Elevated Levels of Naturally Occurring Radioactive Material Associated with Shale Gas Extraction*

John Mauro[1]

Search and Discovery Article #80266 (2012)**
Posted October 22, 2012

*Adapted from oral presentation given at AAPG Eastern Section meeting, Cleveland, Ohio, 22-26 September 2012

[1] SC&A, Inc., Vienna, VA (jmauro@scainc.com)

Abstract

Naturally occurring radioactive materials, such as radium-226, are ubiquitous in the earth's crust, and, in some geologic formations and their associated ground waters, the levels of these radionuclides are often elevated. In addition, the extraction and processing of the minerals and ground water associated with these formations can further enhance the concentration of these radionuclides, resulting in products, byproducts, and waste products containing what is referred to as technologically enhanced sources of naturally occurring radioactive materials or TENORM. Examples of TENORM include solids associated with mineral extraction and beneficiation industries, including heap leach extraction, in situ-mineral extraction, thorium and rare earths processing, phosphate industry tailings, sludge at water and wastewater treatment facilities, spent resin from individual water softeners, coal ash, oil and gas production sludge and scale, and geothermal energy production waste.

With respect to shale gas extraction, elevated levels of naturally occurring uranium and radium have been found in rock cuttings and drilling fluid/mud. For example, uranium has been found in shale at 59.4 pCi/g as compared to about 0.85 pCi/g in typical soil. Hydraulic fracturing flowback water (HFFW or cocktail) has been observed to contain up to18,950 pCi/L gross alpha (which consists primarily of radium-226 and its progeny), and produced water has been observed to contain up to 15,000 pCi/L of gross alpha emitters. These concentrations can be compared to the drinking water standards set forth in 40CFR141 of 15 pCi/L gross alpha and 5 pCi/L combined Ra-226 and Ra-228 water.

The industry and state and federal regulatory authorities are currently involved in the analysis of the radionuclide composition of rock cuttings, drilling fluid/mud, HFFW, and produced water for the purpose of evaluating the potential health risks associated with the handling, management, transport, and disposal of these materials. To date, it has been found that the limiting radionuclide is Ra-226, and that its concentration in the various products, byproducts, and waste products associated with the shale gas extraction industry is highly variable depending on location. In addition, investigations to date reveal that under most circumstances TENORM does not pose a hazard to workers, members of the public, or to the environment. However, our understanding of the nature and extent of TENORM associated with this

industry is incomplete, and careful attention must be given to characterizing the composition of the products, byproducts, and waste products associated with shale gas extraction in order to ensure that the industry complies with all applicable state and federal regulations and guidelines, many of which are still in the formative stages.

Selected References

Eisenbud, M., and T.F. Gesell, 1997, Environmental Radioactivity: from natural, industrial, and military sources, 4th edition: Academic Press, San Diego, California, 656 p.

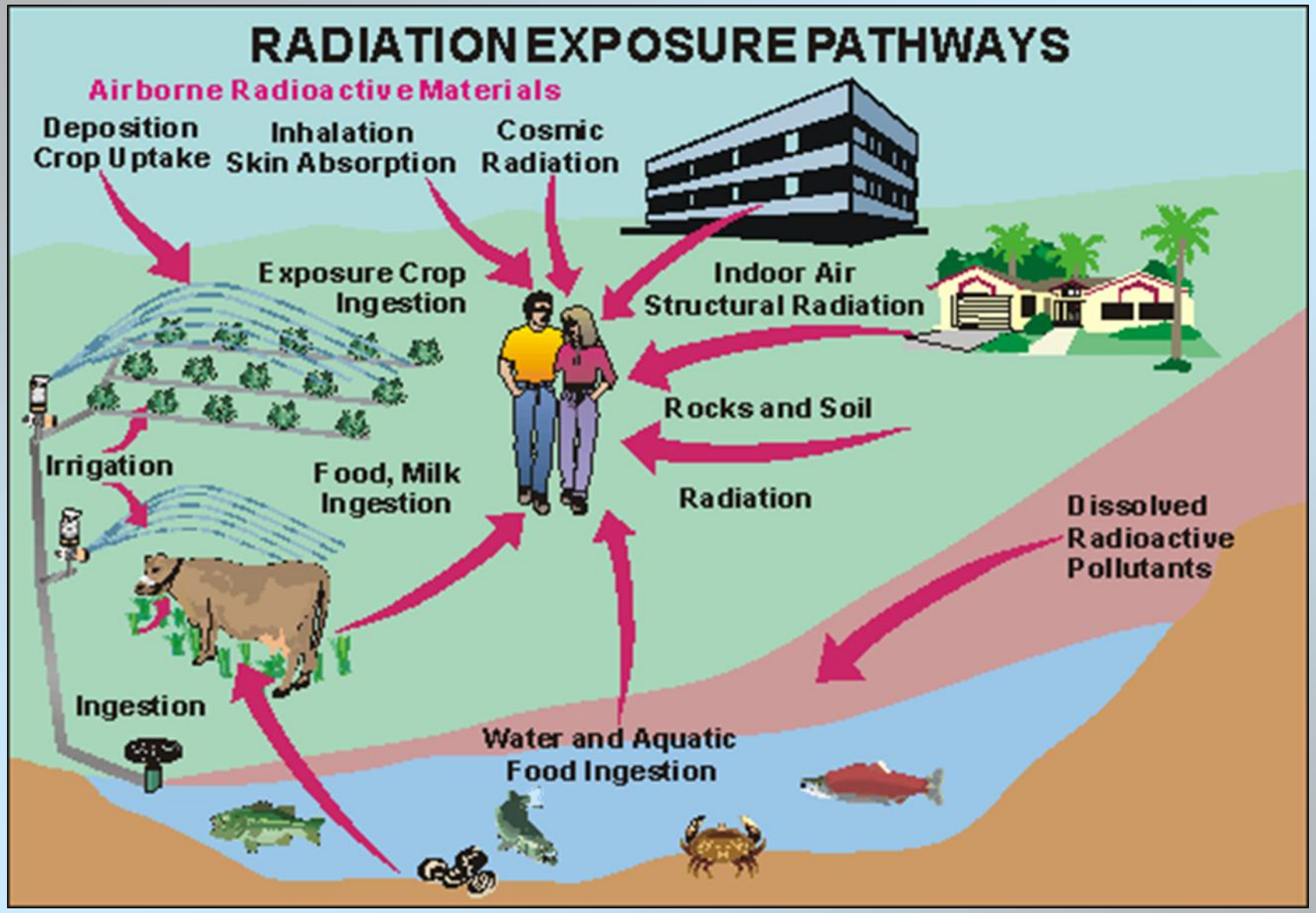
Hanlon, J.A., 2011, Regulating Natural Gas Drilling in Marcellus Shale under the NPDES Program: Memorandum from EPA Director Office of Wastewater Management to Water Division Directors, Regions 1-10 March 17. Web accessed 12 October 2012. http://www.mde.state.md.us/programs/Land/mining/marcellus/Documents/EPA_Marcellus_QsAs_cover_memo.pdf

Elevated Levels of Naturally Occurring Radioactive Material Associated with Shale Gas Extraction

