



## Supplement to The October 11, 2019 TFI Phosphogypsum Reuse Petition: 2019 Radium-226 Results for U.S. Phosphogypsum Stacks

December 5, 2019

### I. INTRODUCTION

On October 11, 2019, The Fertilizer Institute (“TFI”) submitted a petition (hereinafter, the “Petition”) to the U.S. Environmental Protection Agency (“EPA”) requesting approval for the beneficial use of phosphogypsum (“PG”) in road construction (*i.e.*, road base and/or concrete pavement), provided the PG contains no more than an average radioactivity concentration of 35 picocuries per gram (“pCi/g”) of Radium-226 (“Ra-226”).<sup>1</sup>

In conjunction with the submission of the Petition, TFI members agreed to perform radiological testing of PG. In particular, at EPA’s request, two TFI members utilized EPA EML HASL-300 Method Ga-01-R (a 21-day method), entitled “Gamma Radioassay,”<sup>2</sup> for the sampling event. These members’ PG stacks are located in Central Florida (where the highest Ra-226 levels would be expected), Louisiana, and the Western U.S. Because Northern Florida PG is used as a soil amendment and is regularly tested for Ra-226 using a different EPA-approved method, it is not included in this discussion.<sup>3</sup>

### II. 2019 PG SAMPLING PROGRAM AND RESULTS

Nine stacks located in four states were tested. Ten samples were taken from each stack, for a total of 90 samples.<sup>4</sup> A summary of the results is as follows:

- The average Ra-226 concentration of all 90 samples is 18.6 pCi/g.
- Ra-226 concentrations in individual samples range from 6.3 pCi/g to 27.9 pCi/g.
- The average concentration in the Western U.S. is 15.8 pCi/g.
- As expected, the highest average PG stack result is from Central Florida (24.7 pCi/g), with the average of all six Central Florida stacks being 19.6 pCi/g.

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<sup>1</sup> This level was proposed first and foremost with public safety in mind. The risk assessment analysis provided with the Petition demonstrates that PG containing Ra-226 levels up to 148 pCi/g is safe for road construction (*i.e.*, this concentration corresponds to EPA’s regulatory risk of 3 in 10,000). See Arcadis, Radiological Risk Assessment in Support of Petition for Beneficial Use of Phosphogypsum at 5-2 (Oct. 2019) (hereinafter, the “Risk Assessment”). Further, an average Ra-226 concentration of 35 pCi/g represents the high-end of expected levels of Ra-226 in U.S. PG stacks (as discussed in Section III.B, below). PG containing <10 pCi/g has been used as an agricultural amendment for many years. This PG use is not the subject of the Petition.

<sup>2</sup> This method provides conservative Ra-226 values, reported by the laboratory as “Bismuth-214 (Ra-226).”

<sup>3</sup> PG used for agriculture purposes is tested for Ra-226 using EPA Method 903.1, entitled “Radium-226 in Drinking Water Radon Emanation Technique.” Although a comparison cannot be made between the current sampling program and 2019 results from the three Northern Florida PG stacks tested using Method 903.1, for reference, the average 2019 sampling results for these three stacks (representing 30 samples) is 6.47 pCi/g.

<sup>4</sup> To preserve confidentiality, the results from the 2019 sampling program were blinded so the identity of the company and stack location were not disclosed.

Table 1 sets forth results from the 2019 sampling program. The values highlighted in yellow are the highest test result for each stack, and the values highlighted in green are the lowest results. The average and median values are also provided for each stack.

Table 1 – Aggregate PG Ra-226 Data from September 2019 (Reported as pCi/g)

