Risks of Shale Gas Exploration and Hydraulic Fracturing to Water Resources in the United States

Avner Vengosh, Robert B. Jackson, Nathaniel Warner, Thomas H. Darrah

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Risks of Shale Gas Exploration and Hydraulic Fracturing to Water Resources in the United States

Short term

Long term

- Stray gas contamination;
- Surface water contamination via disposal of inadequately treated wastewater;
- Spills;



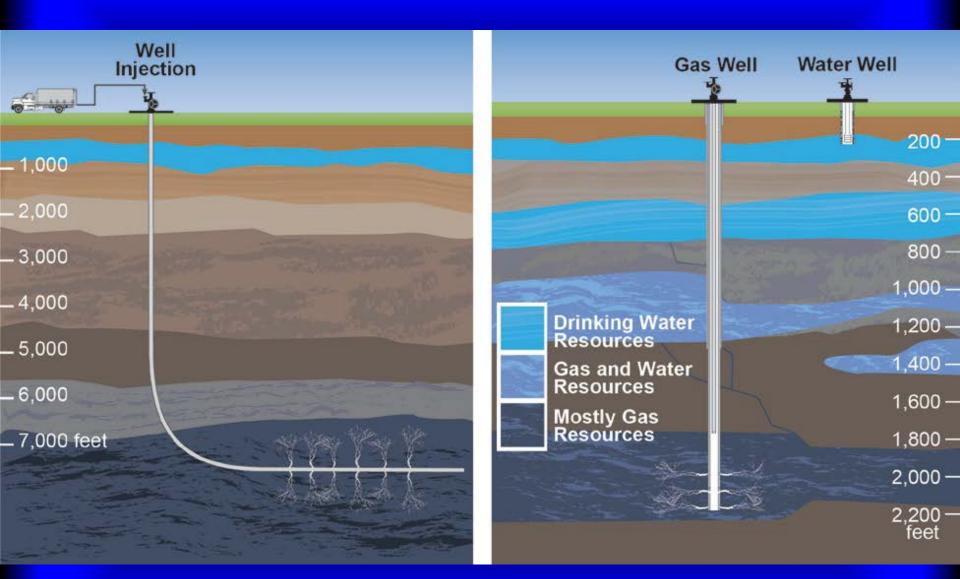
- Water availability in water scarce areas;
- Groundwater contamination through natural fracture networks;
- Groundwater contamination through abandoned and improperly sealed conventional oil and gas wells;
- Accumulation of residual contaminants and radiation in areas of wastewater disposal and spills;

Stray gas contamination

The risks:

- Occurrence of elevated levels of methane and in shallow drinking water wells can pose a potential flammability or explosion hazard to homes near shale gas drilling sites;
- Shut-down of private drinking water wells, need for alternative water resources;
- Houses and property devaluation;





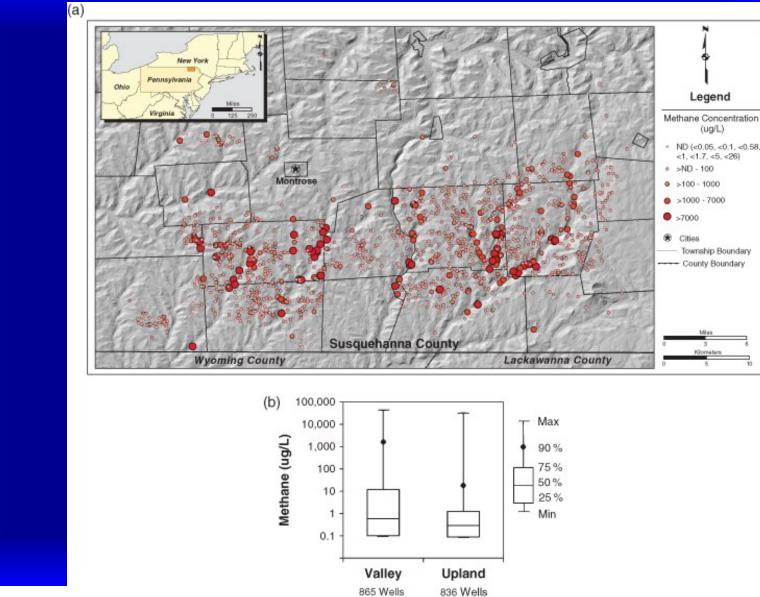
Source: EPA Progress Report 2012

The debate on stray gas contamination

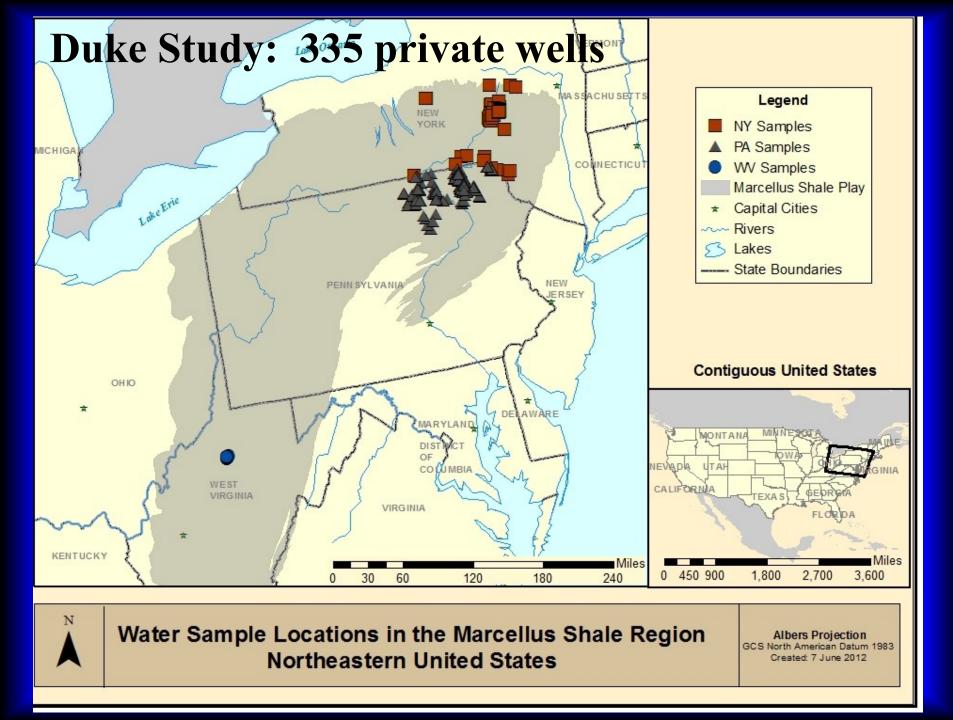
No risk:

Methane is ubiquitous in groundwater, with higher concentrations observed in valleys vs. upland; methane concentrations are best correlated to topographic and hydrogeologic features, rather than shale-gas extraction (Molofsky et al., 2013). High risk in a subset of wells near shale gas sites : Evidence for stray gas contamination in a subset of wells less than a km from shale gas sites in northeastern PA (Osborn et al., 2011; Darrah et al., 2012).

Methane is ubiquitous in PA groundwater



Molofsky et al., 2013; Groundwater, 3 333–349



Methane contamination of drinking water accompanying gas-well drilling and hydraulic fracturing

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*Center on Global Change, Nicholas School of the Environment, *Division of Earth and Ocean Sciences, Nicholas School of the Environment, and 'Biology Department, Duke University, Durham, NC 27708

Edited* by William H. Schlesinger, Cary Institute of Ecosystem Studies, Millbrook, NY, and approved April 14, 2011 (received for review January 13, 2011)

Directional drilling and hydraulic-fracturing technologies are dramatically increasing natural-gas extraction. In aguifers overlying the Marcellus and Utica shale formations of northeastern Pennsylvania and upstate New York, we document systematic evidence for methane contamination of drinking water associated with shalegas extraction. In active gas-extraction areas (one or more gas wells within 1 km), average and maximum methane concentrations in drinking-water wells increased with proximity to the nearest gas well and were 19.2 and 64 mg $CH_4 L^{-1}$ (n = 26), a potential explosion hazard; in contrast, dissolved methane samples in neighboring nonextraction sites (no gas wells within 1 km) within similar geologic formations and hydrogeologic regimes averaged only 1.1 mg L⁻¹ (P < 0.05; n = 34). Average δ^{13} C-CH₄ values of dissolved methane in shallow groundwater were significantly less negative for active than for nonactive sites (-37 \pm 7‰ and -54 \pm 11‰, respectively; P < 0.0001). These δ^{13} C-CH₄ data, coupled with the ratios of methane-to-higher-chain hydrocarbons, and δ^2 H-CH₄ values, are consistent with deeper thermogenic methane sources such as the Marcellus and Utica shales at the active sites and matched gas geochemistry from gas wells nearby. In contrast, lower-concentration samples from shallow groundwater at nonactive sites had isotopic signatures reflecting a more biogenic or mixed biogenic/ thermogenic methane source. We found no evidence for contamination of drinking-water samples with deep saline brines or fracturing fluids. We conclude that greater stewardship, data, andpossibly-regulation are needed to ensure the sustainable future of shale-gas extraction and to improve public confidence in its use.

groundwater | organic-rich shale | isotopes | formation waters | water chemistry

ncreases in natural-gas extraction are being driven by rising energy demands, mandates for cleaner burning fuels, and the economics of energy use (1-5). Directional drilling and hydraulic-fracturing technologies are allowing expanded natural-gas extraction from organic-rich shales in the United States and elsewhere (2, 3). Accompanying the benefits of such extraction (6, 7)are public concerns about drinking-water contamination from drilling and hydraulic fracturing that are ubiquitous but lack a strong scientific foundation. In this paper, we evaluate the potential impacts associated with gas-well drilling and fracturing on shallow groundwater systems of the Catskill and Lockhaven formations that overlie the Marcellus Shale in Pennsylvania and the Genesee Group that overlies the Utica Shale in New York (Figs. 1 and 2 and Fig. S1). Our results show evidence for methane contamination of shallow drinking-water systems in at least three areas of the region and suggest important environmental risks accompanying shale-gas exploration worldwide.