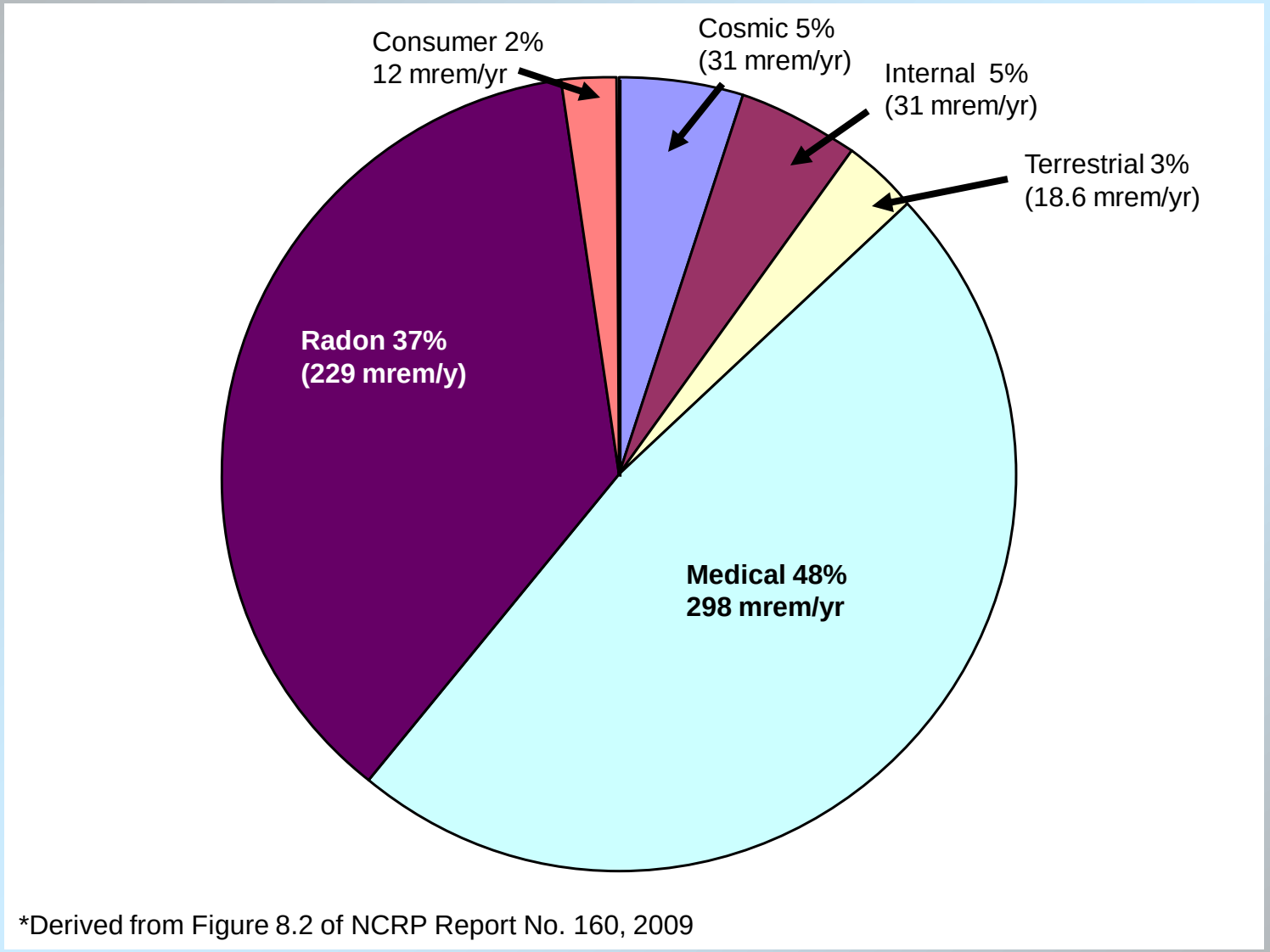
Presented to
AAPG Eastern Section

John Mauro, PhD, CHP
SC&A, Inc.
Vienna, VA

September 24, 2012
jmauro@scainc.com

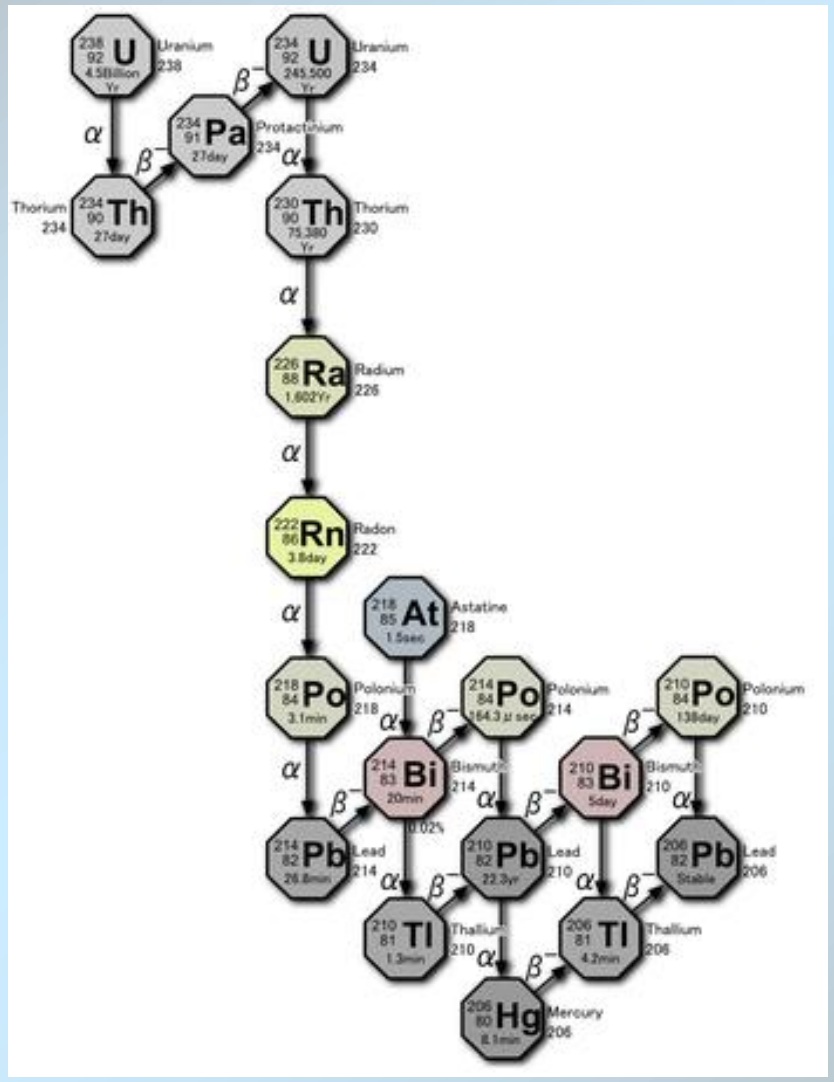

RADIATION EXPOSURE PATHWAYS
Airborne Radioactive Materials
Deposition
Crop Uptake
Inhalation
Skin Absorption
Cosmic
Radiation
Exposure Crop
Ingestion
Indoor Air
Structural Radiation
Rocks and Soil
Irrigation
Food, Milk
Ingestion
Radiation
Dissolved
Radioactive
Pollutants
Ingestion
Water and Aquatic
Food Ingestion

Average Total Effect Dose per Person in U.S. Population for 2006*
Total = 620 mrem per year (0.620 rem per year)



*Derived from Figure 8.2 of NCRP Report No. 160, 2009

Naturally Occurring Radionuclides



Units of Quantities of Radioactive Material and Radiation Exposure

We will be talking about picocuries (pCi) and millirems (mrem)

What is a picocurie?

