageously used to carry out useful analyses of interest to academic research, various industries, as well as environmental studies, when coupled with scintillation detectors with high resolution and good efficiency. The analyses can be optimized by judicious use of simulation codes and data reduction software. The results can definitely be much improved by having higher neutron fluxes and using appropriate electronics to suppress the continuous Compton background in the scintillation detectors at low gamma energies. The valuable experience gained at KFUPM in NAA with both delayed and prompt gamma rays using neutron generators over the past 30+ years is very relevant to the applications program of nuclear reactors.

Acknowledgments

Work on accelerator-based NAA and PGNAA has benefitted from the support of KFUPM through internal university funding as well as from government support through NSTIP funding. This support as well as the support of the Physics Department at KFUPM are gratefully acknowledged.

NAA: Neutron Activation Analysis
TNC: Thermal Neutron Capture
NIS: Neutron Inelastic Scattering
PGNAA:Prompt Gamma Neutron Activation Analysis
TSP: Sodium triphosphate
MDC: Minimum Detectable Concentration
σ_{MDC}: Error on MDC
σ_{th}: Thermal Capture Cross Section.

References

1. Neutron Generators for Analytical Purposes, International Atomic Energy Agency, Vienna, (2012)

2. H. Al-Juwair, G. Blume, R. J. Jaarsma, C. R. Meitzler, and K. H. Purser, "350 keV accelerator facility for th University of Petroleum & Minerals", Nuclear Instruments and Methods in Physics Research, Vol. **B24/25**, pages 810-812 (1987).

3. Genie 16 neutron generator. Inside N. R C (2013) https://www.nrc.gov/docs/ML1408/ML14085A270.pdf

4. A. Aksoy, M. N. Al-Haddad, A. A. Naqvi, K. Al-Nahdi, "An ^{241}Am-Be source based thermal neutron activation analysis facility at KFUPM", Journal of Radioanalytical and Nuclear Chemistry, Articles, **Vol. 181**, No. 1, pages 131-140 (1994)

5. A. R. Zeisler, N. Vajda, G. Kennedy, G. Lamaze, G. L. Molna´r, Activation Analysis, Handbook of Nuclear Chemistry, 2nd Edition, Chapter 30, Page 1553 (2011)

6. Z. Re´vay, R. M. Lindstrom, E. A. Mackey, T. Belgya, Neutron-Induced Prompt Gamma Activation Analysis (PGAA), Handbook of Nuclear Chemistry, 2nd Edition, Chapter 31, Page 1619 (2011)

7. F. Z. Khiari, A. Aksoy, M. N. Al-Haddad, "Accelerator-based slow neutrons at KFUPM", Book of Abstracts for the Fourteenth International Conference on the Applications of Accelerators in research and Industry, Nov. 6-9 (1996) Denton Texas, USA.

8. Ministry of Petroleum & Minerals, Directorate General of Mineral Resources, Mineral resources of Saudi Arabia (1994)

9. Al-Majalla No. 793, pages 49-51 (1995)

10. P. J. O'Connor and J. F. Pyne, "Radioelement and REE content of Namurian Phosphorites, County Clare, Ireland", Trans. Instn. Min. Metall. (Sect. B. Appl. Earth Sci.), **Vol. 95**, pages B211-213, (1986)

11. S. M. McLennan and S. R. Taylor, "Rare Earth mobility associated with Uranium mineralization", Nature, **Vol. 282**, pages 247-249 (1979)

12. F. Z. Khiari, A. Aksoy, A. Shahin, "Analysis of phosphate rock samples for vanadium using accelerator-based thermal neutrons", Journal of Radioanalytical and Nuclear Chemistry, **Vol. 261**, Nos. 3, pages 507-511, (2004)

13. A. Aksoy, F. Z. Khiari, A. Rahman, "Study of Pt-Re/Alumina reforming catalysts by neutron activation analysis", Journal of Radioanalytical and Nuclear Chemistry, **Vol. 230**, Nos. 1-2, pages 75-78, (1998)