The drilling of organic-rich shales, typically of Upper Devonian to Ordovician age, in Pennsylvania, New York, and elsewhere in the Appalachian Basin is spreading rapidly, raising concerns for impacts on water resources (8, 9). In Susquehanna County, Pennsylvania alone, approved gas-well permits in the Marcellus formation increased 27-fold from 2007 to 2009 (10).

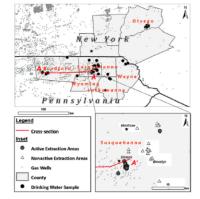


Fig. 1. Map of drilling operations and well-water sampling locations in Pennsylvania and New York. The star represents the location of Binghamton, New York. (*Nres*?) A close-up in Susquehanana County, Pennsylvania, showing areas of active (closed circles) or nonactive (open triangles) extraction. A drinking-water well is classified as being in an active extraction area if a gas well is within 1 km (see *Methods*). Note that drilling has already spread to the area around Brooklyn, Pennsylvania, primarily a nonactive location at the time of our sampling (see inset). The stars in the inset represent the towns of Dimock, Brooklyn, and Montrose, Pennsylvania.

Concerns for impacts to groundwater resources are based on (i) fluid (water and gas) flow and discharge to shallow aquifers due to the high pressure of the injected fracturing fluids in the gas wells (10); (ii) the toxicity and radioactivity of produced water from a mixture of fracturing fluids and deep saline formation waters that may discharge to the environment (11); (iii) the potential explosion and asphyxiation hazard of natural gas; and (iv) the large number of private wells in rural areas that rely on shallow groundwater for household and agricultural use—up to one million wells in Pennsylvania alone—that are typically unregulated and untested (8, 9, 12). In this study, we analyzed groundwater from 68 private wells from 36- to 190-m deep in

- Freely available online through the PNAS open access option.
- ¹To whom correspondence should be addressed. E-mail: jackson@duke.edu. This article contains supporting information online at www.pnas.org/lookup/suppl/
- doi:10.1073/pnas.1100682108/-/DCSupplemental.

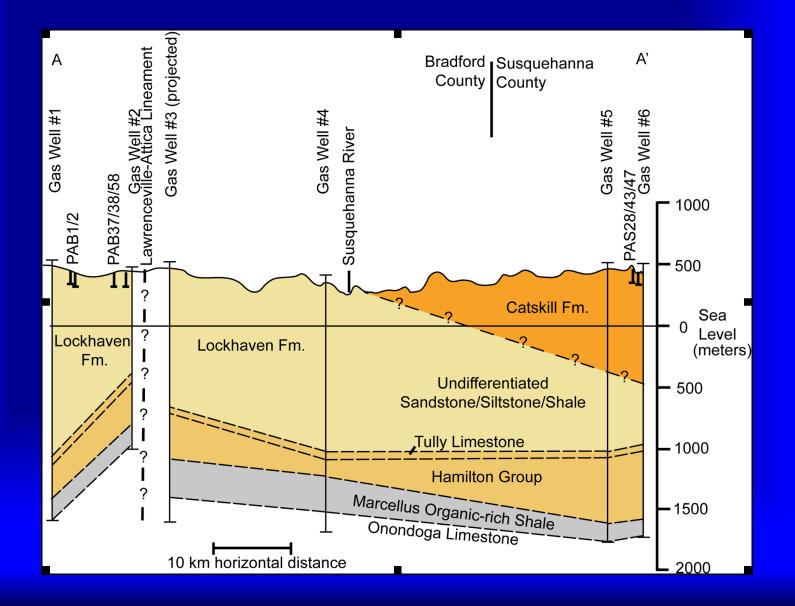
Proceedings of National Academy of Sciences, May 17, 2011

Author contributions: S.G.O., A.V., and R.B.J. designed research; S.G.O. and N.R.W. performed research; A.V. contributed new reagents/analytic tools; S.G.O., A.V., N.R.W., and R.B.J. analyzed data; and S.G.O., A.V., N.R.W., and R.B.J. wrote the paper.

The authors declare no conflict of interest

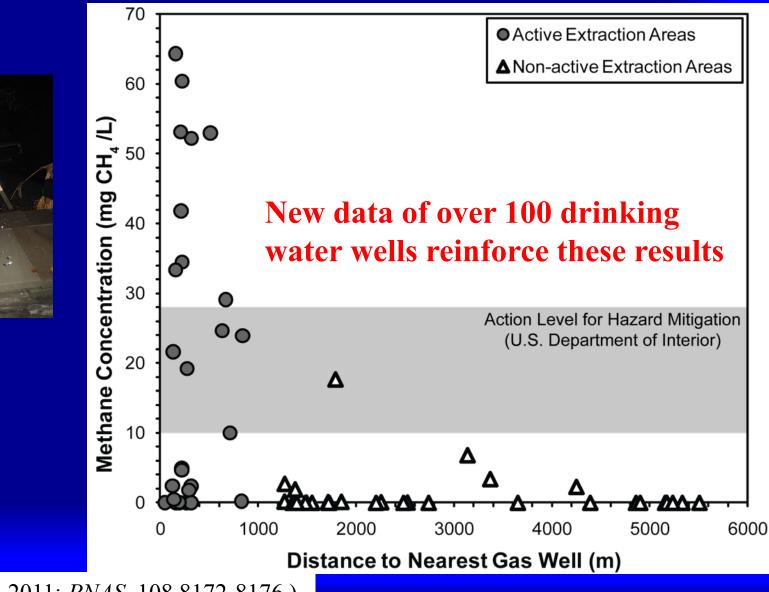
^{*}This Direct Submission article had a prearranged editor.

Hydro-geological cross section

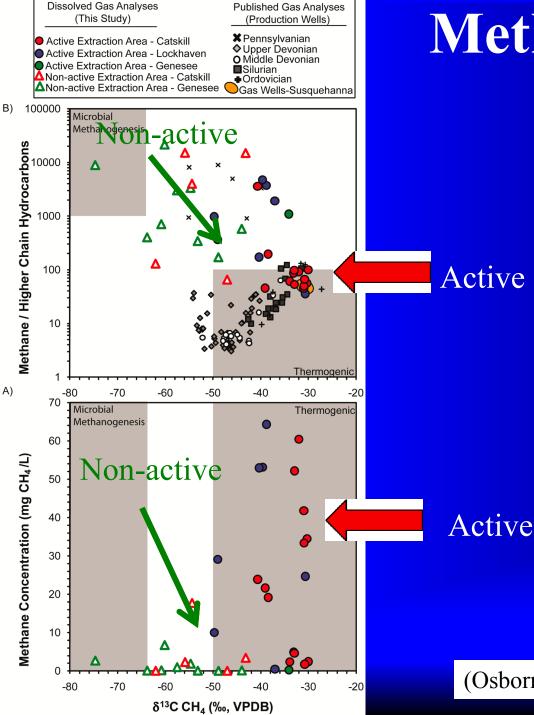


Definition of active versus non-active wells: Private wells located <1km from a shale gas had typically higher methane





(Osborn et al., 2011; *PNAS*, 108,8172-8176)



Methane sources?

A distinction between active wells with a thermogenic isotopic fingerprint and nonactive wells with a mixed composition

(Osborn et al., 2011; PNAS, 108,8172-8176)

Possible mechanisms for leakage of stay gas to water resources

Risks to Drinking Water

Chemical-laden wastewater ponds can leak or overflow (center), nect to natural fissures or old wells (inset, right),

Once a drill pad and wastewater pond are established, a driller may which happened in Pennsylvania in September because of flooding sink a dozen wells or more to fully tap the shale gas. Three spots by Tropical Storm Lee. Concrete that encases the vertical pipe can may have the greatest potential to contaminate groundwater. crack (inset, left), and new fissures opened by the fracking can con-