By definition, 1 gram of radium-226 = 1 Curie (Ci)

1 Ci = 3.7×10^{10} disintegrations per second (dps)

"Pico" means 1/trillionth or 10^{-12}

Typical Levels of Naturally Occurring Radionuclides in the Environment*

Media	Uranium-238	Radium-226	Thorium-232
Typical Rock/soil (pCi/g)	0.2–1.6	0.2–1.6	0.2–2.2
Typical Groundwater (pCi/L)		0.12–2.19	

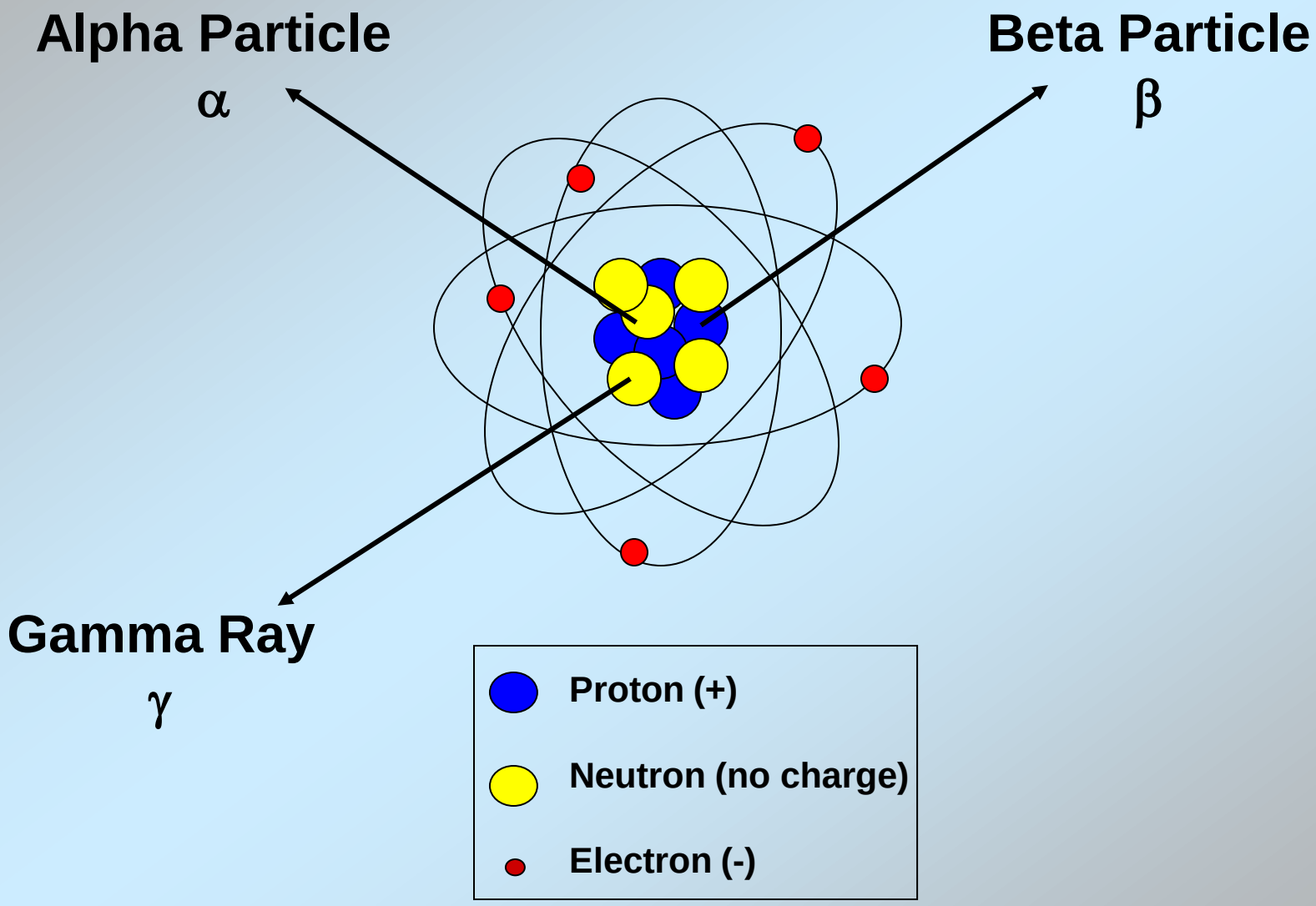
* From Eisenbud, M. and T. Gesell, *Environmental Radioactivity*, 4th Edition (1997)

What is a mrem?

A mrem is a measure of the amount of ionizing radiation energy deposited per gram of tissue

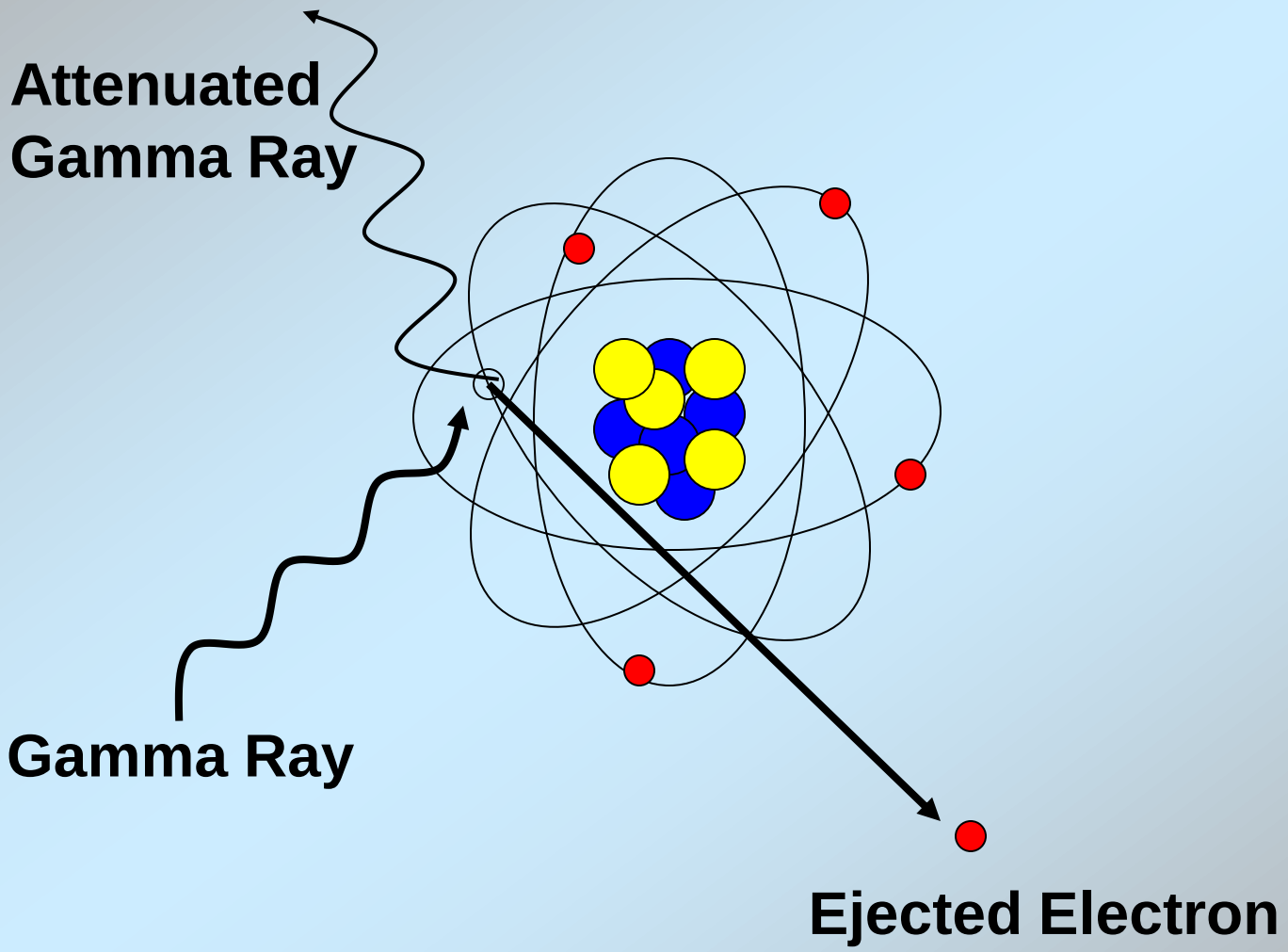
If you receive 1 mrem of uniform whole body exposure,
1.8 billion ion pairs will have been produced in every gram of tissue in your body