Sample No.	Central Florida						Louisiana	Western U.S.	
	Stack 1	Stack 2	Stack 3	Stack 4	Stack 5	Stack 6	Stack 7	Stack 8	Stack 9
1	23.3	24.9	16.6	19.9	27.9	19.2	19.8	24.2	8.51
2	27.0	24.5	17.3	9.0	17.7	7.23	18.9	24.3	8.38
3	16.1	25.4	17.5	25.8	23.8	17.1	16.2	22.3	7.82
4	24.1	26.6	19.3	20.3	22.4	27.2	15.9	21.3	6.83
5	18.9	24.2	10.3	21.6	14.6	24.8	16.1	25.5	6.32
6	21.7	20.6	18.6	19.5	17.7	19.1	21.9	24.8	8.17
7	21.4	24.2	10.4	24.7	18.2	21.4	16.7	25.1	7.37
8	25.0	25.6	11.0	18.5	18.9	12.6	19.2	25.3	6.88
9	19.5	27.8	13.8	17.1	23.8	7.27	18.7	25.2	6.80
10	14.9	23.1	15.3	22.7	16.6	13.3	17.9	24.3	7.50
<b>Average</b>	21.19	24.69	15.01	19.91	20.16	16.92	18.13	24.23	7.46
<b>Median</b>	21.55	24.70	15.95	20.10	18.55	18.10	18.30	24.55	7.44

**III. ANALYSIS OF THE 2019 PG SAMPLING RESULTS**

**A. Comparison of the 2019 Results to The Risk Assessment Included With The Petition**

The data provided above assist in the assessment of regulatory risks. The Risk Assessment submitted in support of the Petition demonstrates that PG containing an average of 148 pCi/g corresponds to the EPA safe Ra-226 risk goal of 3 in 10,000 (see Table 2 below, reprinted from the risk assessment submitted as part of the Petition).<sup>5</sup>

Table 2 – Radionuclide Risk Assessment from the Petition

Average Ra-226 in PG*** (pCi/g)	Exposure Duration Dose (Mrem)	Risk
10	40.7	0.20 per 10,000
20	81.5	0.41 per 10,000
27	110	0.55 per 10,000
30	122	0.61 per 10,000
35	143	0.71 per 10,000
40	163	0.81 per 10,000
50	204	1.02 per 10,000

\*\*\* Although we are not aware of any PG with average Ra-226 concentrations anywhere near 148 pCi/g, such PG could in principle be used for road construction and still achieve the EPA’s safe level of a risk of 3 in 10,000.

<sup>5</sup> Risk Assessment at 5-2 (Table 5-1).

When viewed in light of the risk assessment, the 2019 PG sampling results demonstrate that the use of PG in the proposed application is consistent with EPA’s view of a safe level of risk. For example:

- The average Ra-226 concentration from the 2019 stack sampling (18.6 pCi/g) is below the radioactivity concentration limit of 35 pCi/g proposed in the Petition, and well below the average Ra-226 concentration of 148 pCi/g that corresponds to an EPA safe risk level of 3 in 10,000.
- No individual concentration from the 2019 stack sampling (with concentrations ranging from 6.32 pCi/g to 27.9 pCi/g) exceeds 35 pCi/g.
- The average concentration of the six Central Florida PG stacks (19.6 pCi/g) as well as the individual PG stack averages (ranging from 15.01 to 24.69 pCi/g) are below the radioactivity concentration limit of 35 pCi/g proposed in the Petition, and well below the average Ra-226 concentration of 148 pCi/g that corresponds to an EPA safe risk level of 3 in 10,000.
- The average concentration in the Western U.S. (15.8 pCi/g) is below the radioactivity concentration limit of 35 pCi/g proposed in the Petition, and well below the average Ra-226 concentration of 148 pCi/g that corresponds to an EPA safe risk level of 3 in 10,000.

Thus, the 2019 sampling results demonstrate that the average of all 2019 stack samples and every individual sample result are well below the proposed radioactivity concentration limit of 35 pCi/g, and significantly below EPA’s safe risk level of 148 pCi/g set forth in the Petition.

**B. Comparison to Prior EPA PG Results**

For context, in a 1992 Background Document,<sup>6</sup> EPA summarized data from a 1988 sampling of Central Florida PG stacks. The information from EPA’s Background Document is reprinted below.

Table 3 – Reprint of EPA 1988 Central Florida PG Ra-226 Sampling Results

Phosphogypsum Stack	Mean Concentration (pCi/g dry)*	Concentration Range (pCi/g dry)
Gardinier	33± 2	31-37
W. R. Grace	30 ± 9	19-48
Royster	30 ± 11	16-49
Conserve	34 ± 18	23-81
Estech	25 ± 4	19-31

\*Mean concentration with the standard deviation of samples from 10 locations on each stack.