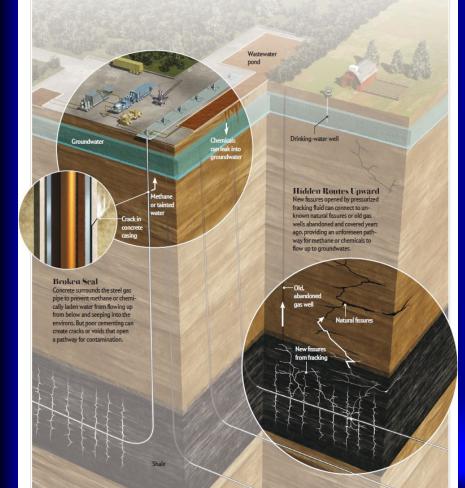
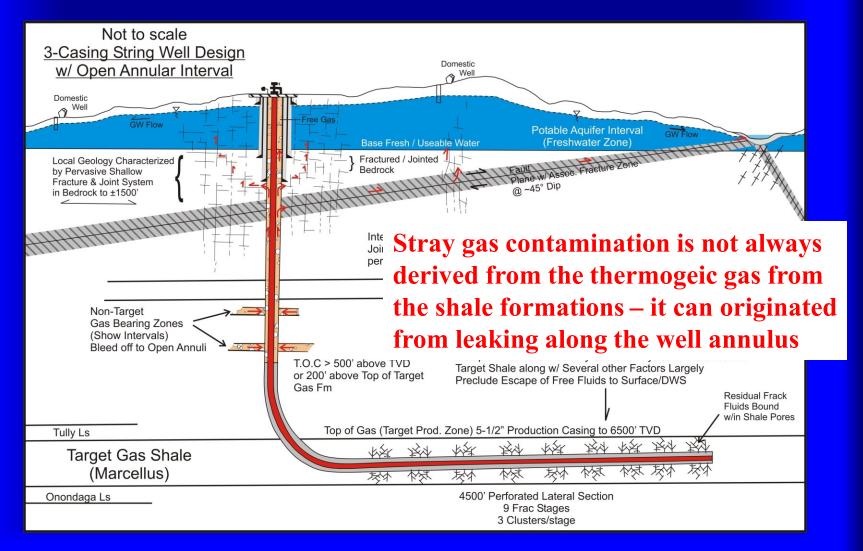


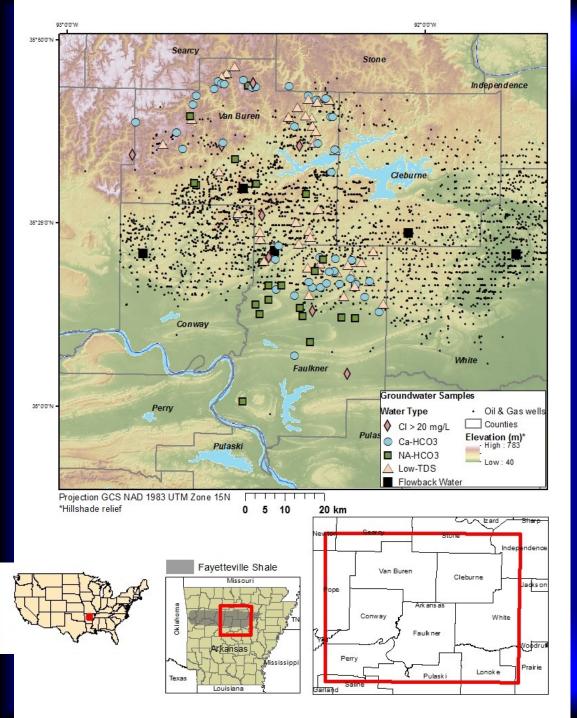


Figure from Scientific American Magazine, Nov 2011

Possible mechanisms for leakage of stay gas to water resources

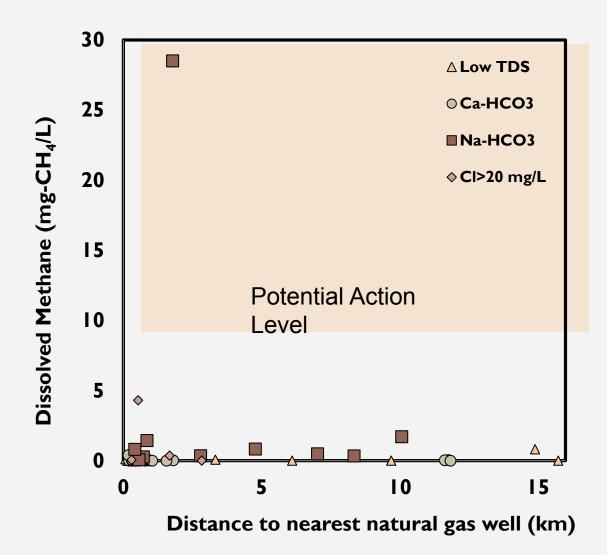


From Penoyer, (2011), Natural Resource Stewardship & Science

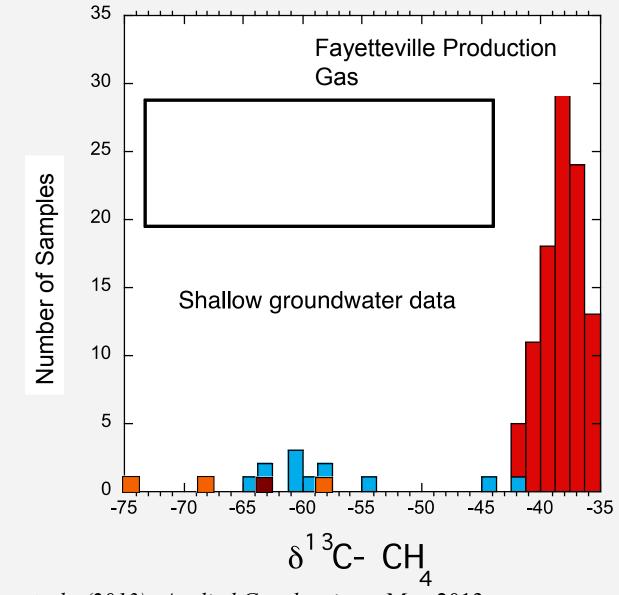


GROUNDWATER IN FAYETTEVILLE SHALE NORTH-CENTRAL ARKANSAS

Warner et al., (2013); Applied Geochemistry, May 2013



Warner et al., (2013); Applied Geochemistry, May 2013



Warner et al., (2013); Applied Geochemistry, May 2013

Stray gas contamination- conclusions

- Methane is indeed ubiquitous in groundwater in some areas overlying shale plays (e.g., Marcellus);
- Geochemical and isotopic evidence for stray gas contamination in a subset of wells near shale gas drilling sites in northeastern PA but not in AK;
- Stray gas contamination can result from leaking of natural gas along the well annulus from shallower formations and/or the the target formation through poorly constructed or failing well casings.



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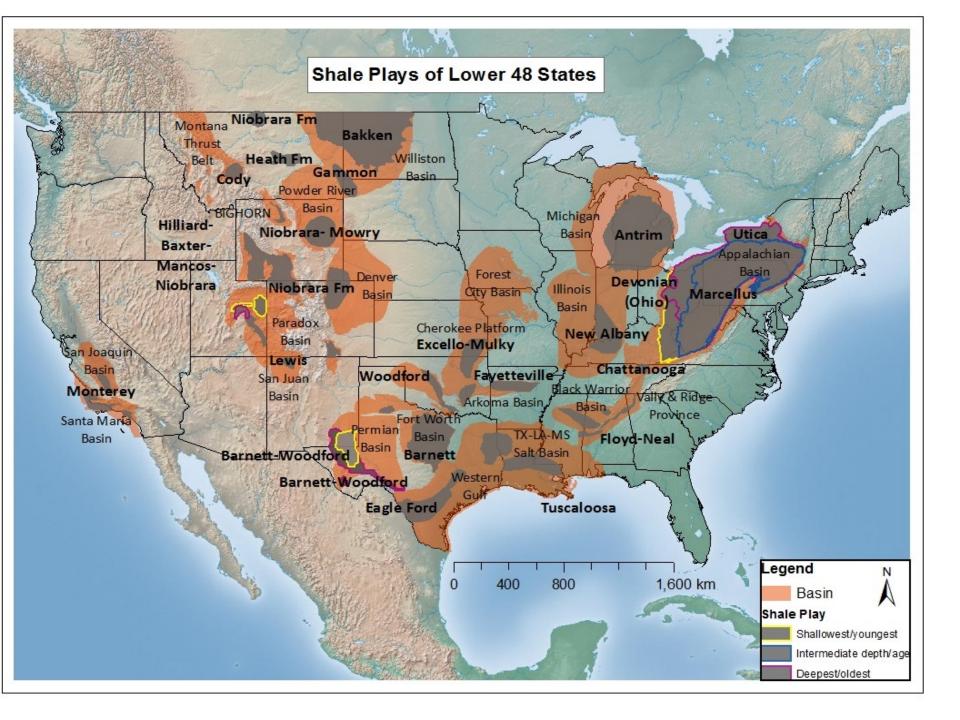
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Disposal of inadequately treated shale gas wastewater: contamination of waterways





Shale gas water footprint

Hydraulic fracturing requires large quantities of fracturing fluid

- Marcellus: 12–19 million liter (ML) per well;
- Oklahoma: 11.3 ML
- Marcellus shale gas well generates on average 5.2 ML of wastewater (12% drilling fluids, 32% flowback; 55% brine)
- Total Marcellus wastewater production in 2011 was 3144 ML (3.14x10⁶ m³) relative to ~800 ML from conventional oil and gas wells. 1200 ML was disposed at treatment facilities.

Sources: Lutz et al., (2013) WRR, 49, 647-656

What's in shale gas wastewater?

