ASSESSMENT OF URANIUM LEVELS IN ATMOSPHERIC PARTICULATE MATTER (PM$_{10}$) IN AND AROUND THE TUMMALAPALLE URANIUM MINING SITE, ANDHRA PRADESH, INDIA

Murad Basha A., Yasovardhan N., Satyanarayana S.V. *,1, Subba Reddy G.V. 2, Padma Savitri P., Vishwa Prasad K.3 and Vinod Kumar A.4

1. Department of Chemical Engineering, JNTUACE, Anantapur, Andhra Pradesh (INDIA)
2. Department of Chemistry, JNTUACE, Pulivendula, Andhra Pradesh (INDIA)
3. HPU Division, Nuclear Fuel Complex, Hyderabad (INDIA)
4. Environmental Assessment Division, BARC, Mumbai (INDIA)

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ABSTRACT

Recently, there is a rapid increase of nuclear based generated power, associated industries and its mining activities in India. This has raised the questions concerning a possible increase of uranium levels over that existing in the environment. The present work explores the evaluation of ambient air quality with respect to the Atmospheric Particulate Matter (PM$_{10}$) and Uranium concentration in PM$_{10}$ at Tummalapalle Uranium mining site as well as its surrounding villages. In order to monitor ambient air quality, ten PM$_{10}$ Air Samplers (10 locations) were installed in and around 30 km radial distance from Uranium mining site. Air Samples were collected twice in a month for one year duration. The uranium concentration in PM$_{10}$ was estimated using UV-Flourimetry technique after acid digestion of PM$_{10}$ present on the glass fiber/EPM 2000 filter paper. The Uranium concentration was found to be in the range of 5 - 30 ng/m$^3$. These values are found to be normal compared with the reported literature values which are in the range of 20 - 200 ng/m$^3$.

Key Words: Nuclear power, Uranium, PM$_{10}$, UV–Flourimetry, Air quality

INTRODUCTION

In present days, the world uses a great deal of energy and the usage of it is increasing dramatically, particularly in developing countries. Uranium is a best alternative source in the production of electricity through nuclear technology to meet the world’s demand. In view of importance of uranium in the generation of electricity with the help of nuclear technology, Government of India (UCIL-Uranium Corporation of India Limited) has established the extraction and processing of Uranium from its ore at Tummalapalle, Pulivendula, Andhra Pradesh, India. Uranium (U) is naturally occurring heavy metal and is found at an average concentration of 0.0003% (3 mg/kg) in the earth’s crust. It occurs in numerous minerals and is also found in lignite, monazite sands, phosphate rock and phosphate fertilizers.$^{1,2}$ Since, uranium is naturally occurring element, it is present in the environment in ultra trace levels, interacts with its surroundings and may reach the humans through inhalation, food and drinking water.$^3$ There is always a chance of release of Uranium into the atmosphere in various stages in the nuclear fuel cycle, including mining, milling, reining, fuel fabrication and fuel reprocessing. Basically uranium is both chemically and radio logically toxic. The literature survey reveals that uranium toxicity primarily affects the kidney, causing damage to the proximal tubule. Further, this metal has been also identified as a potential reproductive toxicant.$^{4,5}$ Particular Matter in air (both Air - PM$_{10}$ and PM$>10$ micron) have been recognized with great environmental significance. Particulate matter is the carrier of many harmful trace
metals (such as As, Be, Cd, Cr, Cu, Hg, Mn, Ni, Pb, Sb, Se, V, Zn, U etc), and can be deposited into the human respiratory system. The most essential step of figuring out the distribution characteristics of PM\textsubscript{10} in the industrial complex area is to establish long-term concentration profiles of PM\textsubscript{10} and the associated components including toxic metals. The workers and residents in and around industrial complexes are assumed to be exposed to elevated levels of the toxic metals associated with the PM\textsubscript{10} compared to residents who are living further away from the industrial areas.

**AIMS AND OBJECTIVES**

To analyze the Uranium concentration in ambient air associated with PM\textsubscript{10} and exclusively focused on quantitative estimation and variation of uranium in PM\textsubscript{10} at Tummalapalle Uranium mining site and its surrounding villages. The study area Tummalapalle, Andhra Pradesh, India lies and confined in the tropical region where the climate is characterised by very hot summers, mild winters and monsoon rains. In summer season, a temperature ranges from maximum 46°C during daytime to a minimum of 15°C at night. In winter maximum temperature during day goes up to 40.6°C and minimum temperature at night becomes as low 10.6°C.

