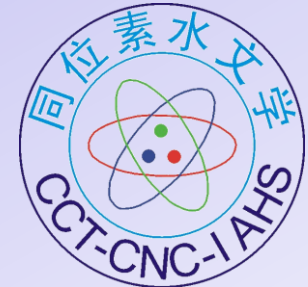


中国科学院地质与地球物理研究所
Institute of Geology and Geophysics, Chinese Academy of Sciences
中国科学院地球科学研究院
Institutions of Earth Science, Chinese Academy of Sciences



《第一届中国同位素水文学论坛》

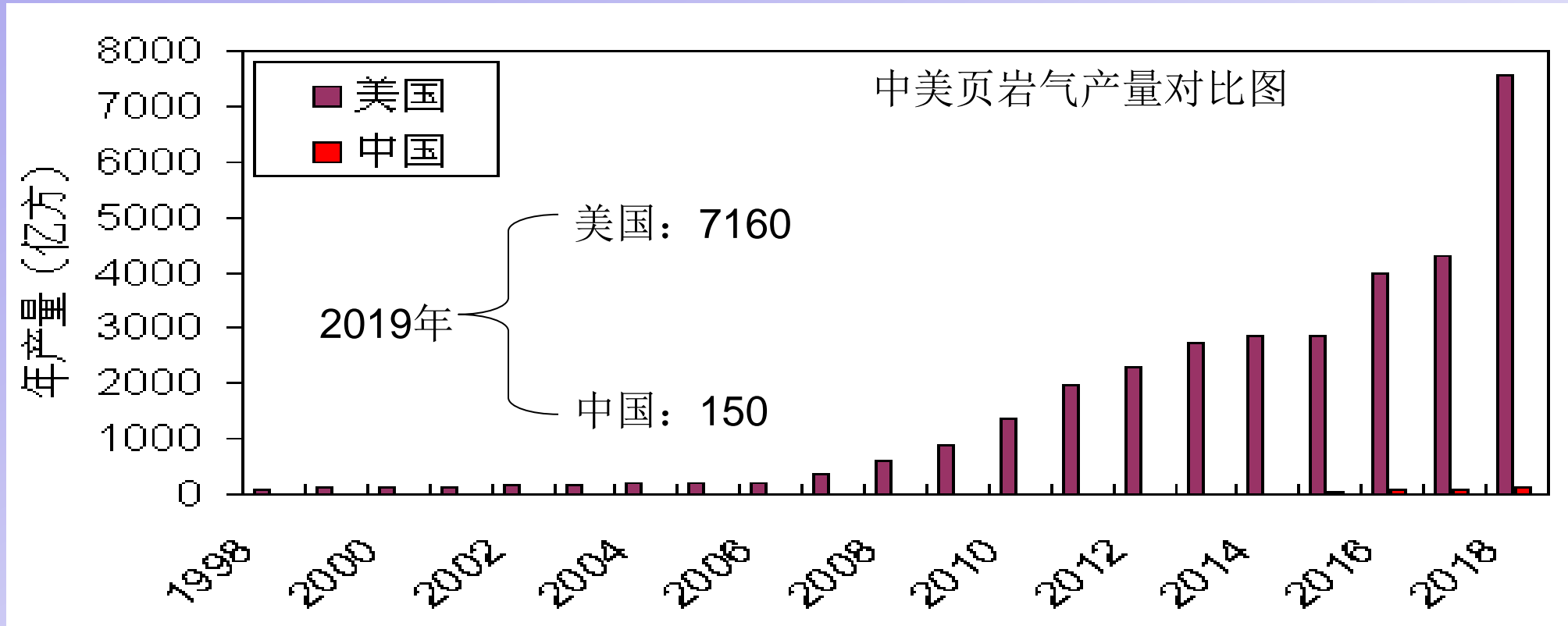
页岩气水力压裂中地球化学过程的同位素示踪

黄天明

中国科学院地质与地球物理研究所

2020年11月3日，北京

页岩气革命



- 美国历经20余年攻关，2006年**水平钻井**和**水力压裂**技术获得重大突破，使页岩气产量急剧增加，2011年成为世界第一大产气国，价格持续减低，引发了**页岩气革命**，并改变了美国能源结构，影响了世界能源格局，消除了人们对油气稀缺性的担心
- 由于美国页岩气革命的成功，中国、**英国**、波兰等国拟大力发展页岩气

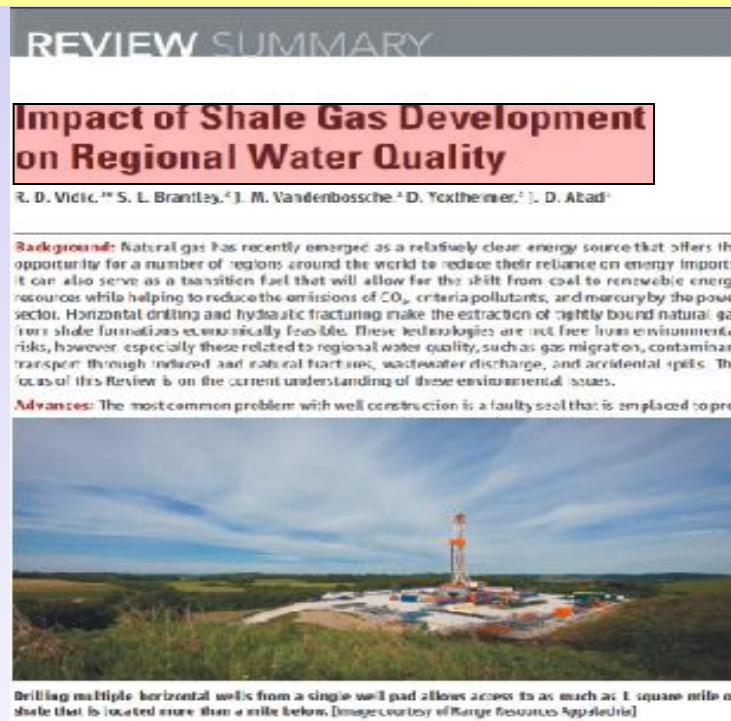
页岩气开发的环境问题

环境问题是制约页岩气开发的主要因素，由于担心**地下水污染**和**诱发地震**，一些欧洲国家（如法国、德国、保加利亚）、美国和澳大利亚一些州都**禁止水力压裂和页岩气开发**，英国2019年11月叫停水力压裂