14. A. Aksoy, F. Z. Khiari, N. A. al-Shayea, "Analysis of soil samples from Saudi Arabia using neutron activation spectrometry", 8th Congress of International Association of Engineering Geology and the Environment, Vancouver, British Columbia, Canada, September 21-25, (1998)

15. Database of Prompt Gamma Rays from Slow Neutron Capture for Elemental Analysis, International Atomic Energy Agency, Vienna, (2007)

16. MCNP® USER'S MANUAL Code Version 6.2 October 27, (2017)

17. A. A. Naqvi, M. M. Nagadi, Khateeb-ur-Rehman, M. Maslehuddin, S, Kidwai, "Monte Carlo simulations for design of the KFUPM PGNAA facility", Radiation Physics and Chemistry vol. **66**, pages 89-98 (2003)

18. M. S. Abdelmonem, A. A. Naqvi, Ghada Al-Misned, Hasan Al-Ghamdi, "Performance improvement of a PGNAA setup due to change in moderator design: A Monte Carlo study", Journal of Radioanalytical and Nuclear Chemistry, vol. **271**, pages 685–690 (2007)

19. A. A. Naqvi, F. Z. Khiari, F. A. Liadi, Khateeb-ur-Rehman, M. A. Raashid, A. H. Isab, "Moisture effect in prompt gamma measurements from soil samples", Applied Radiation and Isotopes vol. **115**, pages 61–66 (2016)

20. G. AlMisned, T. Al-Abdullah, F. A. Liadi, A. Hawsawi, "Comparison between NIS and TNC channels for the detection of nickel in soil samples", Applied Radiation and Isotopes, vol. **186**, pages 1-5 (2022)

21. Ghada AlMisned, Tariq Al-Abdullah, Fatai Liadi, Adil Hawsawi, "Improving a PGNAA Technique to Detect Heavy Metals in Solid Samples", Applied Sciences, vol. **12**, pages 1-10, (2022)

22. Fatai A. Liadi, A. A. Naqvi, F. Z. Khiari, M. Raashid, "Phosphorus, sodium and sulphur measurements in compound samples using 2.5 MeV neutron activation", Journal of Radioanalytical and Nuclear Chemistry, vol. **331**, pages 1155–1161 (2022)

23. T. Al-Abdullah, A. A. Naqvi, F. Z. Khiari, M. Raashid, A. H. Isab , "Sulfur analysis in bulk samples using a DD portable neutron generator based PGNAA setup", Journal of Radioanalytical and Nuclear Chemistry, vol. **326**, pages 555–561 (2020)

24. A. A. Naqvi, F. Z. Khiari, F. A. Liadi, Khateeb ur-Rehman, M. Raashid, A. A. Isab, "Multiple Gamma-Ray Detection Capability of a $CeBr_3$ Detector for Gamma Spectroscopy", Journal of Spectroscopy, vol. **2017**, 8 pages (2017)

25. A. A. Naqvi, F. Z. Khiari, F. A. Liadi, Khateeb-ur-Rehman, A. A. Isab, "Performance tests of a large volume cerium tribromide ($CeBr_3$) scintillation detector", Applied Radiation and Isotopes, vol. **114,** pages 50-56 (2016)

26. A.A. Naqvi, Fares A. Al-Matouq, F. Z. Khiari, A. A. Isab, Khateeb-ur-Rehman, M. Raashid, "Prompt gamma tests of $LaBr_3$:Ce and BGO detectors for detection of hydrogen, carbon and oxygen in bulk samples", Nuclear Instruments and Methods in Physics Research A **684**, pages 82–87 (2012)

Element →	K [wt%]	Mn [ppm]	V [ppm]	Al [wt%]	Na [ppm]	Cl [ppm]	Ba [ppm]	Sr [wt%]	Mg [ppm]
Minimum	0.6	126	6.0	1.2	2490	352	349	0.4	3130
Maximum	1.1	165	26	2.1	3900	5090	534	2.3	6876
Average	0.8	151	11	1.6	3090	1490	436	1.5	5327
Std. Dev.	0.2	18.0	10	0.4	603	1640	63.0	0.7	1195

Table 3