• Salinity (Marcellus brine – 250,000 mg/L ; 10 fold seawater);

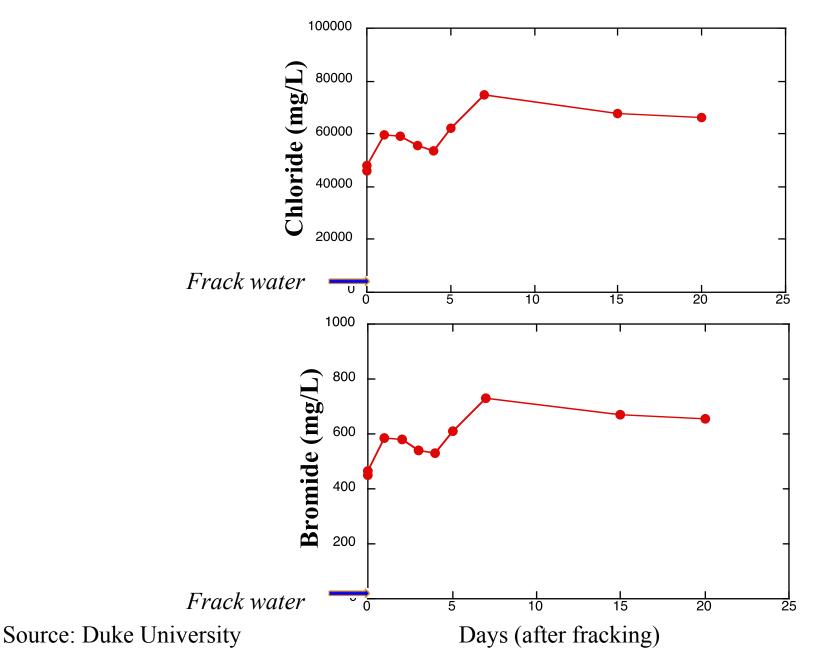
High bromide, bromide presence in water enhances the formation of carcinogenic disinfection by-products (e.g., bromodichloromethane) upon chlorination of downstream potable water;

• High concentrations of toxic elements (barium, arsenic, selenium, lead);

• High concentrations of naturally occurring radioactive materials (NORMs); (**5000 pCi/L**, drinking water standard=5 pCi/L)

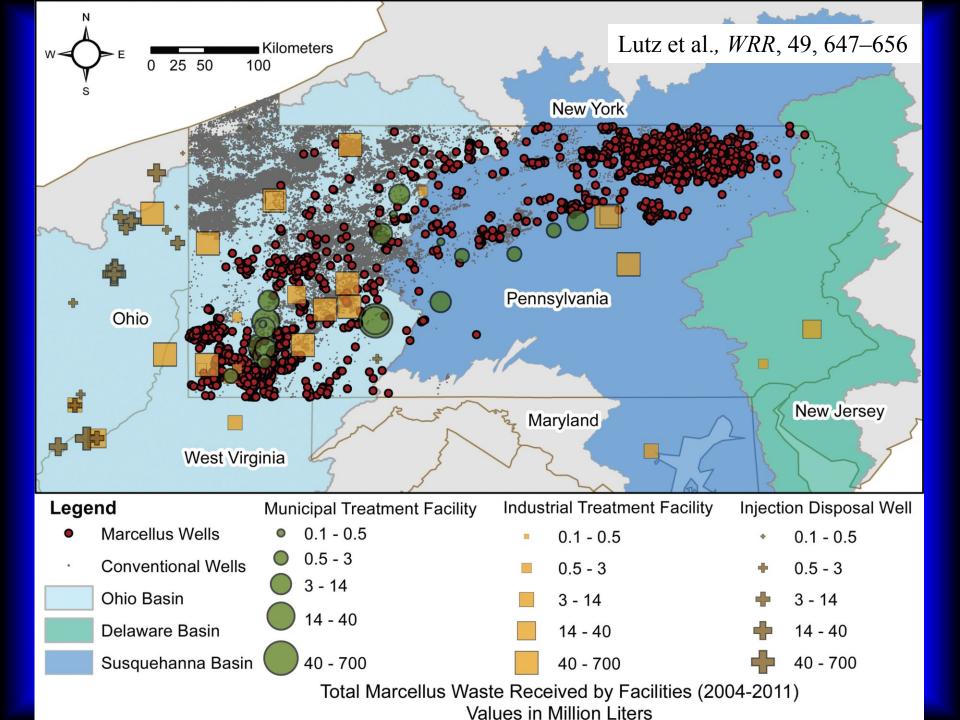
•Hydrocarbon residuals, oil, organics

Flowback from the Marcellus gas well

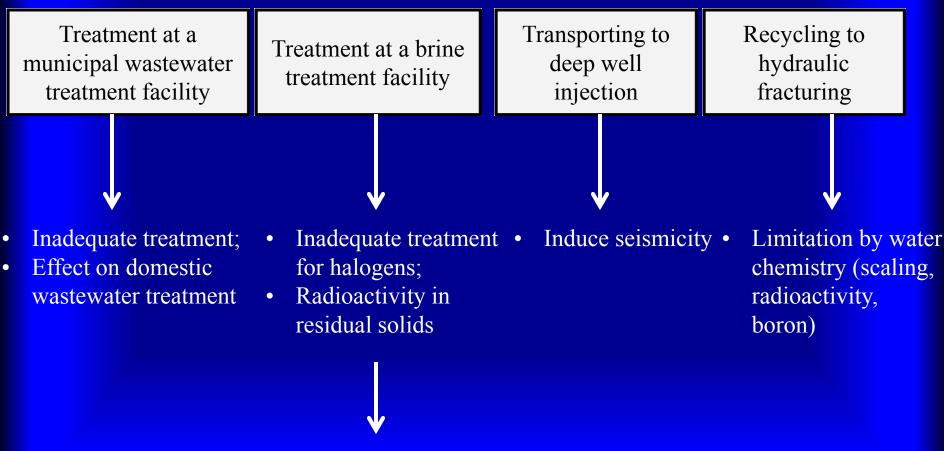


Wastewater management

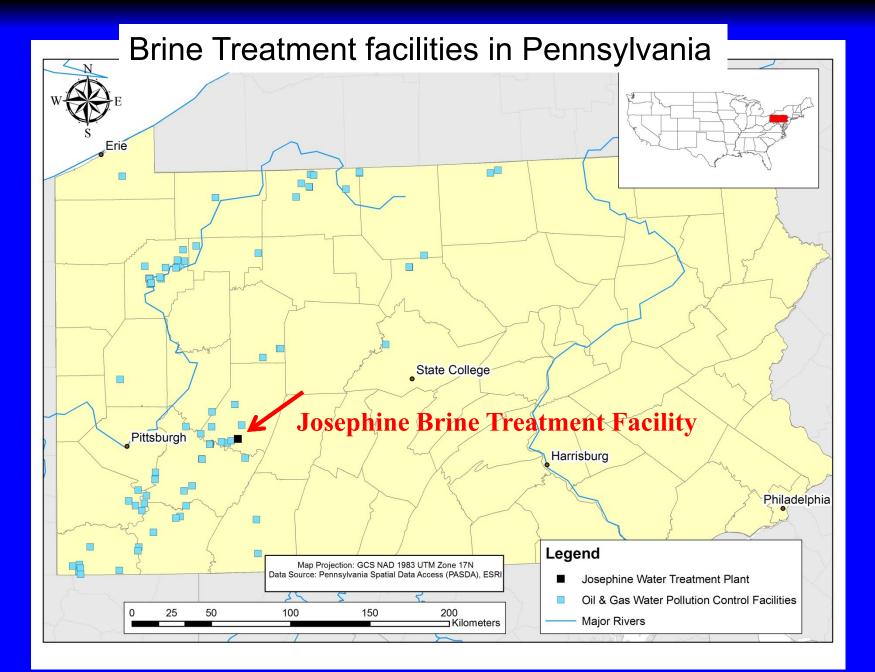
- Treatment at a municipal wastewater treatment facility followed by discharge to a local waterway;
- Treatment at a private industrial wastewater facility followed by discharge into a local waterway;
- Transporting to underground injection well site;
- Recycling to hydraulic fracturing (~70% in 2011 for Marcellus);
- Road spreading of brines for ice and dust control (currently not permitted in PA).