“世界上尚无人能证明使用这种技术进行开采不会给环境和健康带来严重危害”-法国生态、可持续发展与能源部部长



Tollefson (2013, Nature)



Vidic et al. (2013, Science)

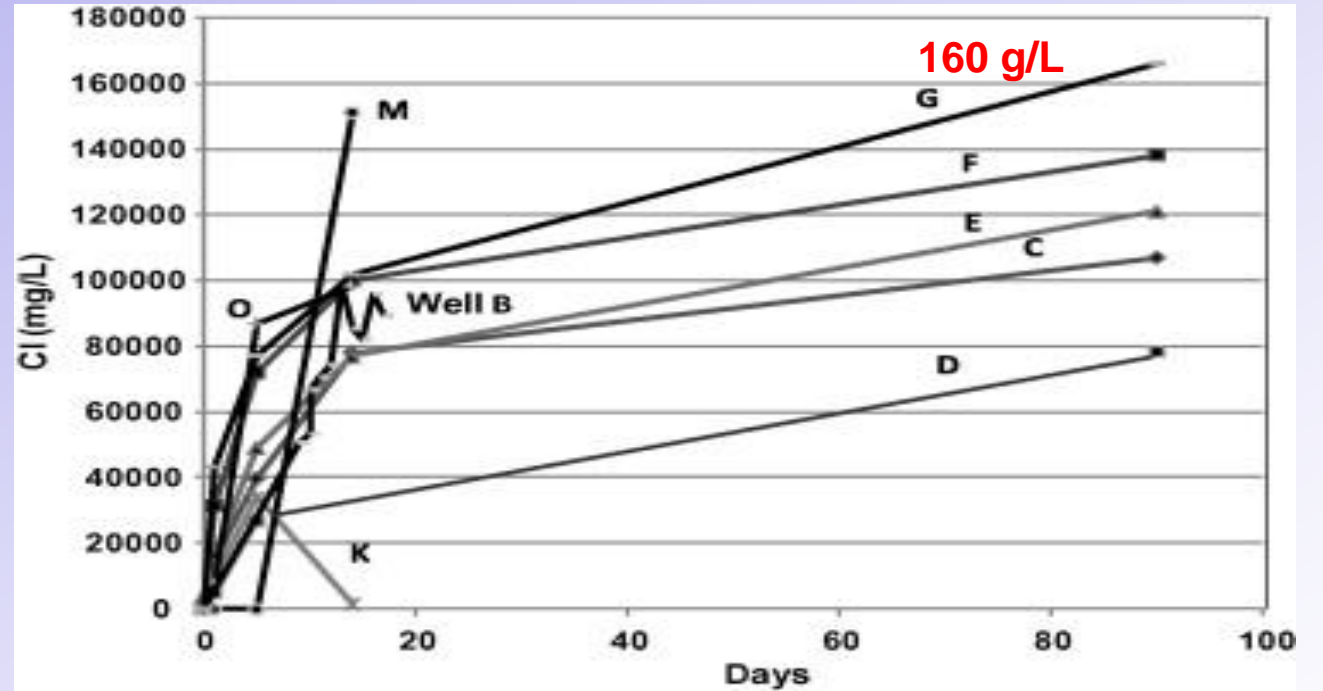
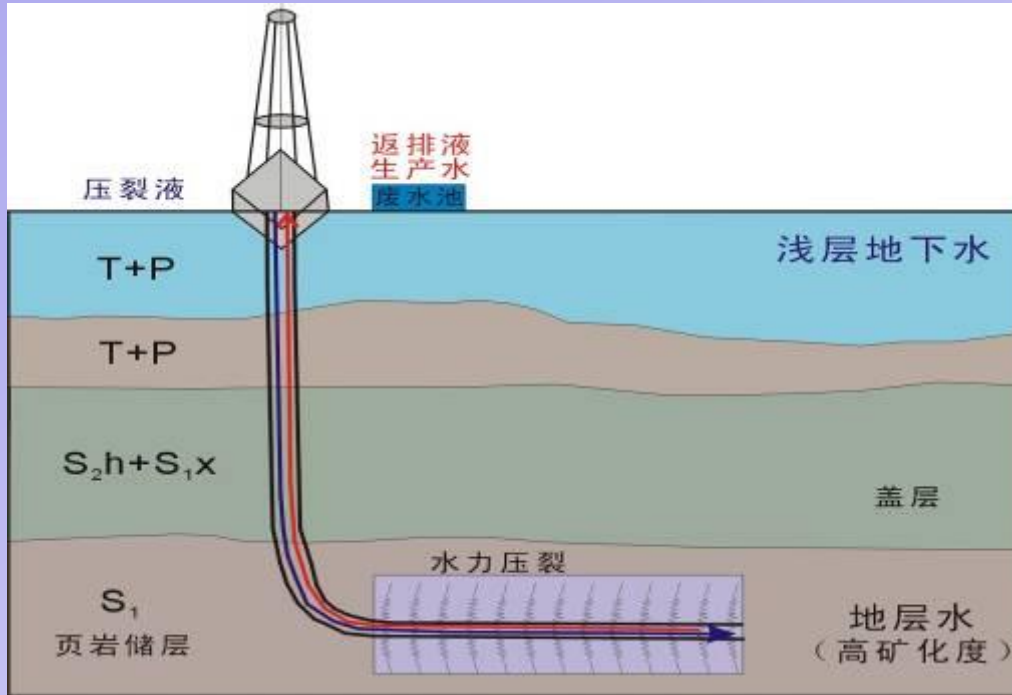
科学问题