Types of Emissions from Radioactive Materials

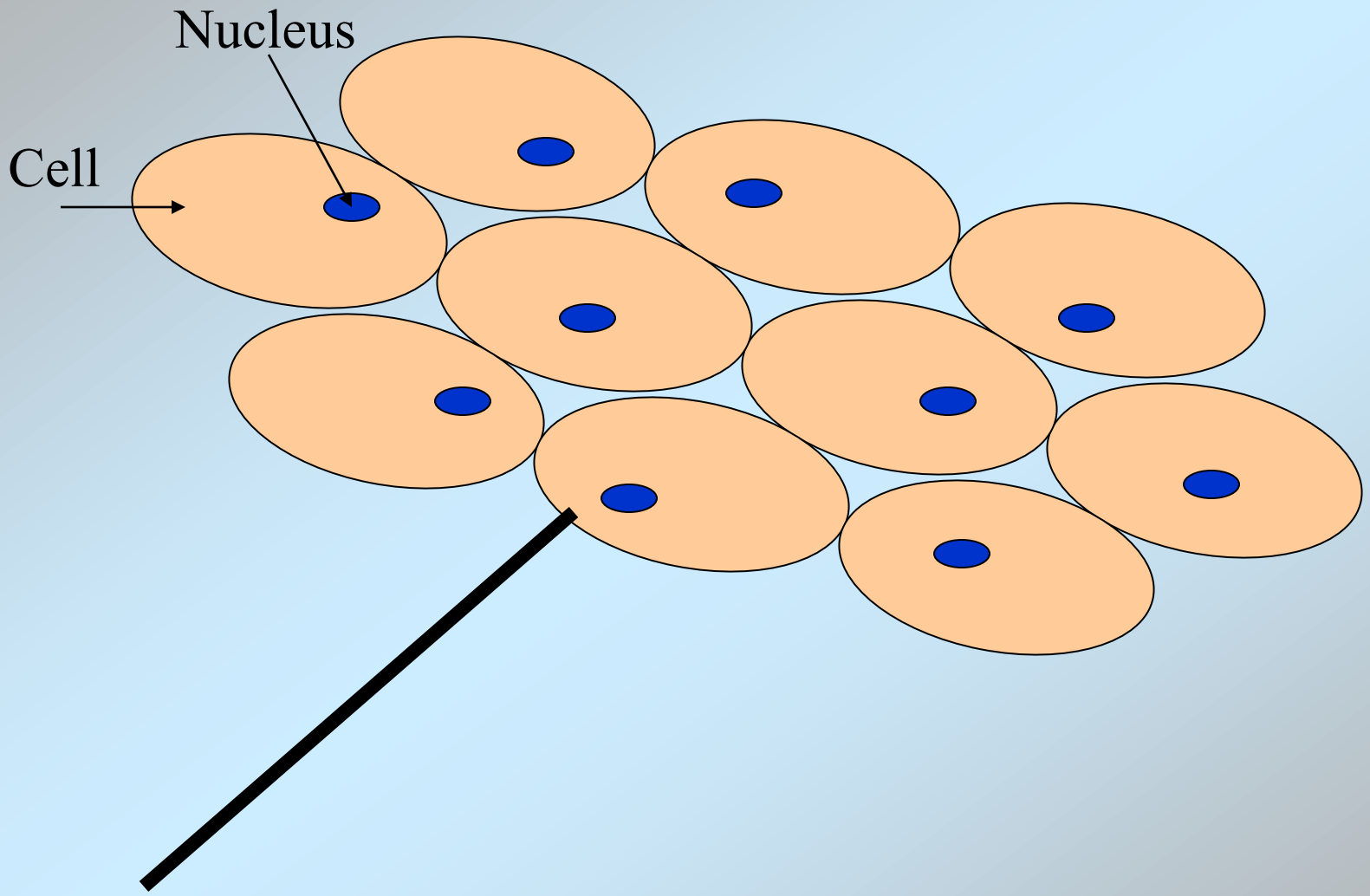


Ionizing Radiation

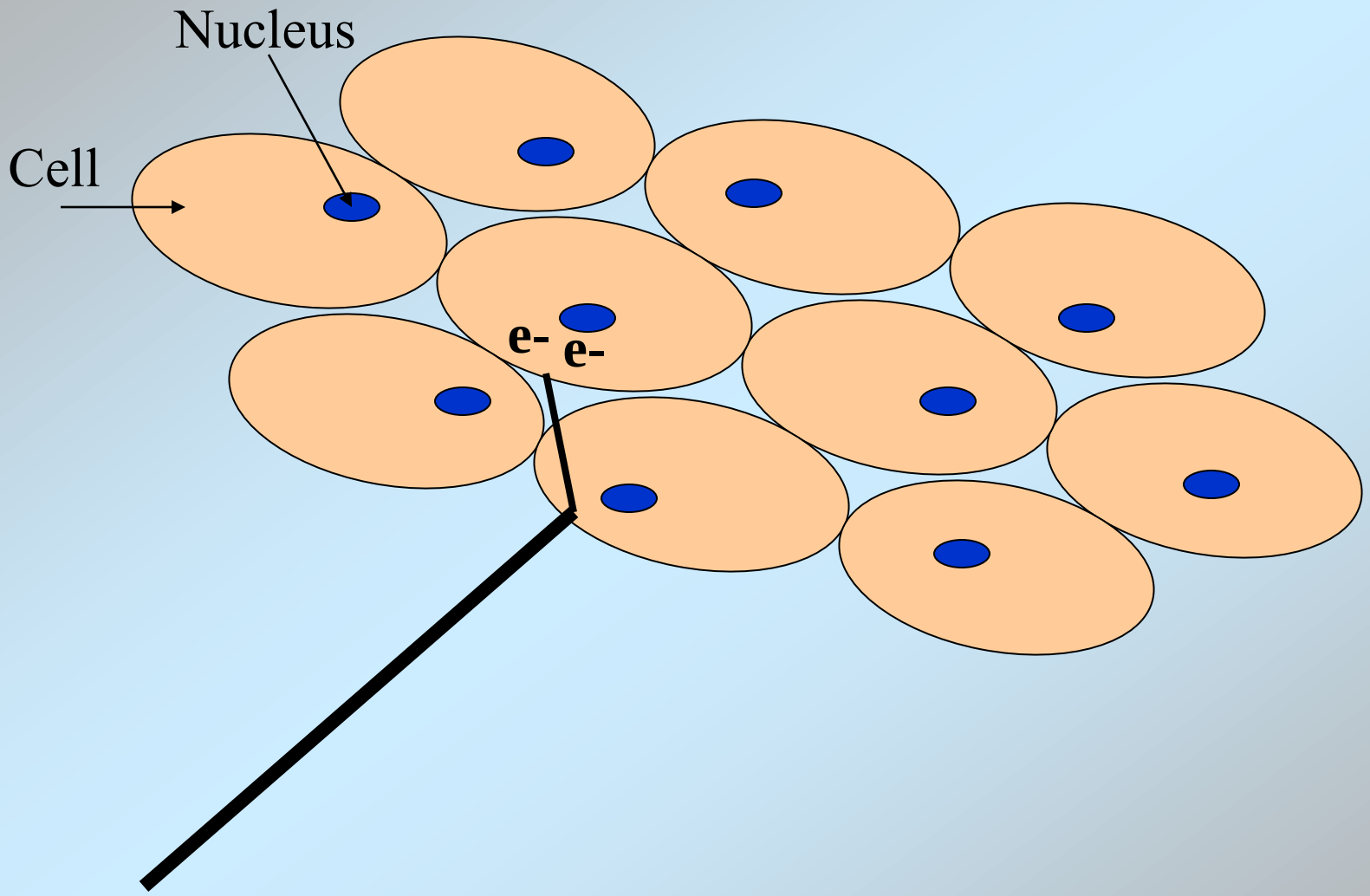
Radiation that has Enough Energy to Ionize an Atom
(typically 35–40 electron volts)