Short-term risks for wastewater management options



Josephine Brine Treatment Facility



A schematic illustration of the impact of a brine treatment facility

Shale gas waste water (high Cl, Na, Br, Ba, **Ra**)

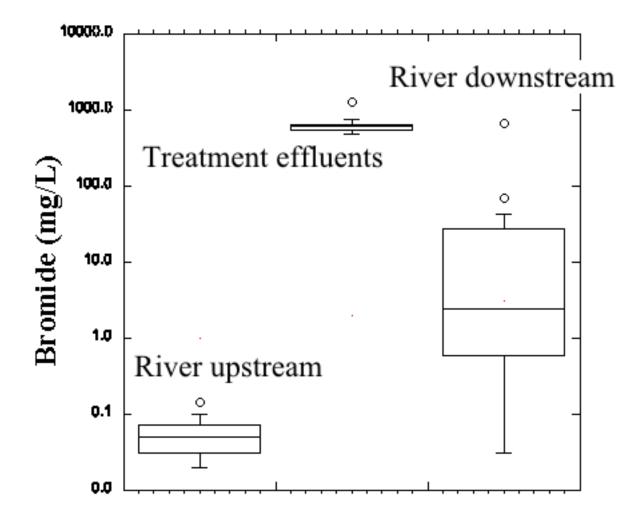
Brine Treatment Facility

Wastewater treatment does not remove all contaminants

Chloride flux (2010-2011) = 32x10³ and 143x10 tons/year for PA Treated waste water (high Cl, Na, Br)

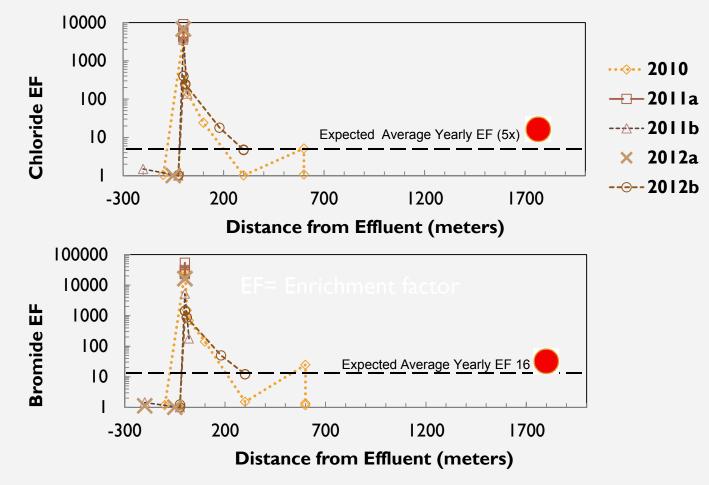
Salinity and high bromide in surface water

Source: Warner (2013) PhD thesis, Duke University



Source: Wa

Enrichment factor of halogens in downstream river water



EF= enrichment factor relative to upstream river

Source: Warner (2013) PhD thesis, Duke University

Surface water contamination via disposal of inadequately treated wastewater - conclusions

- Local contamination of streams and rivers;
- Despite of the dilution, downstream river contains higher Br than background levels → risk of formation of carcinogenic disinfection by-products upon chlorination of downstream potable water;
- Zero discharge policy is required.

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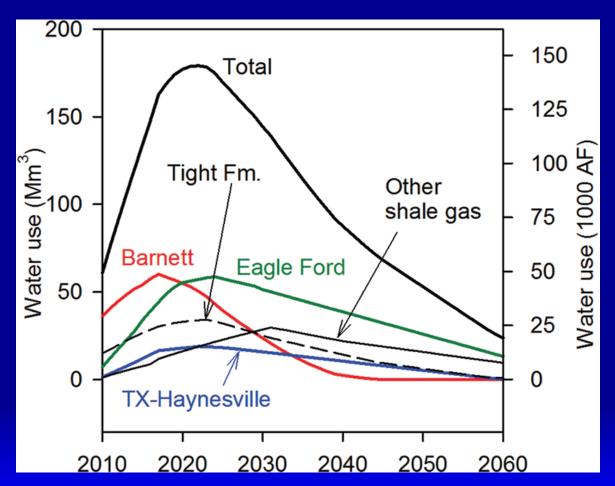
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Hydraulic fracturing requires large quantities of fracturing fluid

- Marcellus: 12–19 million liter (ML) per well;
- Oklahoma: 11.3 ML per well;
- Barnett Shale: 10.6 ML per well;
- Total water use for shale gas:
 - Marcellus (PA): \sim 42-66x10⁶ m³ (2011)
 - Oklahoma State wide: $16x10^6 \text{ m}^3(2011) \rightarrow 1\%$ of statewide fresh water use
 - Barnett Shale, TX: $\sim 30 \times 10^6 \text{ m}^3 \rightarrow 7\%$ of Dallas water use

Sources: Lutz et al., *(2013) WRR*, 49, 647–656; Murray, (2013); *ES&T*, 47, 4918–4925; Nicot and Scalon, (2012), *ES&T*

Shale gas water footprint



Time evolution in Texas of fracking net water use distributed among the Barnett, Tx-Haynesville, Eagle Ford, and other shale-gas plays

Sources: Nicot and Scalon, (2012), ES&T

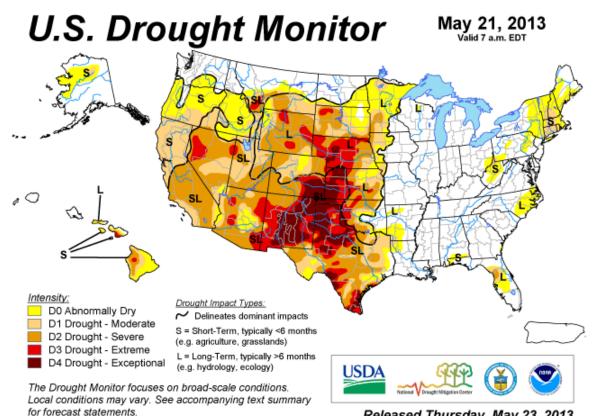
Overall water footprint

- Shale gas water footprint a few % of total freshwater withdrawal;
- Thermoelectric withdrawal (2005) 142 Bgal/day* (196 x 10⁹ m³/year) ~ 40% of total freshwater withdrawal in the USA.

*Source: Kenny et al. (2009), U.S. Geological Survey Circular 1344, 52 p.

Shale gas water footprint

Although the overall water use for shale gas and hydraulic fracturing is low in comparison to other users, in some water-scare areas, such as in TX, water use for shale gas constitutes a large fraction of groundwater resources, that could lead to potential water shortage.



Released Thursday, May 23, 2013 Author: Brad Rippey, U.S. Department of Agriculture

http://droughtmonitor.unl.edu/

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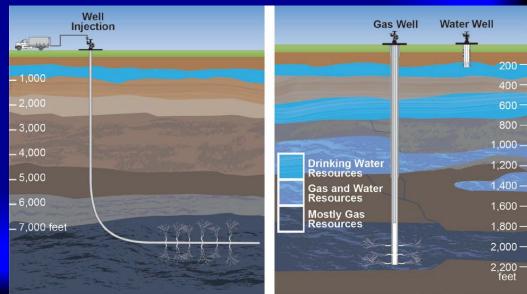
• Water availability in water scarce areas;

Long term

- Groundwater contamination through natural fracture networks;
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The long-term risk: Groundwater contamination through natural fracture networks

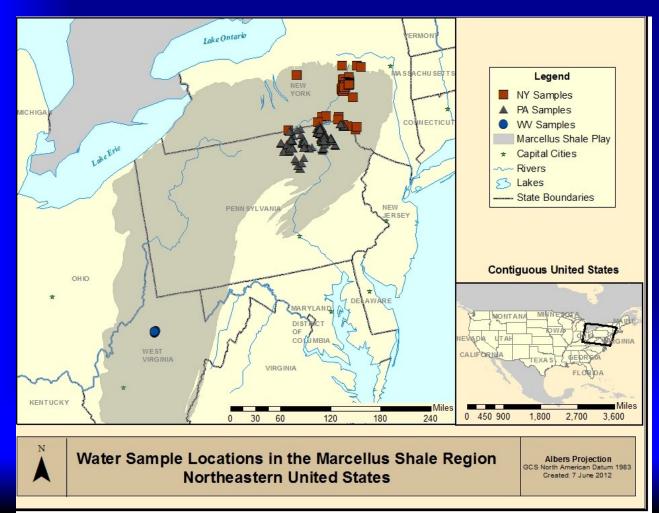
Modeling simulation: Advective transport of saline water through faults and fracture system could reach the overlying aquifers in less than 10 years



Myers (2012), Groundwater, 50,872-882

Duke Study:

Evidence for hydraulic connectivity – deep brine/gas can flow to shallow aquifers in PA



Geochemical evidence for possible natural migration of Marcellus Formation brine to shallow aquifers in Pennsylvania

Nathaniel R. Warner^a, Robert B. Jackson^{a,b}, Thomas H. Darrah^a, Stephen G. Osborn^c, Adrian Down^b, Kaiguang Zhao^b, Alissa White^a, and Avner Vengosh^{a,1}

"Division of Earth and Ocean Sciences, Nicholas School of the Environment, Duke University, Durham, NC 27708; "Center on Global Change, Nicholas School of the Environment, Duke University, Durham, NC 27708; "Geological Sciences Department, California State Polytechnic University, Pomona, CA 91768

Edited by Karl K. Turekian, Yale University, North Haven, CT, and approved May 10, 2012 (received for review January 5, 2012)

The debate surrounding the safety of shale gas development in the Appalachian Basin has generated increased awareness of drinking water quality in rural communities. Concerns include the potential for migration of stray gas, metal-rich formation brines, and hydraulic fracturing and/or flowback fluids to drinking water aguifers. A critical guestion common to these environmental risks is the hydraulic connectivity between the shale gas formations and the overlying shallow drinking water aquifers. We present geochemical evidence from northeastern Pennsylvania showing that pathways, unrelated to recent drilling activities, exist in some locations between deep underlying formations and shallow drinking water aguifers. Integration of chemical data (Br. Cl. Na. Ba, Sr. and Li) and isotopic ratios (⁸⁷Sr/⁸⁶Sr, ²H/H, ¹⁸O/¹⁶O, and ²²⁸Ra/²²⁶Ra) from this and previous studies in 426 shallow groundwater samples and 83 northern Appalachian brine samples suggest that mixing relationships between shallow ground water and a deep formation brine causes groundwater salinization in some locations. The strong geochemical fingerprint in the salinized (Cl > 20 mg/L) groundwater sampled from the Alluvium, Catskill, and Lock Haven aquifers suggests possible migration of Marcellus brine through naturally occurring pathways. The occurrences of saline water do not correlate with the location of shale-gas wells and are consistent with reported data before rapid shale-gas development in the region; however, the presence of these fluids suggests conductive pathways and specific geostructural and/or hydrodynamic regimes in northeastern Pennsylvania that are at increased risk for contamination of shallow drinking water resources, particularly by fugitive gases, because of natural hydraulic connections to deeper formations.