**MATERIAL AND METHODS**

All reagent used for the analysis were of analytical grade and Milli – Q water is used for distilled water prior to use. The sampling was all dilutions. All the glassware was cleaned by soaking in dilute nitric acid and was rinsed with performed around the 30 km radial distance from the Uranium mining site. The sampling locations were fixed on the basis of wind roses and locations were distributed according to the radial distances from the site as follows:

- 5 locations within 5 km radial distance, considered as Core Zone
- 3 locations between 5-10 km radial distance, considered as Buffer Zone 1
- 2 locations between 10-30 km, considered as Buffer Zone 2

The sampling locations and its longitude and latitudes were measured through GPS locator and the values are given in Table 1. The location map is shown in Fig.1. The samples of PM\textsubscript{10} were collected twice in a month from November - 2010 to February - 2012. The Envirotech High Volume air sampler Model APM 460 BL was used to collect the samples by running the equipment for a period of 24 hours with an average flow rate of 1.3 LPM (lit/min). The samples of the particulate matter with aerodynamic diameter less than 10 μm (PM\textsubscript{10}) were collected by passing air over pre-weighed glass fiber/EPM 2000 filter paper.

<table>
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<tr>
<th>S/N</th>
<th>Location</th>
<th>Zones</th>
<th>Radial distance from the site (km)</th>
<th>Latitude/ Longitude</th>
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<td>2</td>
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<td></td>
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<td>14°18'24.12&quot;/78°17'46.38&quot;</td>
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<td>4</td>
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<td></td>
<td>3.93</td>
<td>14°19'50.64&quot;/78°18'05.94&quot;</td>
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<td>4.55</td>
<td>14°21'52.14&quot;/78°16'11.76&quot;</td>
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<td>Buffer Zone-1</td>
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<td>14°22'56.58&quot;/78°15'11.94&quot;</td>
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<td>8</td>
<td>L\textsubscript{8}</td>
<td></td>
<td>9.90</td>
<td>14°22'10.44&quot;/78°21'29.46&quot;</td>
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<td>9</td>
<td>L\textsubscript{9}</td>
<td>Buffer Zone-2</td>
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<td>14°26'55.74&quot;/78°14'04.50&quot;</td>
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<td></td>
<td>17.70</td>
<td>14°10'49.68&quot;/78°11'30.06&quot;</td>
</tr>
</tbody>
</table>
Fig. 1: Geographical distribution of the sampling locations

The wind rose for Tummalapalle uranium mining site collected for the period September – 2011 to June – 2012 is shown in Fig. 2. The filter papers used for the collection of PM$_{10}$ were pre-conditioned (24 h in a dessicator under the following conditions: temperature of 15 – 30 °C and humidity less than 40%). They were pre-weighed and after the particulates collected, the filters were reconditioned for another 24 h in the dessicator and reweighed for mass of PM$_{10}$. All the procedures during handling of filters were strictly quality controlled to avoid any possible contamination and followed the guidelines of CPCB, India.$^{17,18}$

Fig. 2: Wind rose for Tummalapalle uranium mining site

Uranium analysis

The PM$_{10}$ sample was collected on glass fiber/EPM 2000 filter paper which was digested in several stages. Initially, the digestion was carried out by taking approximately 20 ml of aquaregia (HNO$_3$: HCl - 1:3) until dryness on hotplate. Then, around 20 ml of HClO$_4$: HNO$_3$ (1:4) was added and digested on hotplate till complete dryness of the sample. Finally, the resultant sample was digested 2-3 times with Conc. HNO$_3$ on the hot plate until to dryness. The digest was rinsed with 4N HNO$_3$ and filtered off on Whatmann 41 filter paper. The filtrate was kept on the hot plate up to dryness. To the final residue, 10 ml of 4N HNO$_3$ and 10 ml of aluminum nitrate were added. From this mixture, the uranium is extracted using ethyl acetate by solvent extraction process. The separated organic layer is subjected to evaporation. The residue
 RESULTS AND DISCUSSION

PM$_{10}$ (µg/m$^3$) concentration and Uranium (ng/m$^3$) concentration in PM$_{10}$ sample at different sampling villages with its sampling dates, annual Minimum (Min), Geometric Mean (GM) and Maximum (Max) values are given in Table 2.