追踪水力压裂中**水足迹**及刻画**地球化学过程**,

对于:

- 解低渗油气储层水岩相互作用
- 改善水力压裂效果等具有重要意义
- 追溯地下水污染

返排液水源组成



Haluszczak et al., 2013, AG

页岩中没有自由移动水，返排液及生产水是均是压裂液

Balashov et al.,
2015, *AAPG Bull*



页岩中存在大量地层水，返排液及生产水是两者混合，且返排后期以地层水为主

Kondash et al.,
2018, *Science Advances*

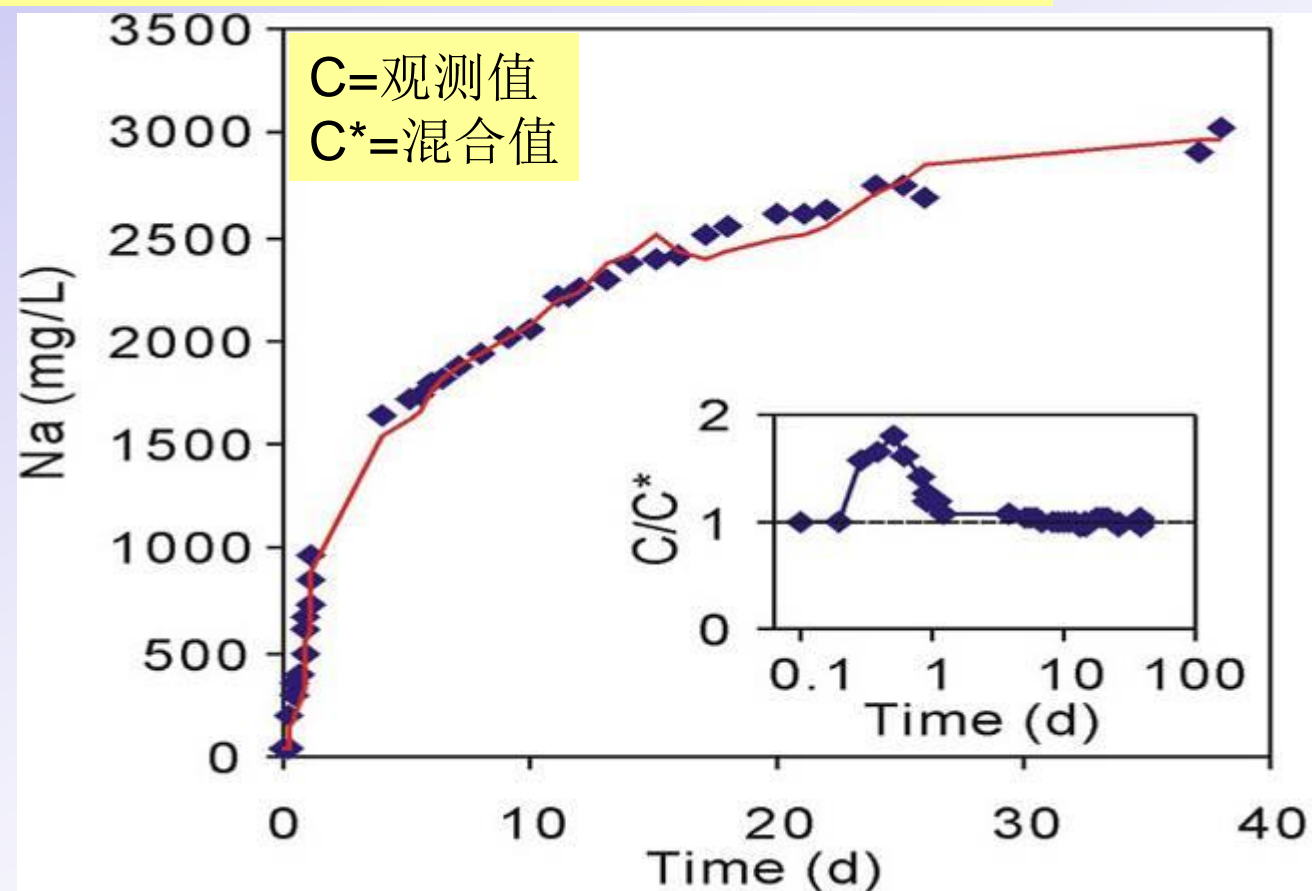