Although the method of analysis from the 1988 sampling effort is unknown, the mean (average) results (ranging from 25 to 34 pCi/g) are higher than the average results from the 2019 sampling of Central Florida PG stacks (ranging from 15.01 to 24.69 pCi/g). However, both the 1988 and 2019 sampling results from Central Florida PG stacks demonstrate that the average concentrations are all below 35 pCi/g (the limit proposed in the Petition), and significantly below 148 pCi/g (the Ra-226 concentration corresponding to a 3 in 10,000 risk level).

<sup>6</sup> EPA, Potential Uses of Phosphogypsum and Associated Risk, Background Information Document, 402-R92-002 at 2-7 (Table 2-4) (May 1992).

**C. Comparison to International PG Sampling Results**

In addition to the 2019 and prior EPA sampling of U.S. PG stacks, data exist on the average Ra-226 concentrations in PG from 23 countries (including other data from U.S. stacks) that further confirm the 2019 U.S. PG stack data. According to published PG stack data, the Ra-226 concentrations range from 1 pCi/g (35 Bq/kg) to 38.4 pCi/g (1,420 Bq/kg)).<sup>7</sup> Representative information from the international review of PG Ra-226 data is set forth in Table 4.

Table 4 – NORM Radionuclide Activity in Phosphogypsum

Country	Ra-226 Concentration in becquerel/kilogram (Bq/kg) (converted to pCi/g)	Number of Samples	Reference
Australia	246 (6.7 pCi/g)	28	Beretka and Matthew (1985) Cooper (2005 and Msila et al. (2016)
Brazil	410 (11.1pCi/g)	268	De Conceicao and Bonotto (2006); Mazzilli and Saucia (1999) Santos (2006) Xhixha et al (2013)
Germany	305 (8.2 pCi/g)	2*	Trevisi et al. (2012)
Greece	440 (11.9 pCi/g)	31	Trevisi et al. (2012) and Turhan (2008)
Israel	747 (20.2 pCi/g)	1*	Gezer et al. (2012)
Jordan	378 (10.2 pCi/g)	16	Al-Jundi et al. (2008) Gezer et al. (2012)
Korea	618 (16.7 pCi/g)	1*	Gezer et al. (2012)
Morocco	1420 (38.4 pCi/g)	1*	Xhisha et al. (2013)
Spain	491 (13.3 pCi/g)	91	Trevisii et al. (2012)
The United Kingdom	1020 (27.6 pCi/g)	91	Trevisi et al. (2012)
United States	750 (20.3 pCi/g)	95	Hull and Brunett (1996); Roper et al. (2013)

\* Sample too small to be statistically significant.

Even though the analytical methods used in the international compilation are not known, the average for the U.S. (20.3 pCi/g) corresponds closely with the 18.6 pCi/g average concentration across all nine stacks from the 2019 sampling program. These data (as well as the 2019 TFI data and the 1988 EPA PG data) illustrate

<sup>7</sup> See Chapter 6 at Table 6.16 in “Naturally Occurring Radioactive Materials in Construction” (2017). Chapter 6: “From raw materials to NORM by-products” is authored by T. Kovacs, et al., and is available from ScienceDirect, at <https://www.sciencedirect.com/book/9780081020098/naturally-occurring-radioactive-materials-in-construction>). Section 6.6.1.2 Radiological properties from Naturally Occurring Radioactive Materials in Construction, which contains Table 6.16, is also available online at <https://www.sciencedirect.com/topics/engineering/phosphogypsum>).

that the variation of individual Ra-226 concentrations in PG is relatively low, particularly compared to the safe level of 148 pCi/g.

All of the data support the view that it is very unlikely that any PG in U.S. stacks will exceed either (1) 148 pCi/g (EPA's PG safe level), or (2) 35 pCi/g (the level proposed in the Petition for reuse). Thus, no sampling should be required prior to shipping PG for road construction uses.

#### **IV. CONCLUSION**

Key highlights from this Supplement to TFI's Petition are set forth below:

- As expected, the highest levels of naturally occurring radiation are associated with the Central Florida samples.
- All of the data demonstrate that the individual, average, and medium Ra-226 concentrations from U.S. PG are below 35 pCi/g, and well below the 148 pCi/g level corresponding to EPA's safe level of 3 in 10,000.

These results verify that the Petition contains a reasonable maximum limit for the beneficial use of PG in U.S. road construction.