O:1 formation water | isotopes | Marcellus Shale | water chemistry

The extraction of natural gas resources from the Marcellus Shale in the Appalachian Basin of the northeastern United States (1, 2) has increased awareness of potential contamination in shallow aquifers routinely used for drinking water. The current debate surrounding the safety of shale gas extraction (3) has focused on stray gas migration to shallow groundwater (4) and the atmosphere (5) as well as the potential for contamination from toxic substances in hydraulic fracturing fluid and/or produced brines during drilling, transport, and disposal (6–9).

The potential for shallow groundwater contamination caused by natural gas drilling is often dismissed because of the large vertical separation between the shallow drinking water wells and shale gas formations and the relatively narrow zone (up to 300 m) of seismic activity reported during the deep hydraulic fracturing of shale gas wells (10, 11). Recent findings in northeastern Pennsylvania (NE PA) demonstrated that shallow water wells in close proximity to natural gas wells (i.e., <1 km) yielded, on average, higher concentrations of methane, ethane, and propane with thermogenic isotopic signature. By comparison, water wells farther away from natural gas development had lower combustible gas concentrations and an isotopic signature consistent with a mixture between thermogenic and biogenic components (4). In contrast, when inorganic water geochemistry from active drilling areas was compared to nonactive areas and historical background values, no statistically significant differences were observed (4). Increasing reports of changes in drinking water quality have nevertheless been blamed on the accelerated rate of shale gas development.

/IRONMENT/ SCIENCES

The study area in NE PA consists of six counties (Fig. 1) that lie within the Appalachian Plateaus physiographic province in the structurally and tectonically complex transition between the highly deformed Valley and Ridge Province and the less deformed Appalachian Plateau (12, 13). The geologic setting and shallow aquifer characteristics are described and mapped in greater detail in multiple sources (4, 14-19) and in SI Methods. The study area contains a surficial cover composed of a mix of unconsolidated glacial till, outwash, alluvium and deltaic sediments, and postglacial deposits (the Alluvium aquifer) that are thicker in the valleys (17-19) (Fig. S1). These sediments are underlain by Upper Devonian through Pennsylvanian age sedimentary sequences that are gently folded and dip shallowly $(1-3^{\circ})$ to the east and south (Fig. S2). The gentle folding creates alternating exposure of synclines and anticlines at the surface that are offset surface expressions of deeper deformation (12, 20). The two major bedrock aquifers are the Upper Devonian Catskill and the underlying Lock Haven Formations (14, 15, 18, 19). The average depth of drinking water wells in the study area is between 60 and 90 m (Table S1). The underlying geological formations, including the Marcellus Shale (at a depth of 1,200–2,500 m below the surface) are presented in Fig. 2, Fig. S2 A and B, and SI Methods.

In this study, we analyze the geochemistry of 109 newly-collected water samples and 49 wells from our previous study (4) from the three principal aquifers, Alluvium (n = 11), Catskill (n = 102), and Lock Haven (n = 45), categorizing these waters into four types based on their salinity and chemical constituents (Figs. 1 and 2, and *S1 Text*). We combine these data with 268 previously-published data for wells in the Alluvium (n = 57), Catskill (n = 147), and Lock Haven (n = 64) aquifers (18, 19) for a total of 426 shallow groundwater samples. We analyzed major and trace element geochemistry and a broad spectrum of isotopic tracers (δ^{180} , δ^2 H, 87 Sr/ 86 Sr, 228 Ra/ 226 Ra) in shallow Warner et al., 2012 Proceedings of National Academy of Sciences, July 9, 2012

158 wells - new measurements

268 wells – previously published data

Author contributions: N.R.W., R.B.J., and A.V. designed research; N.R.W., R.B.J., S.G.O., A.D., A.W., and A.V. performed research; N.R.W., R.B.J., T.H.D., K.Z., and A.V. analyzed data; and N.R.W., R.B.J., T.H.D., and A.V. wrote the paper.

The authors declare no conflict of interest

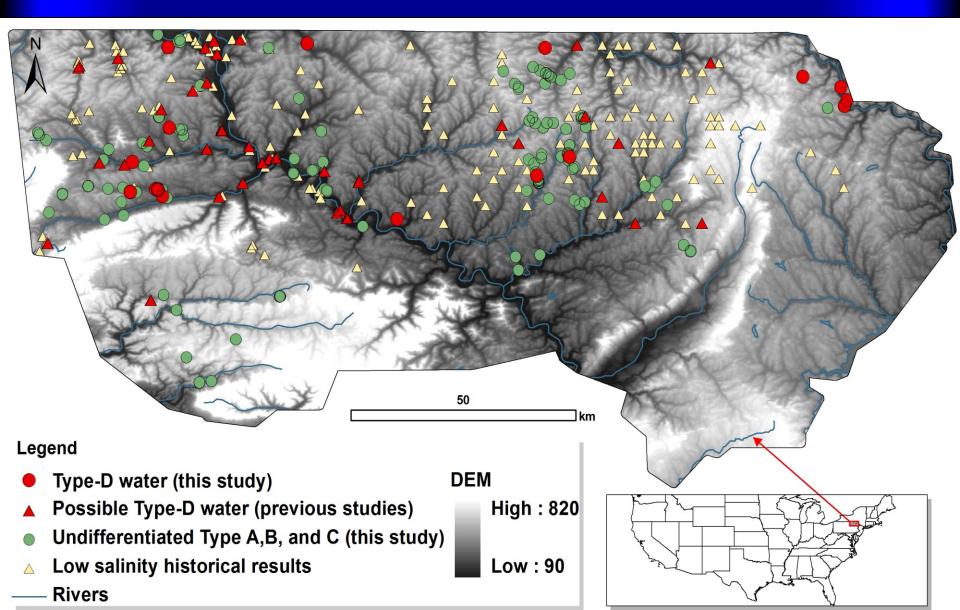
This article is a PNAS Direct Submission.

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¹To whom correspondence should be addressed. E-mail: vengosh@duke.edu.

S This article contains supporting information online at www.pnas.org/lookup/suppl/ doi:10.1073/pnas.1121181109/-/DCSupplemental.

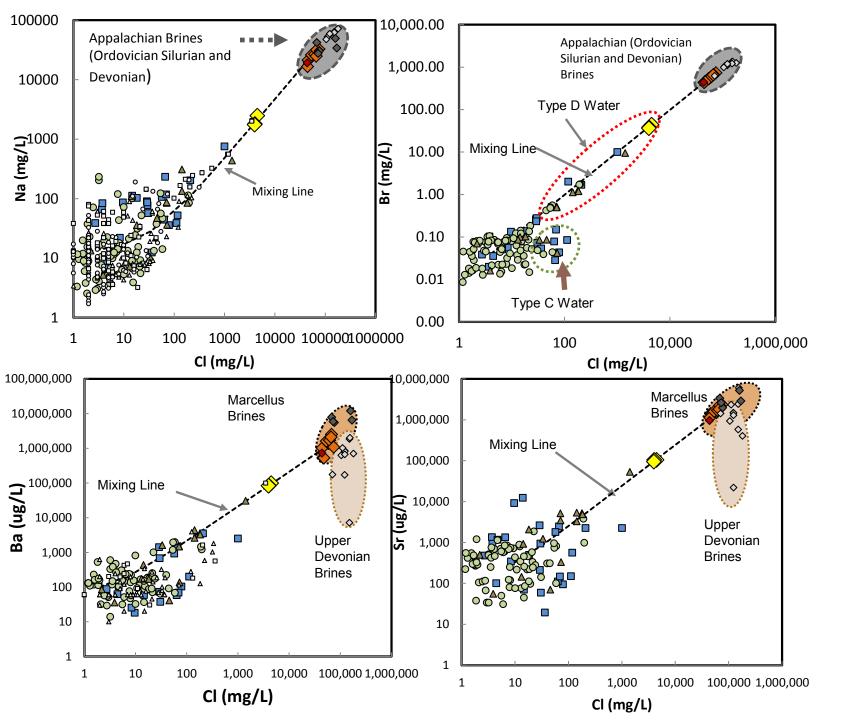
Occurrence of saline groundwater enriched in barium in shallow aquifers



Salt Springs Park, Susquehanna County, Pennsylvania



TDS = 7,000 mg/L; CH_4 - over-saturation Ca-Na-Cl composition; high Br/Cl



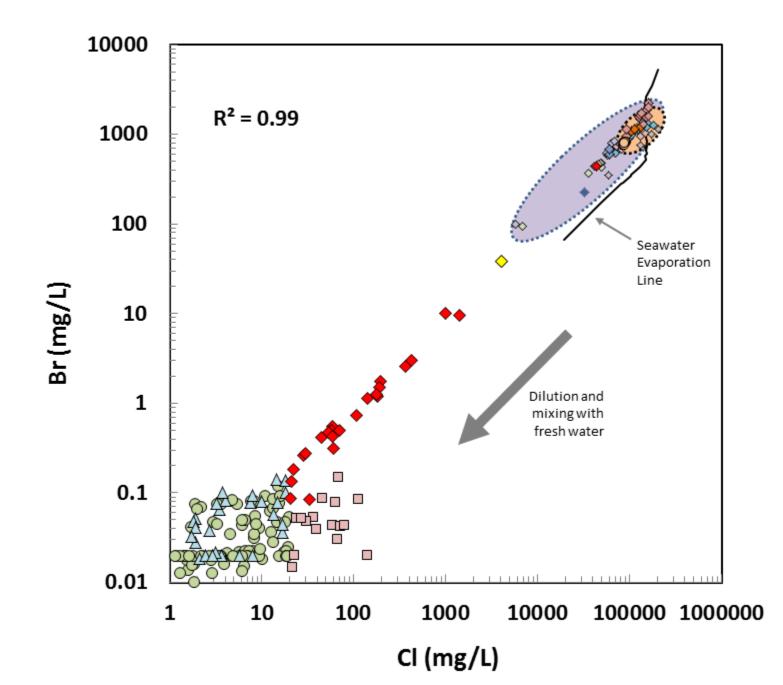


Figure 3.