返排液溶质来源（或地球化学过程）

除了混合，还有地球化学反应

目前尚未对压裂及返排中的地球化学过程进行全面刻画，一些重要的地球化学作用（如黄铁矿氧化）是否发生也不明确

主要原因：

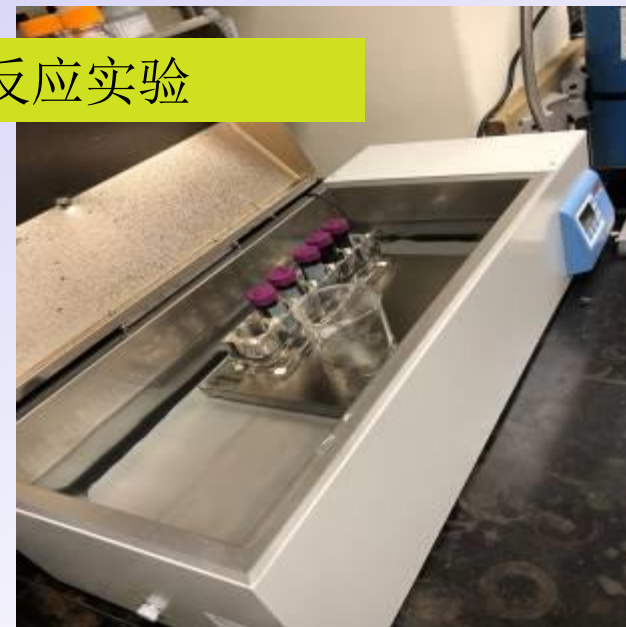
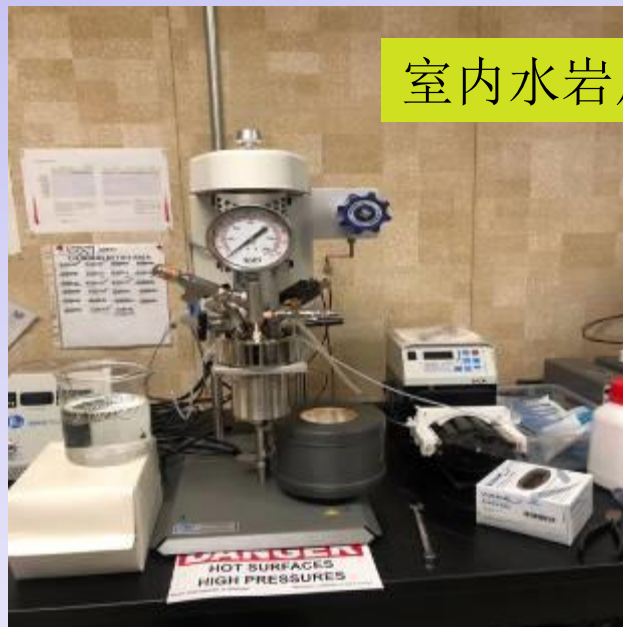
- 返排液分辨率不高不足以刻画
- 室内水岩反应实验和野外结果不匹配



野外试验

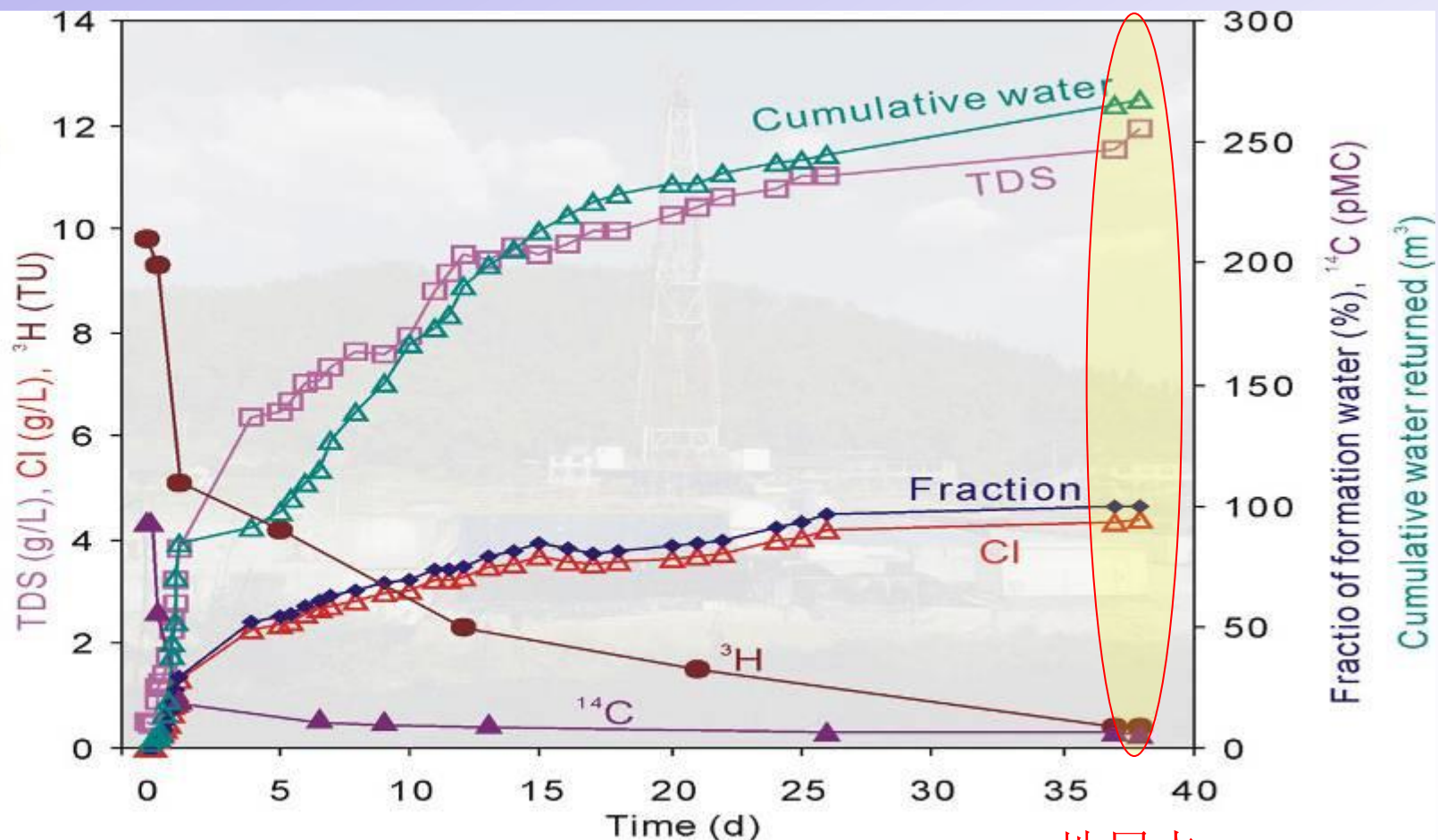
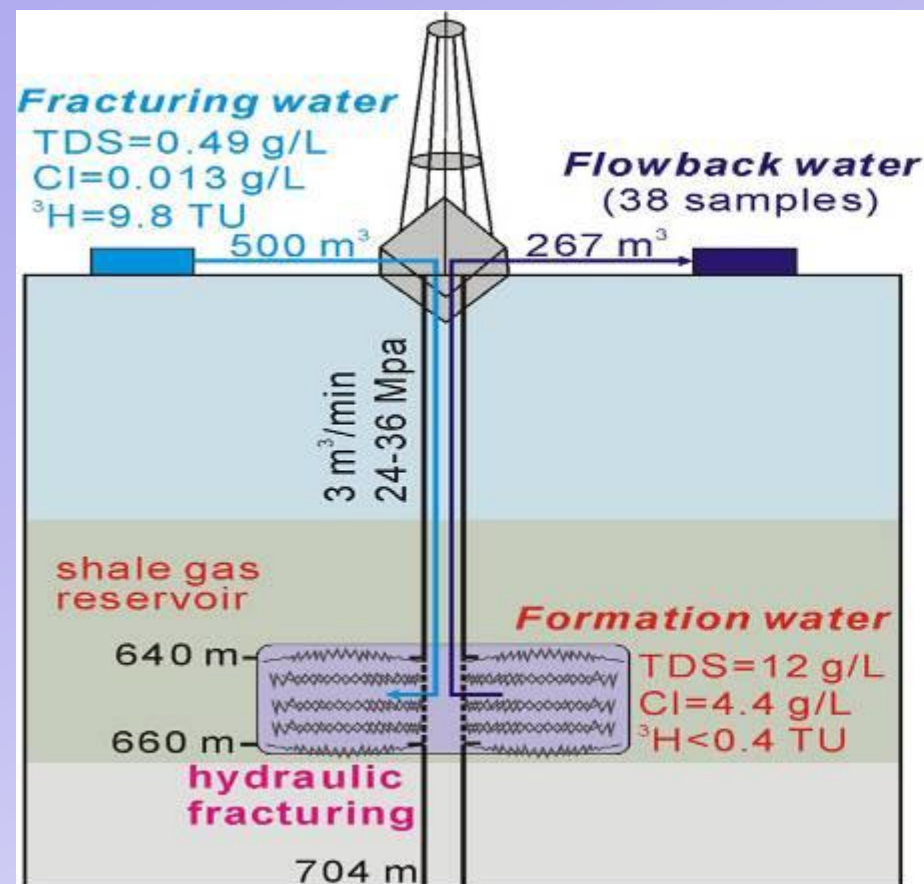