Air – PM$_{10}$ concentration

The PM$_{10}$ concentrations were observed in the range of 30.2 – 87.1 µg/m$^3$ at L$_1$; 22.9 – 109.6 µg/m$^3$ at L$_2$; 13.5 – 64.7 µg/m$^3$ at L$_3$; 24.3 – 78.1 µg/m$^3$ at L$_4$; 19.2 – 99.0 µg/m$^3$ at L$_5$; 15.4 – 134.4 µg/m$^3$ at L$_6$; 24.7 – 82.5 µg/m$^3$ at L$_7$; 15.2 – 67.6 µg/m$^3$ at L$_8$; 22.5 – 75.1 µg/m$^3$ at L$_9$; and 4.5 – 13.5 µg/m$^3$ at L$_{10}$. It has been observed that L$_6$ is showing high Air - PM$_{10}$ concentration (134.44 µg/m$^3$) with GM value of 45.8 µg/m$^3$, which was sampled in the season of winter. This concentration is higher than the maximum concentration reported in all other sampling locations. The observed Air – PM$_{10}$ value has exceeded once during the observation period, the limit of daily (24 hrs) - Indian National Ambient air quality standards of Air - PM$_{10}$ (100 µg/m$^3$) value.$^{19}$ This site is at a radial distance of 6.64 km from the mining site and the mining operation cannot be the cause to these high values. It was found that there exists an operating stone crushing mill near to the L$_6$ sampling location. The location is also situated beside Pulivendula to Kadapa highway, where huge vehicular movement takes place. The minimum Air - PM$_{10}$ (13.5 µg/m$^3$) concentration has been observed at L$_3$ with GM value of 37.5 µg/m$^3$. The reported value is low even comparing with minimum values at all other sampling locations where it was sampled in the season of monsoon. This sampling site is located in core zone at a radial distance of 3.74 km from the mining site. The sampling locations L$_1$, L$_2$, L$_6$ and L$_7$ were comparatively showed higher concentrations of PM$_{10}$ (GM of 52.9, 52.0, 45.8 and 52.2 respectively). The locations L$_1$ and L$_2$ are located in Core Zone and having the radial distance of 0.53 and 1.17 km, respectively. Dust generation due to movement of tipper lorries carrying ore and other vehicles for local public transportation may be the reason for the elevated values. The locations L$_6$ and L$_7$ are located in Buffer Zone – 1 at radial distance of 6.64 and 7.85 km, respectively. These sampling locations are located beside Pulivendula to Kadapa highway, catering to huge vehicular movement. In addition both the sampling locations there exists stone crushing mills. The sampling location L$_{10}$ frequently observed low concentration of PM$_{10}$ with the GM of 28.1. The location is located in Buffer Zone – 2 having the radial distance of 17.7 km from the mining site. This location is far away from the mining site, and also situated at other side of hill area and can be considered as a control pint for future studies as impact of transport of particulate matter including dust from the mining site, if any will be minimal. The high values of PM$_{10}$ concentration were observed in winter season (i.e., from December to February) in all the sampling locations due to enhanced combustion activities of the villager. In addition lot of agricultural crops are cultivated in winter season, adding to possible dust load generation.

Uranium concentration in air – PM$_{10}$

Uranium concentration in PM$_{10}$ were observed in the range of 3.8 – 32.2 ng/m$^3$ at L$_1$; 1.6 – 27.8 ng/m$^3$ at L$_2$; 1.8 – 22.9 ng/m$^3$ at L$_3$; 3.9 – 31.6 ng/m$^3$ at L$_4$; 1.5 – 30.6 ng/m$^3$ at L$_5$; 2.2 – 31.7 ng/m$^3$ at L$_6$; 3.1 – 28.6 ng/m$^3$ at L$_7$; 1.2 – 31.9 ng/m$^3$ at L$_8$; 1.5 – 24.9 ng/m$^3$ at L$_9$; and 4.5 – 13.5 ng/m$^3$ at L$_{10}$. By considering samples individually, a maximum concentration value (32.2 ng/m$^3$) was found in a sampling location L$_1$ with a GM of 13.5 ng/m$^3$ and these values marginally varied throughout the sampling period when compared with the other sampling location. This location is situated in core zone with the radial distance of 0.53 km from the mining site. Since, the location is very near to the mining site, transportation of ore from one place to other for processing and re-suspension of the deposited dust due to vehicular movement for transportation of public may be the possible reason for these elevated values. The GM’s of the all locations is below 11 ng/m$^3$ except the location L$_1$, where it is 13.5 ng/m$^3$. However, the concentration of Uranium reported in
Table 2: Air – PM$_{10}$ (µg/m$^3$) concentration and Uranium (ng/m$^3$) concentration in Air – PM$_{10}$ of different sampling villages with its sampling dates