Western PA			Eastern PA Plateau		⁸⁷ Sr/ ⁸⁶ Sr	Age
Conemough Gp Allegheny Gp Burgoon Fm			A	llegheny and Pottsville Groups	0 0 0 0 7 7 0 0 0 7 7	Pennsylvaniar
			Huntley Mtn, Pocono, and Mauch Chunk Fms		0.706 0.708 0.712 0.714 0.714 0.716 0.716 0.716 0.718 0.718 0.718 0.720 0.722 0.722	Mississippian
Venango Gp Bradford Gp Elk Gp			Catskill Fm Duncannon Mbr Sherman Mbr Lock Haven Fm			Upper Devonian
Brallier Fm Hamilton Mahantango Fm			Brallier Fm Hamilton Mahantango Fm			
Gp Onondaga Gr	— Ma	rcellus Fm	Gp Onondaga G	Marcellus Fm — —	rceiius Bradford SS	Middle Devonian
Ridgeley Ss Helderberg Gp Bass Islands Dol – Keyser Fm			<u>Ridgeley Ss</u> Helderberg Gp Keyser Fm		Organic Rich Shales	Lower Devonian
Salinas G _l	Salinas Gp Wills Creek Fm		Salinas Gp Tonoloway Fm Wills Creek Fm Bloomsburg Fm		Variation in Paleozoic	Upper Silurian
Gp McKenz		kport Dol – e/Rochester Fm	Clinton Gp	McKenzie/Mifflintown	Seawater ⁸⁷ Sr/ ⁸⁶ Sr	Middle Silurian
Medina Gp	Rose Hill Fm Medina Gp Tuscarora Fm		Rose Hill Fm Tuscarora Fm		Medina	Lower Silurian
Queenston Fm Oswego Fm I Reedsville Sh		Juniata Fm Bald Eagle Fm Utic <u>a Sh</u>	Reedsville	Juniata Fm Bald Eagle Fm Sh Utica Sh	Utica	Upper Ordovician

Figure 2.

Mixing with Marcellus brines

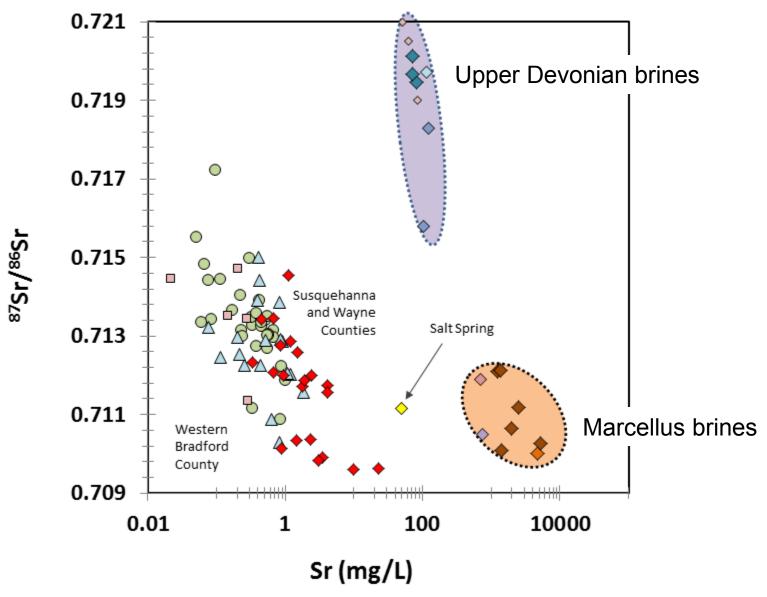
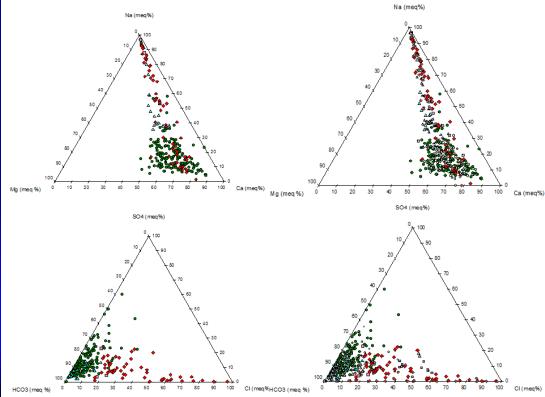


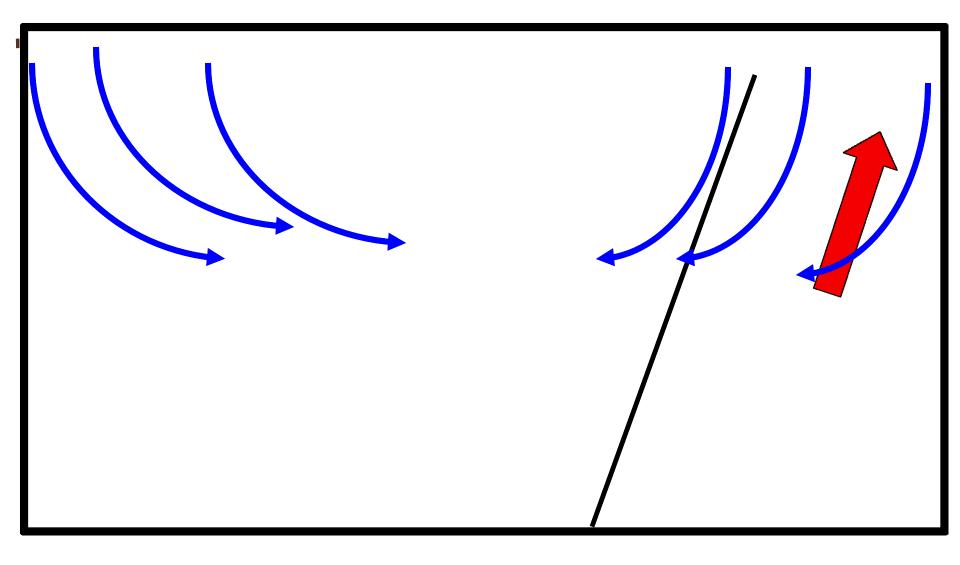
Figure 6.

No link to shale gas exploration:

- Analysis of 1980's USGS data reveals saline water of similar chemical composition (although Br data is not available)
- No geographical proximity to shale gas site (unlike the methane occurrence)
 No geographical proximity to shale gas site (unlike the methane occurrence)



Flowpaths in a differential fractured aquifer: low-saline recharged water and upflow of deep saline groundwater through fracture zones



Risks of Shale Gas Exploration and Hydraulic Fracturing to Water *Resources in the United States*

Short term

- Stray gas contamination;
- Surface water contamination via disposal of inadequately treated wastewater;
- Spills;

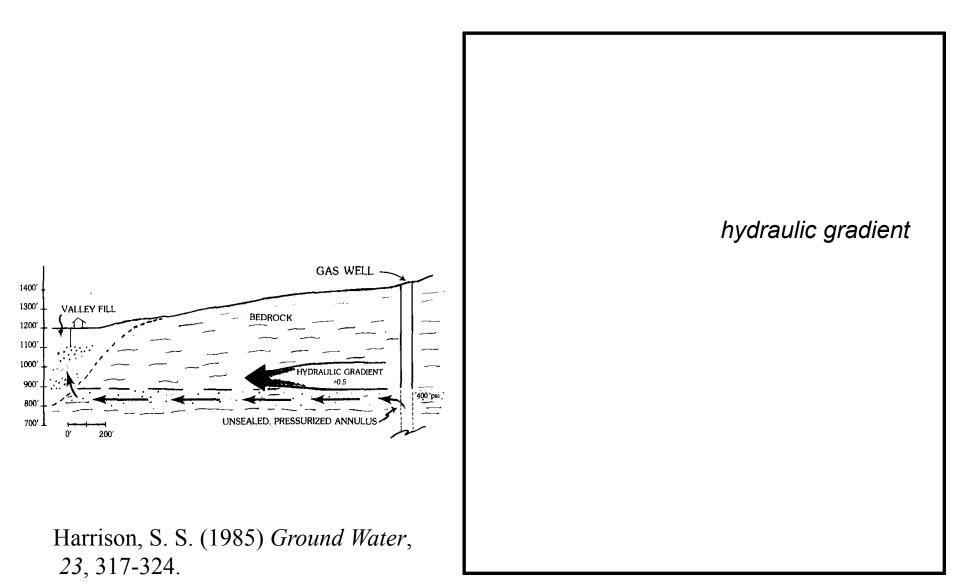


• Water availability in water scarce areas;