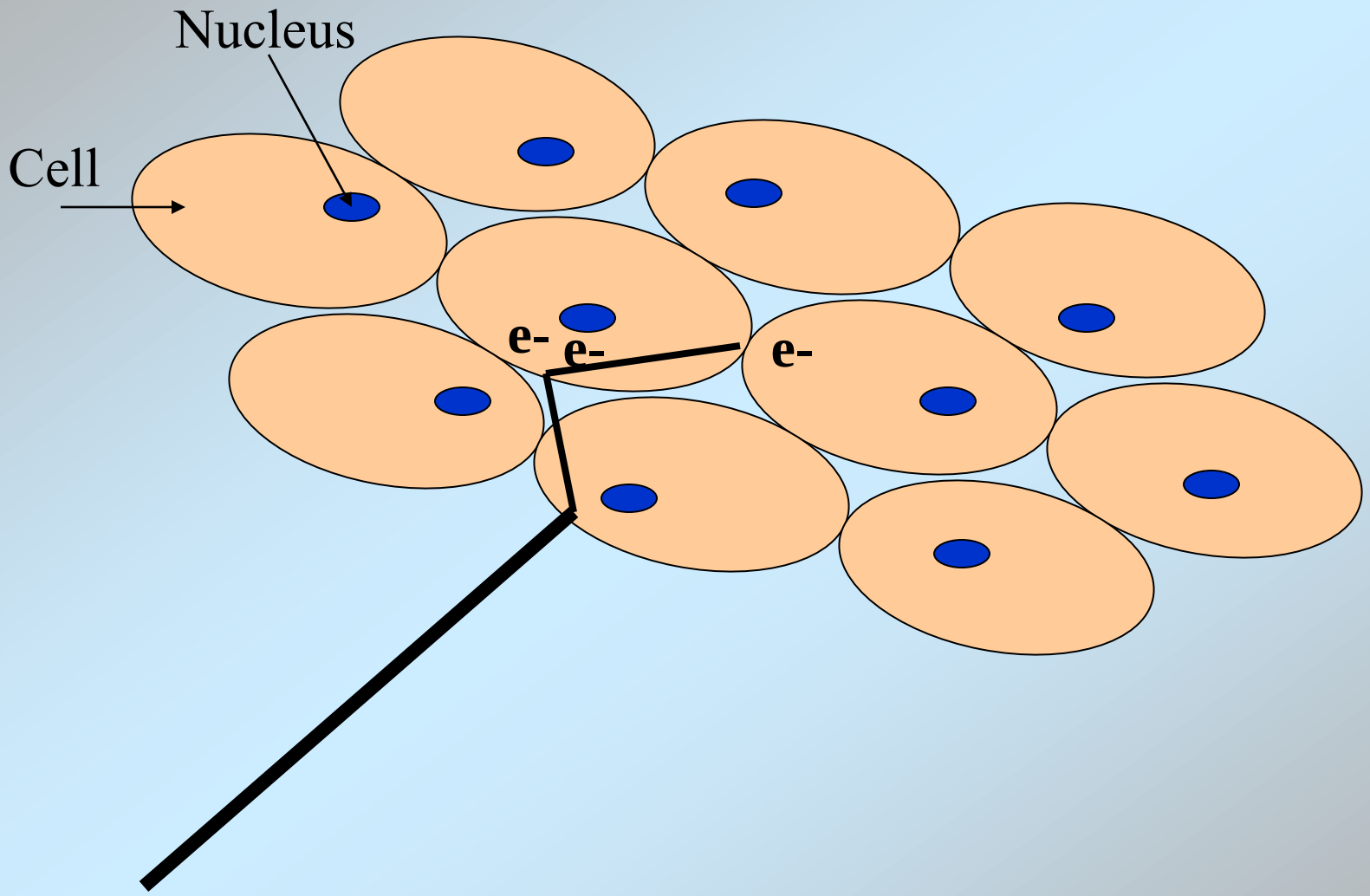
Interaction of Radiation with Tissue



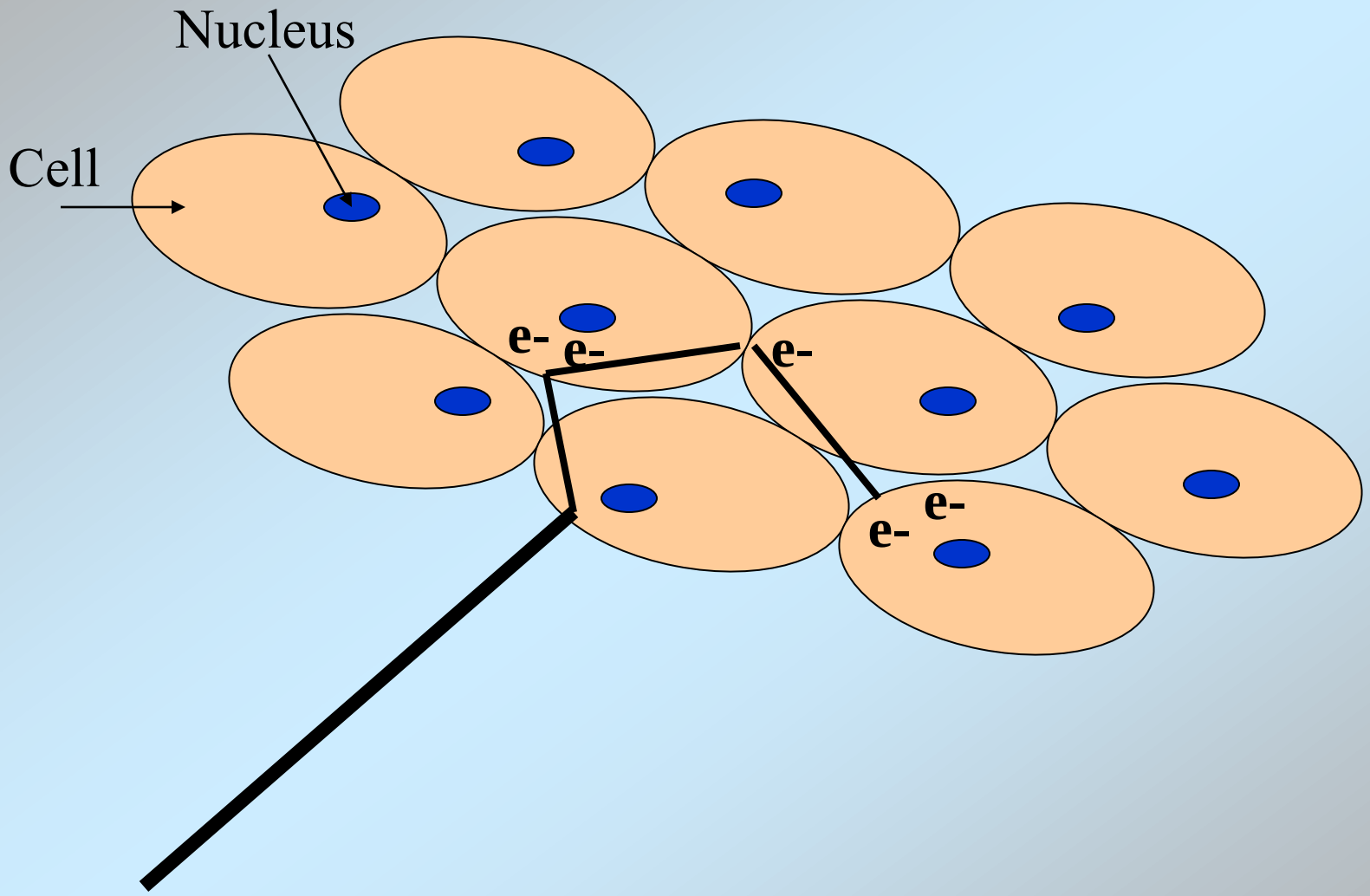
Interaction of Radiation with Tissue



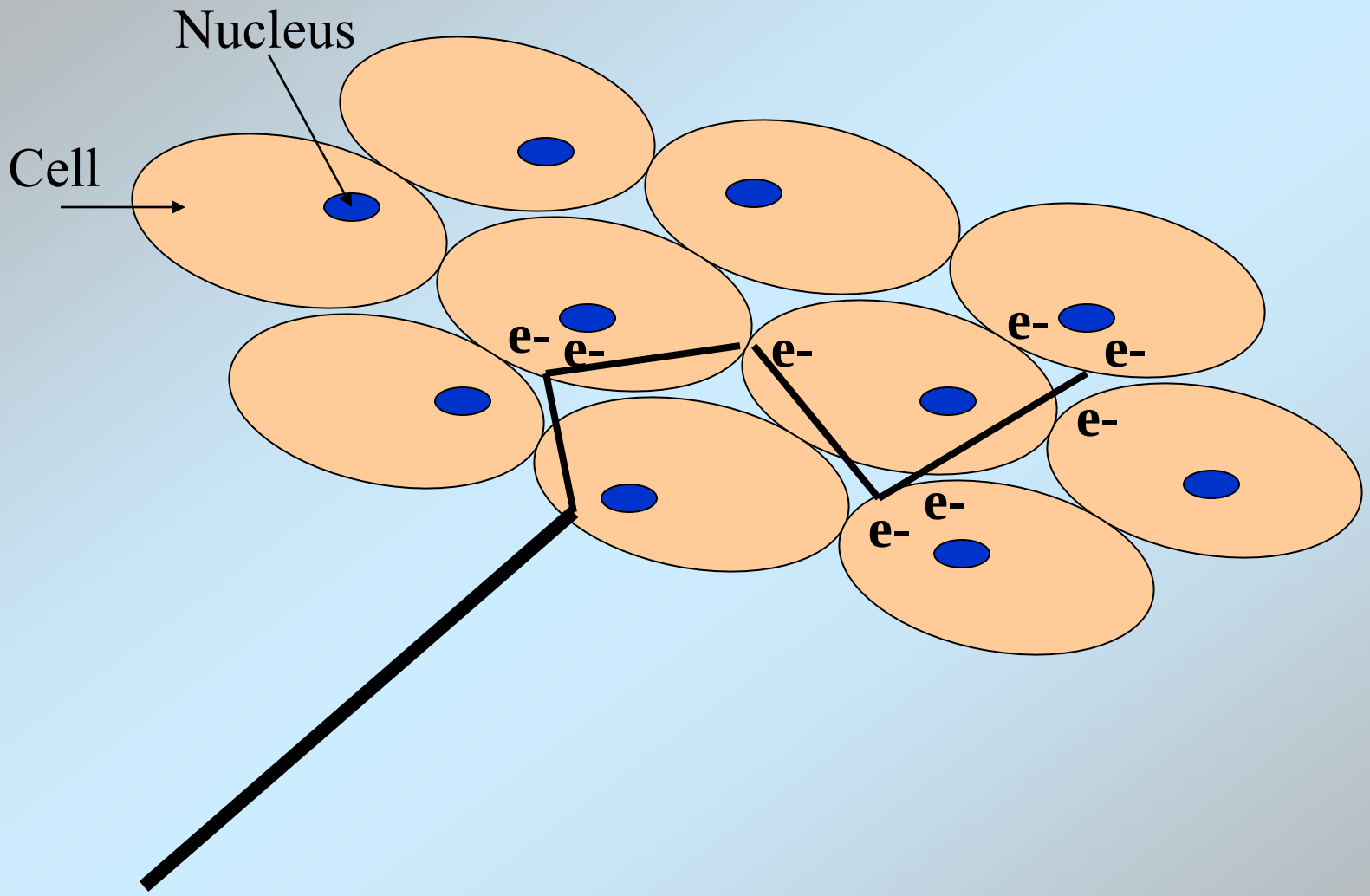
Interaction of Radiation with Tissue



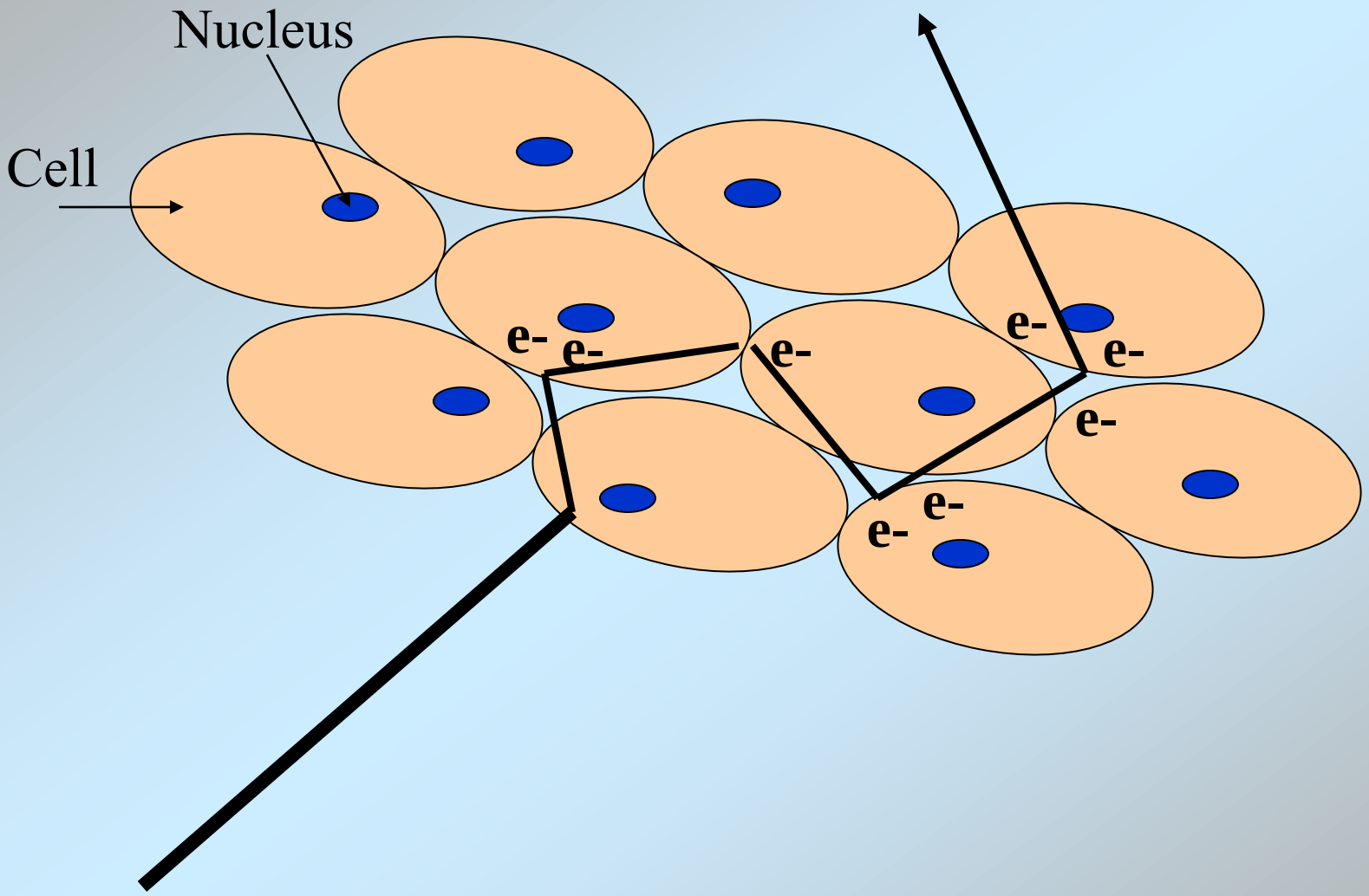
Interaction of Radiation with Tissue



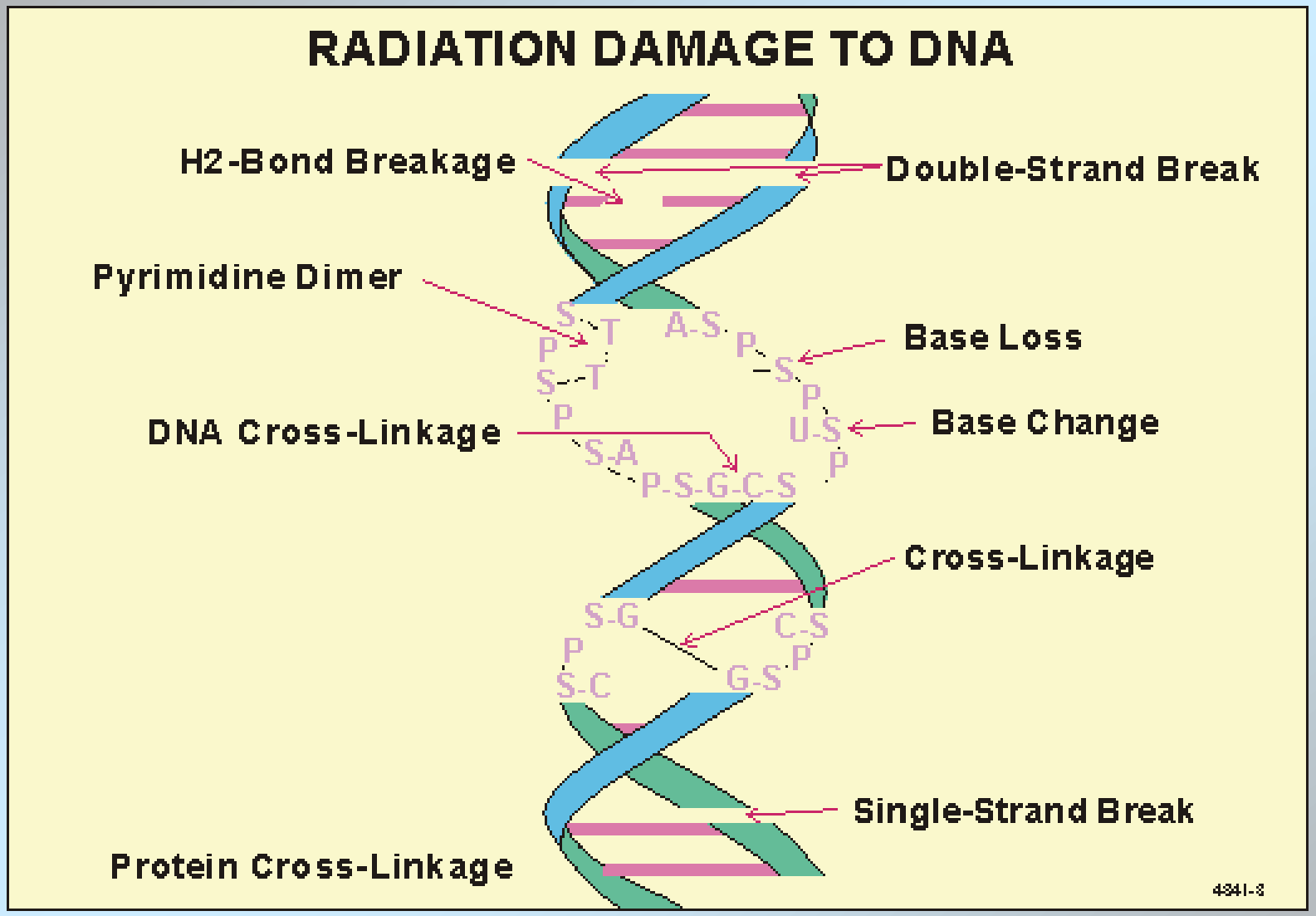
Interaction of Radiation with Tissue



Interaction of Radiation with Tissue



Interaction of Radiation with Genetic Material



Source: Scott, Bobby. 2010. Lovelace Respiratory Research Institute <http://www.radiation-scott.org/radsource/index.htm>

Chronic (Stochastic) Effects

Lifetime risk of fatal cancer per rem = 5×10^{-4}