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<th>Date of sampling</th>
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<th>L$<em>1$ PM$</em>{10}$ U</th>
<th>L$<em>2$ PM$</em>{10}$ U</th>
<th>L$<em>3$ PM$</em>{10}$ U</th>
<th>L$<em>4$ PM$</em>{10}$ U</th>
<th>L$<em>5$ PM$</em>{10}$ U</th>
<th>L$<em>6$ PM$</em>{10}$ U</th>
<th>L$<em>7$ PM$</em>{10}$ U</th>
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</tbody>
</table>

Note: U – Uranium, NS – Not Sample
all the locations were found to be very low when compared with the standard value 150 µg/m³ (regulatory standard for Ontario) based on the toxicity associated with this Uranium and Uranium related compounds for 24 hr average in Air – PM$_{10}$.

Further, as per National Emissions Standard for Hazardous Air Pollutants Radionuclide (NESHAPs) for U-238 is 350 ng/m$^3$ (Fig. 3).

The PM$_{10}$ and Uranium concentration of location 4, L$_{2}$, L$_{6}$ and L$_{7}$ were not showing any significant correlation, throughout the sampling duration from Nov.2010 to Feb.2012. Hence, both Air- PM$_{10}$ and Uranium concentrations are independent with each other and the graphical representation is shown in Fig. 4(a) to Fig. 4(d).

![Graph showing PM$_{10}$ and Uranium concentration in Air – PM$_{10}$](image)

**Fig. 3:** Annual GM of Air – PM$_{10}$ and Uranium concentration in Air – PM$_{10}$

**Daily intake**

The daily intake of Uranium concentration through inhalation is calculated based on 20 m$^3$ of air per day is inhaled by an adult of study area. The daily intake of Uranium in respective locations is tabulated in Table 3. On examining the content in Table 3, the daily intake of Uranium concentration has been observed 0.27 µg/day for L$_{1}$, 0.13 µg/day for L$_{2}$, 0.17 µg/day for L$_{3}$, 0.22 µg/day for L$_{4}$, 0.19 µg/day for L$_{5}$, 0.22 µg/day for L$_{6}$, 0.20 µg/day for L$_{7}$, 0.23 µg/day for L$_{8}$, 0.17 µg/day for L$_{9}$ and 0.23 µg/day for L$_{10}$. The calculated daily intake of uranium is below the standard permissible daily intake limit of uranium concentration (1.5 µg/day).  

![Graph showing PM$_{10}$ and Uranium concentration in Air – PM$_{10}$ for UCIL-Mining site](image)

**Fig. 4(a):** PM$_{10}$ and Uranium concentration in PM$_{10}$ for UCIL-Mining site
Fig. 4(b) : PM$_{10}$ and Uranium concentration in PM$_{10}$ for Tummalapalle

Fig. 4(c) : PM$_{10}$ and Uranium concentration in PM$_{10}$ for Bestuvaripalle

Fig. 4(d) : PM$_{10}$ and Uranium concentration in PM$_{10}$ for Vemula

Table 3 : Daily intake of uranium concentration in different location

<table>
<thead>
<tr>
<th>Location</th>
<th>Daily intake of Uranium (µg/day)</th>
</tr>
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<tbody>
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<td>0.27</td>
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<tr>
<td>L$_2$</td>
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<td>L$_6$</td>
<td>0.22</td>
</tr>
<tr>
<td>L$_7$</td>
<td>0.20</td>
</tr>
<tr>
<td>L$_8$</td>
<td>0.23</td>
</tr>
<tr>
<td>L$_9$</td>
<td>0.17</td>
</tr>
<tr>
<td>L$_{10}$</td>
<td>0.23</td>
</tr>
</tbody>
</table>
CONCLUSION

The ambient air quality with respect to PM$_{10}$ at Tummalapalle Uranium mining site and its surrounding located villages, along with quantitative estimation of Uranium concentration in PM$_{10}$ were evaluated. The observed values at all the sampling locations found to be within the permissible limit of 150 ng/m$^3$ (Ontario standard – 150 ng/m$^3$ and NESHAPs – 350 ng/m$^3$). No abnormal change of PM$_{10}$ in and around the mining plant was observed. The daily intake of uranium is below the standard permissible daily intake limit of uranium concentration (1.5 µg/day). Hence, these levels may not cause any imminent danger to individuals who inhale this air. Further, there was no correlation between the PM$_{10}$ and Uranium concentrations observed in all the sampling locations, indicating that they are independent to each other.

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