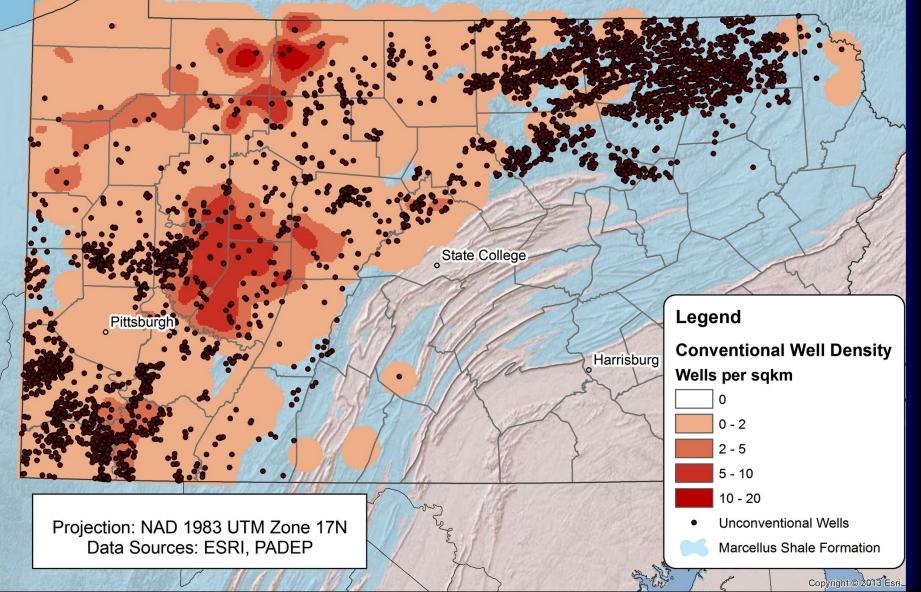
Long term

- Groundwater contamination through natural fracture networks;
- Groundwater contamination through abandoned and improperly sealed conventional oil and gas wells;
- Accumulation of residual contaminants and radiation in areas of wastewater disposal and spills;

The risk: Groundwater contamination through abandoned and improperly sealed conventional oil and gas wells



Areas of high conventional wells density → higher risks of contamination from "short cuts" related to oil and gas wells legacy



Erie

Risks of Shale Gas Exploration and Hydraulic Fracturing to Water *Resources in the United States*

Short term

- Stray gas contamination;
- Surface water contamination via disposal of inadequately treated wastewater;
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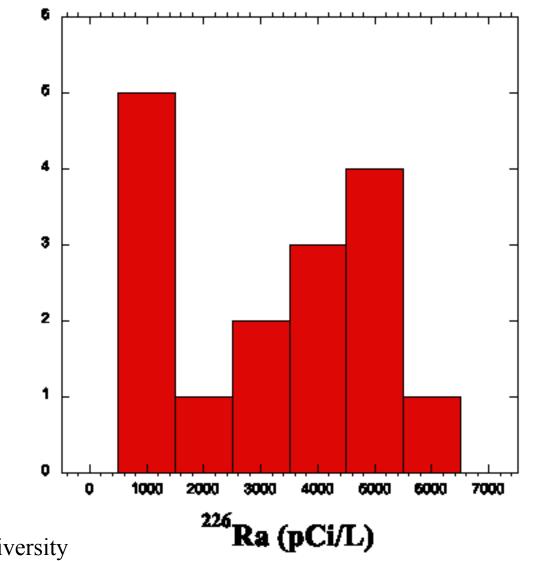
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Long term

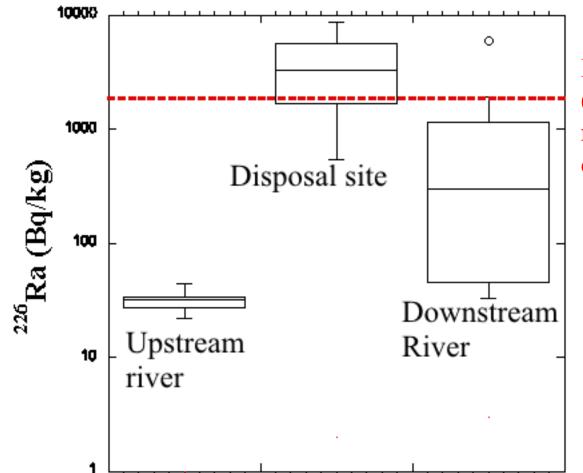
- Groundwater contamination through abandoned and improperly sealed conventional oil and gas wells;
- Accumulation of residual contaminants and radiation in areas of wastewater disposal and spills;

The risk: Accumulation of residual contaminants and radiation in areas of wastewater disposal and spills Radi

uced



Source: Duke University



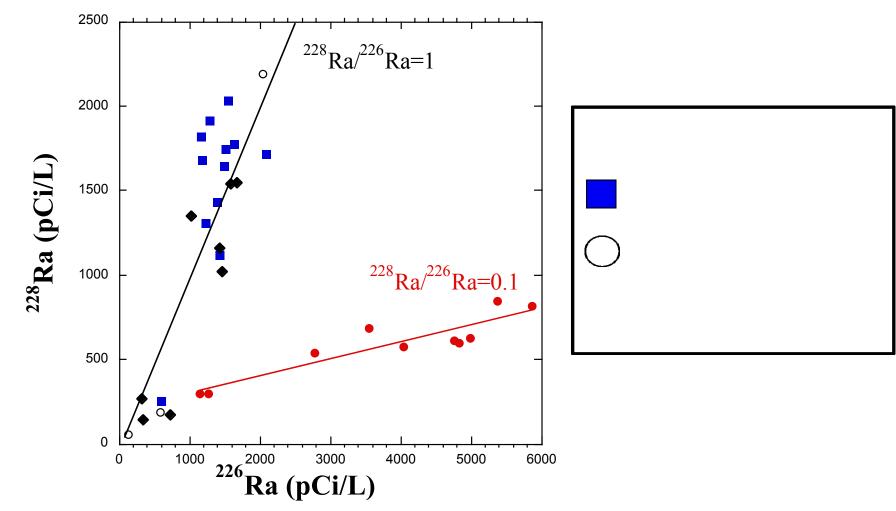
Radiation threshold (requires a licensed radioactive waste disposal facility)

a

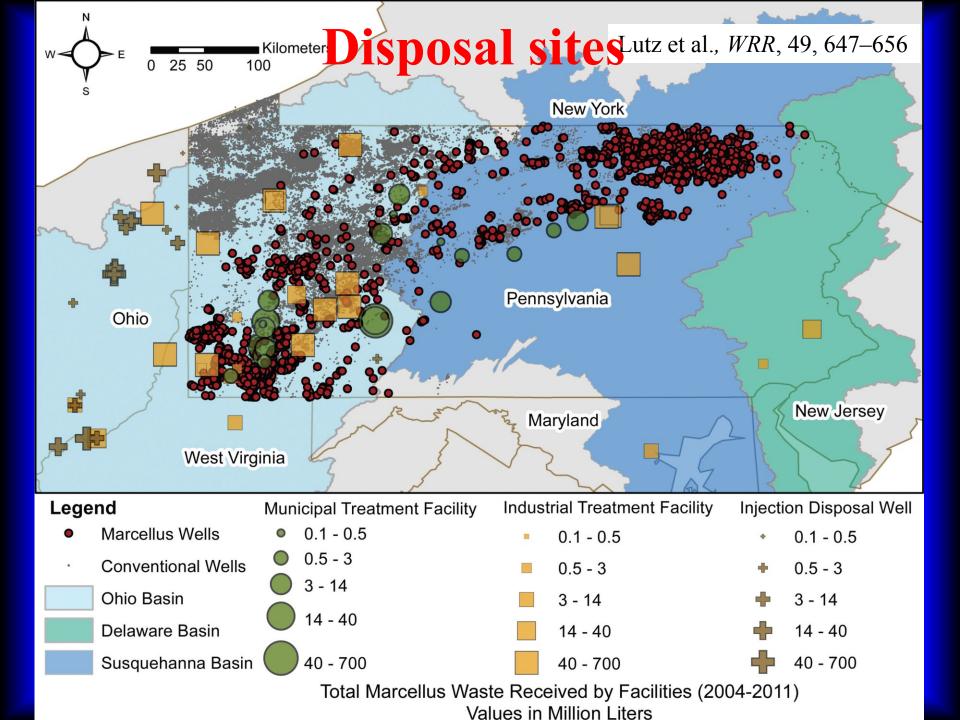
Source: Warner (2013) PhD thesis, Duke University

The used of radium isotopes

Distinction between the Marcellus brines and other (conventional) oil and gas produced waters



Source: Warner (2013) PhD thesis, Duke University



Final comment...



Our knowledge and actual data is limited. We are only at the beginning stage in evaluation of the overall impacts of shale gas development on water resources in the US.

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For more information: http://sites.nicholas.duke.edu/avnervengosh/