704米，垂直井
返排初期小时尺度取样



室内水岩反应实验

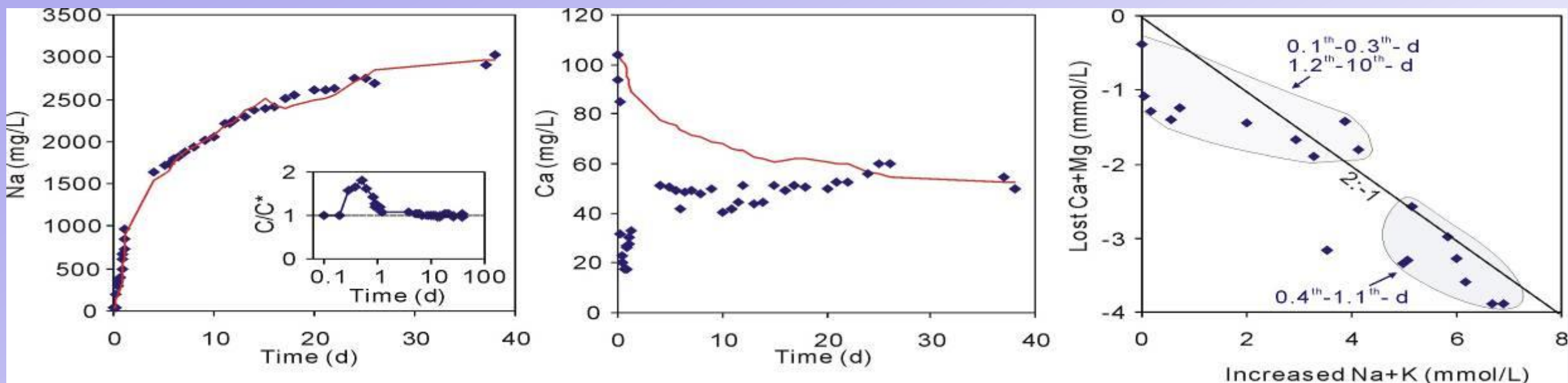
主要成果



地层水

返排液中，初期主要是压裂液；地层水第4天占50%、2周后80%

主要离子演化特征: Na vs. Ca+Mg