Dose which doubles cancer risk = 200 rem

Examples of TENORM
(Technologically Enhanced Naturally Occurring Radioactive Material)

Solids Associated with Mineral Extraction and Beneficiation Industries

- **Heap leach extraction**
- **In-situ mineral extraction**
- **Thorium and rare earths processing**
- **Phosphate industry tailings**

Solids Associated with Water and Wastewater Treatment

- **Sludge at water and wastewater treatment facilities**
- **Spent resin from individual water softeners**

Coal Ash

Oil and Gas Production Sludge and Scale

Geothermal Energy Production Waste

NORM and TENORM in Marcellus Shale

Elevated levels of Uranium, Ra-226 and Ra-228 are found in:

Rock cuttings and drilling fluid/mud (uranium found at 59.4 pCi/g in shale a compared to about 2.8 pCi/g in natural background, and rock cuttings observed to have radium concentrations at about the same values. Radium in typical soil is at about 0.85 pCi/g)**

Hydraulic fracturing flowback water (HFFW or Cocktail): 18,950 pCi/L gross alpha have been observed in HFFW* [Drinking water standards (MCLs in 40CFR141) are 15 pCi/L gross alpha and 5 pCi/L combined Ra-226 and Ra-228]

Produced water: 15,000 pCi/L of gross alpha emitters have been observed in produced water. [Drinking water standards (MCLs in 40CFR141) are 15 pCi/L gross alpha and 5 pCi/L combined Ra-226 and Ra-228]**