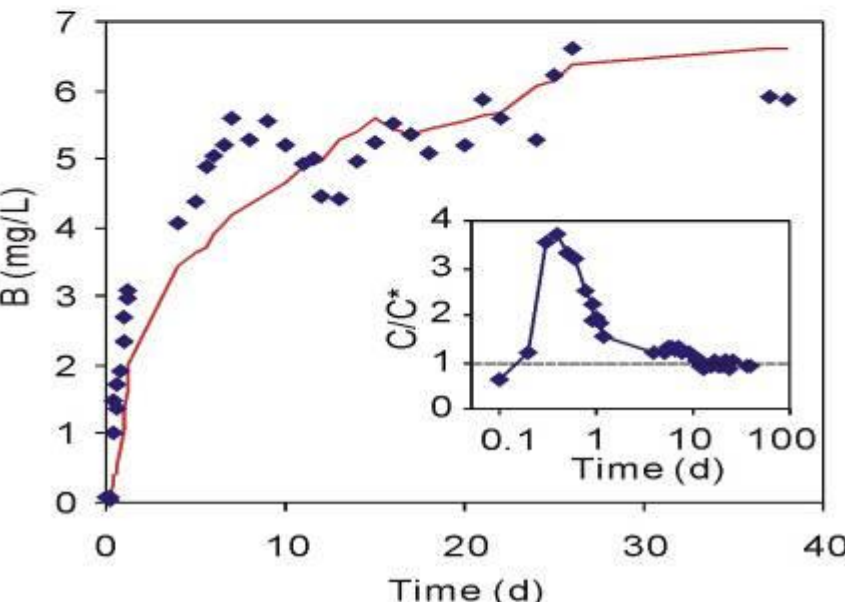
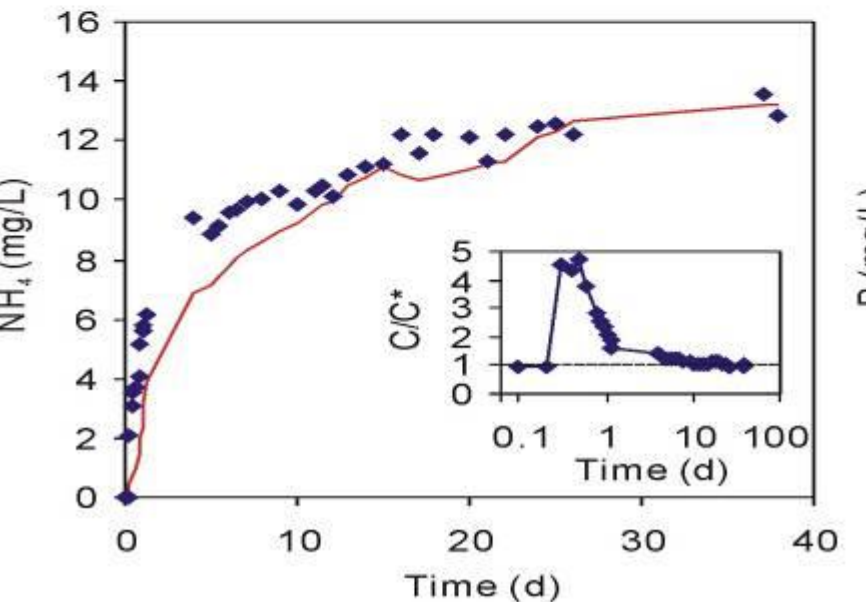
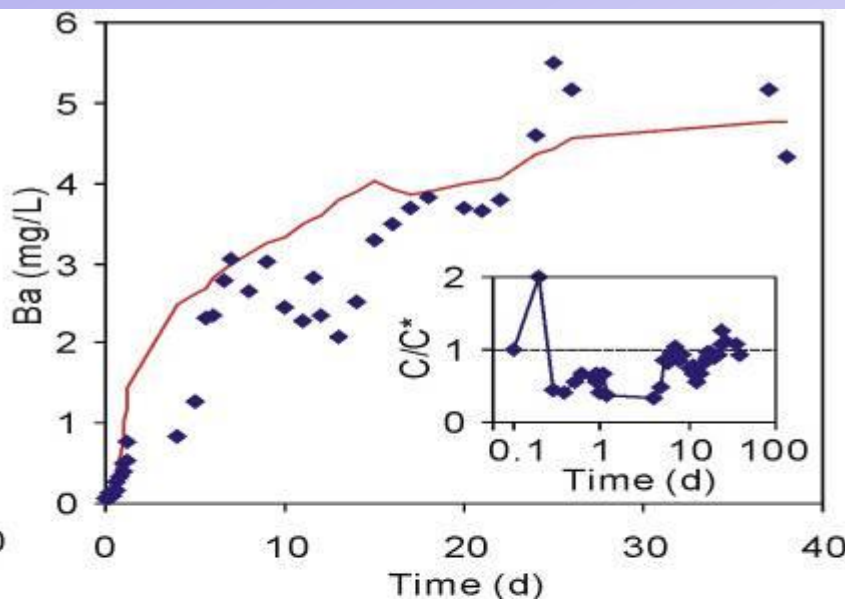
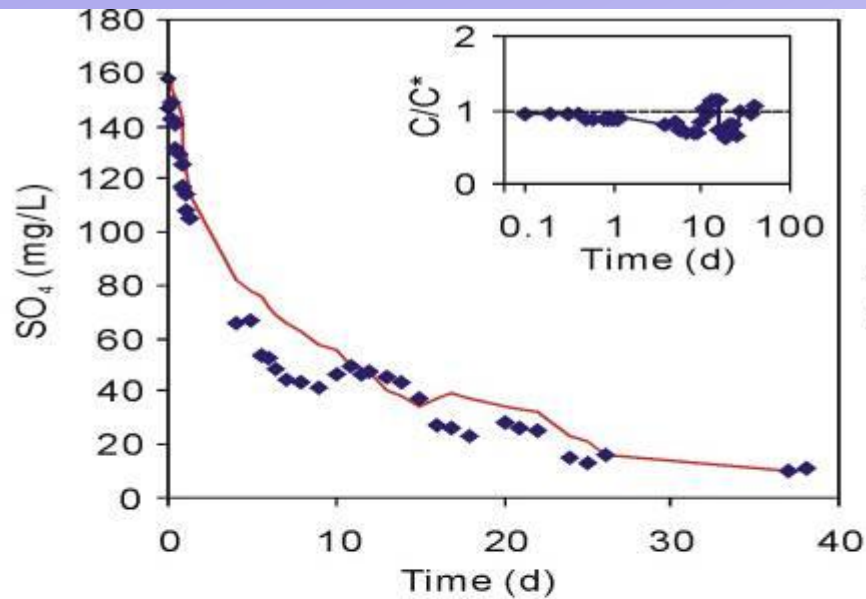
•Na增加

•Ca+Mg减少

CEC=54 meq/kg

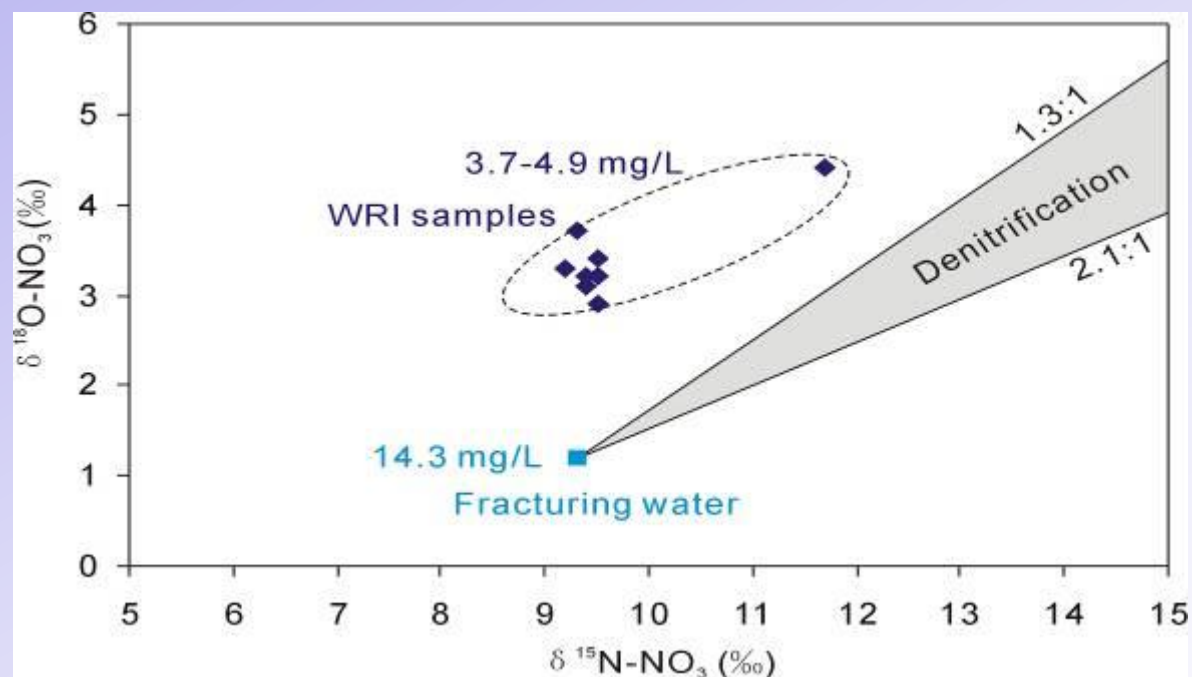
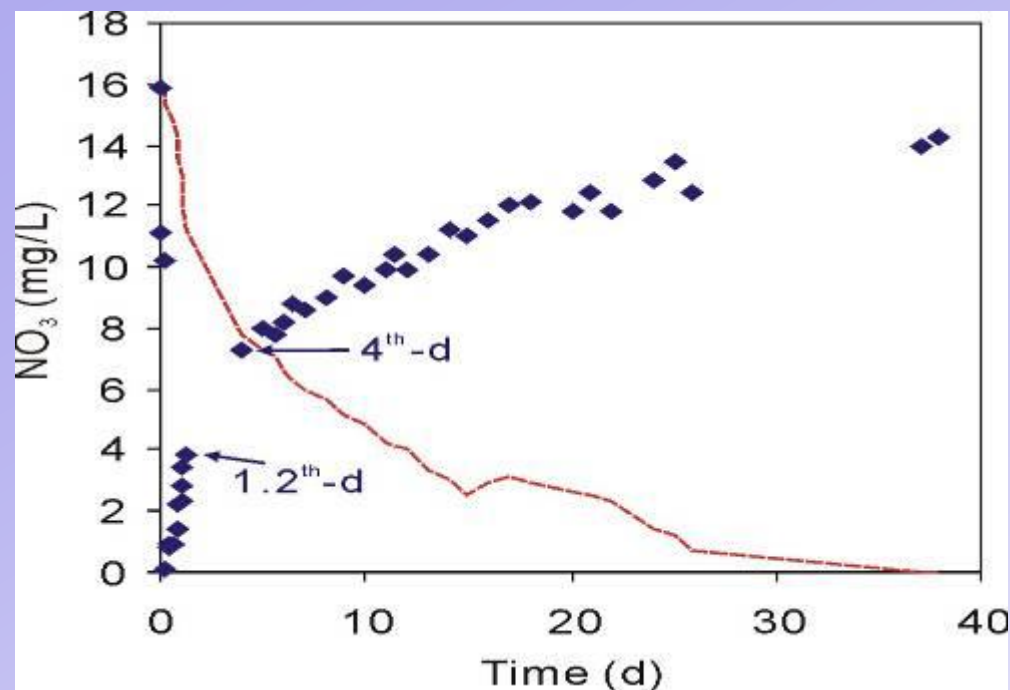
水岩反应时间	Ca (mg/L)	Mg (mg/L)	Na (mg/L)	K (mg/L)
压裂液	106	8.1	38	4
5 min	26	5.2	235	112
1 h	27	5.9	255	118
1 d	27	8.4	235	117
3 d	35	11.1	251	124
7 d	43	14.8	253	126