Solids generated from the evaporation of produced water

*** See Regulating Natural Gas Drilling in Marcellus Shale under the NPDES Program. Memorandum from James A. Hanlon, EPA Director Office of Wastewater Management to Water Division Directors, Regions 1–10 (March 17, 2011).**

**** See "Radioactivity in Marcellus Shale" Radioactive Waste Management Associates (May 19, 2010).**

Some Recent Measurements of Radium in Produced Water from Marcellus Shale

Well	Radionuclide Concentration in Raw Water (pCi/L)		Total Solids of Raw Water	Sludge Activity (pCi/g dry wt)	
	Ra-226	Ra-228	mg/L	Ra-226	Ra-228
A	12,139	1,002	177,643	68.3	5.6
B	1,337	1,339	262,000	5.1	5.1
C	761	713	259,000	2.9	2.8
D	513	378	254,000	2	1.5
E	271	199	163,000	1.7	1.2
F	129	120	137,000	0.9	0.9
G	74	72	195,000	0.4	0.4
H	17,100	1,580	217,000	78.8	7.3
I	17,000	1,610	220,000	77.3	7.3
J	18,300	1,300	158,000	115.8	8.2
K	10,500	1,950	303,000	34.7	6.4
L	11,100	1,580	240,000	46.3	6.6
M	44	1,800	265,000	0.2	6.8
N	8,100	1,150	185,000	43.8	6.2
O	9,700	1,550	235,000	41.3	6.6
P	9,600	1,700	260,000	36.9	6.5
Q	10,900	1,190	182,000	59.9	6.5
R	9,320	1,210	195,000	47.8	6.2
S	10,500	1,420	217,000	48.4	6.5
T	12,700	1,250	207,000	61.4	6
U	12,900	1,360	209,000	61.7	6.5
V	16,900	1,880	248,000	68.1	7.6
w	8,010	1,650	220,000	36.4	7.5
			Mean	40.9	5.5
			SD	31.6	2.4
			GM	17	4.5
			GSD	6.9	2.3

Indoor Radon

A number of EPA publications (as cited in a report by Marvin Resnikoff) report that elevated levels of radon have been observed in homes due to radon entering homes along with natural gas.

Radiation Protection Standards Applicable to TENORM

- American National Standards Institute, *Control and Release of Technologically Enhanced Naturally Occurring Radioactive Material (TENORM).* ANSI/HPS N13.53-2009. March 2009.
- Interagency Steering Committee on Radiation Standards. *ISCORS Assessment of Radioactivity in Sewage Sludge: Modeling to Assess Radiation Dose, ISCORS Technical Report 2004-04*, NUREG-1783; EPA 832-R-03-002; DOE/EH-0670 (February 2005).
- National Council on Radiation Protection and Measurement, *Radiation Protection in the Mineral Extraction Industry*, NCRP Report No. 118 (1993).
- Conference of Radiation Control Program Directors. Part N, *Suggested Regulation Template for TENORM.*
- National Research Council, *Evaluation of Guidelines for Exposure to Technological Naturally Occurring Radioactive Materials*. Committee on Evaluation of Guidelines for Exposure to Naturally Occurring Radioactive Materials. ISBN: 0-309-58070-6 (1999).
- Basic Radiation Safety Standards (BSS) and guidance issued by the National Academy of Sciences, International Commission on Radiation Protection, and the International Atomic Energy Agency dealing with clearance, exemption, exclusion, and intervention of material containing radioactive material.

Radiation Exposure Limits

The primary public and occupational dose limits that will be used by state and federal authorities in evaluating plans for the management and disposition of TENORM at a facility are as follows:*

Occupationally exposed radiation workers	5000 mrem/yr
Occupationally exposed members of the public (non-radiation workers)	100 mrem/yr
Vehicle drivers (non-radiation workers)	100 mrem/yr
Drinking water pathway for members of the public	4 mrem/yr
General public exposed to non-radon airborne emissions	10 mrem/yr
General public all pathways combined	25 mrem/yr

In addition to these limits, exposures should be maintained as low as is reasonably achievable (ALARA).

Radium in drinking water	5 pCi/L
EPA Guidelines on Indoor Airborne Radon	4 pCi/L

*Each state will likely require site owners to demonstrate compliance with these, and possibly additional acceptance criteria and guidelines as part of an operating permit for the facility.

OVERARCHING FEDERAL AND STATE REGULATIONS*

Hydraulic fracturing flowback water (HFFW) and produced water require an NPDES Permit under the Clean Water Act, including pretreatment requirements.

Common constituents of shale gas extraction (SGE) wastewater include calcium, magnesium (hardness), phosphates, nitrates, sodium, potassium, sulfates, chlorides, barium, cadmium, and copper which can interfere with Publicly Owned Treatment Work (POTW) operations. These can inhibit anaerobic digestion processes and biological treatment units and metals can precipitate contaminating biosolids. TDS in SGE wastewater have been measured up to 345,000 mg/L.

PA prohibits direct or indirect discharge of HFFW and produced water to POTWs because of (1) pass through, (2) interference by such water with POTW operations due to high dissolved solids content as allowed by 40 CFR 403.5(b), and (3) land farming issues.

Solids are regulated under the Solid Waste Management Act and Radiation Protection Act.

Effluent guidelines at 40 CFR 435, Subpart C for onshore facilities state "there shall be no discharge of wastewater pollutants into navigable waters from any sources associated with production, field exploration, drilling, well completion, or well treatment (i.e., produced water, drilling muds, drill cuttings and produced sand)."

*** See Regulating Natural Gas Drilling in Marcellus Shale under the NPDES Program. Memorandum from James A. Hanlon, EPA Director Office of Wastewater Management to Water Division Directors, Regions 1–10 (March 17, 2011).**

PADEP GUIDANCE Applicable to TENORM Associated with Marcellus Shale

PaDEP has developed guidance and requirements regarding an action plan for the management of TENORM associated with gas extraction. The guidance and requirements developed by PaDEP are contained in Document Number 250-3100-001, titled *Final Guidance on Radioactivity Monitoring at Solid Waste Processing and Disposal Facilities* (January 2, 2004), and are designed primarily for use by owners and operators of solid waste processing and disposal facilities. However, this same guidance is useful in ensuring the protection of workers and other members of the public at or in the vicinity of the well-field where the solids are produced.

The Radiation Monitoring and Action Plan (RMAP)

Facilities that produce, process, and/or transfer radioactive material that can set off radiation alarms must (1) screen incoming loads for the presence of radioactivity, and (2) have a PaDEP-approved Radiation Monitoring and Action Plan (RMAP). The RMAP must contain the following information:

- Management responsibilities
- Radiation detection equipment and initial procedure
- Designated storage area
- Action levels
- Follow-up actions
- Determination of origin
- Disposition and/or storage of radioactive material
- Training for facility staff
- Procedures for surveying
- Posting suspect containers
- Program oversight
- Periodic plan review and update
- Records of surveys

Pennsylvania Standards Related to the Disposal of TENORM Associated with Marcellus Shale

TENORM shall not be disposed/processed at a solid waste management or disposal facility unless approved in writing by PaDEP, and the disposal/ processing of the material shall not endanger the health and safety of the public and the environment. As applied to the solids TENORM is defined as any naturally occurring material that creates a gamma radiation field in the vicinity of the solid waste container that exceeds 10 microRoentgen per hour (μR/hr) above natural background (where natural background is at or below 10 μR/hr). If any location in the vicinity of the shipment is above this limit, the material is defined as TENORM and must be managed and disposed as TENORM.