主要离子演化特征



- SO_4 在混合线
- Ba减少
- NH_4 增加
- B增加

主要离子演化特征: NO_3



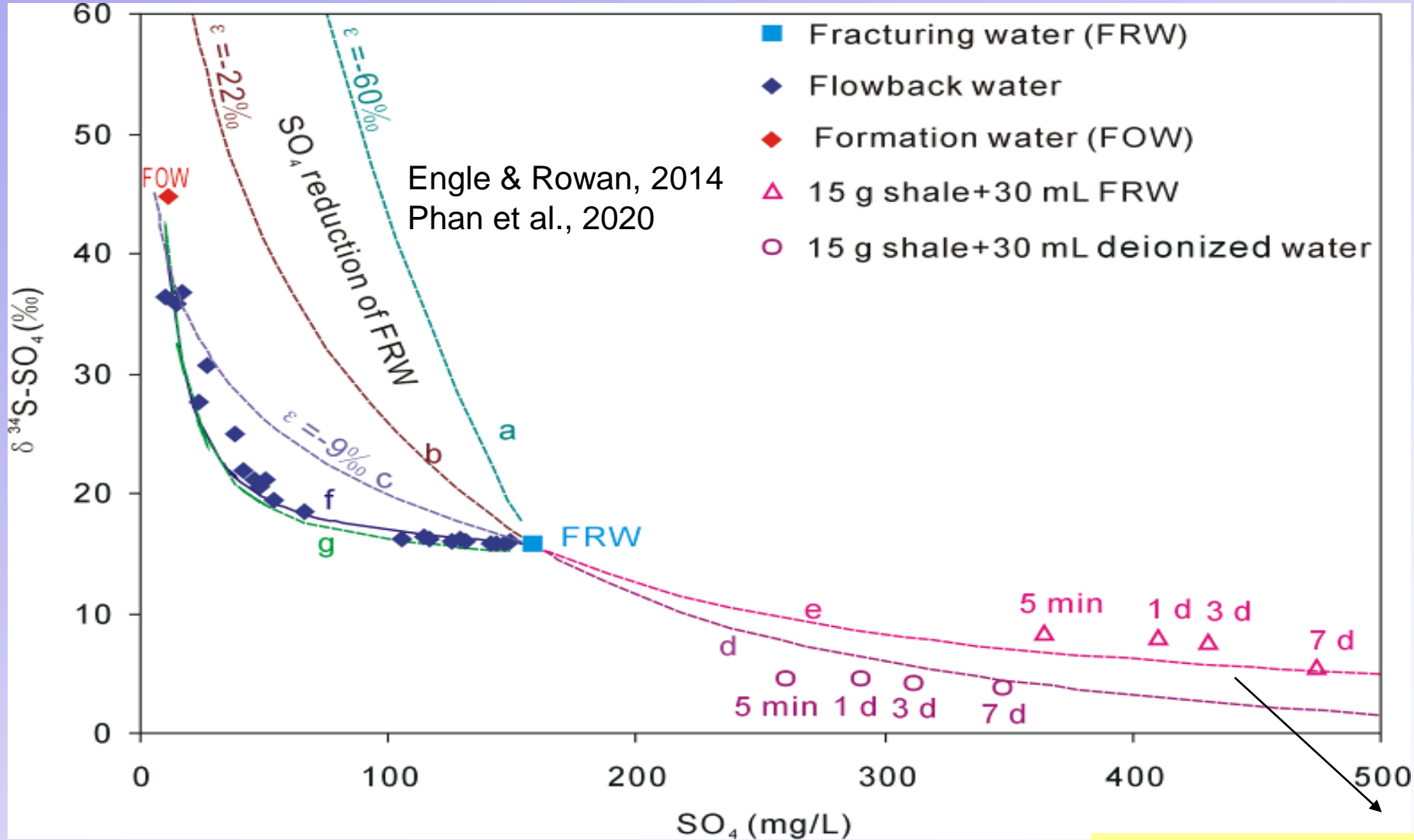
Set	Sample	Description	vol.	NO_3	mass
			mL	mg/L	μg
	fracturing water	fracturing water		14.31	429.3
1	1-1	15 g shale+30 mL fracturing water	24	3.99	95.7
	1-2	+30 mL deionized water	30	2.52	75.7
	1-3	+30 mL deionized water	30	1.51	45.2
	1-4	+30 mL deionized water	30	1.15	34.5
	1-5	+30 mL deionized water	36	0.93	33.5
	Recovery rate				66%
2	2-1	15 g shale+30 mL fracturing water	24	4.12	98.8
	2-2	+30 mL KCl solution	30	7.72	231.6
	2-3	+30 mL KCl solution	30	1.21	36.2
	2-4	+30 mL KCl solution	30	0.32	9.5
	2-5	+30 mL KCl solution	36	0.15	5.3
	Recovery rate				89%

现象: NO_3 含量先减少后降低

原因: 先吸附后解吸附

分馏效应: N同位素分馏不明显

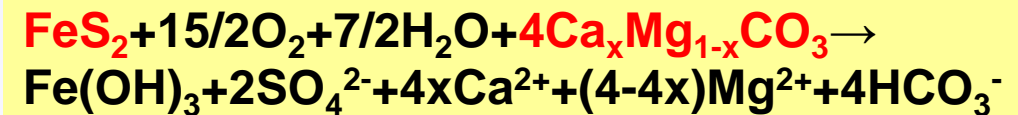
主要离子演化特征: SO_4



尚未进行很好地刻画
Owen et al., 2020

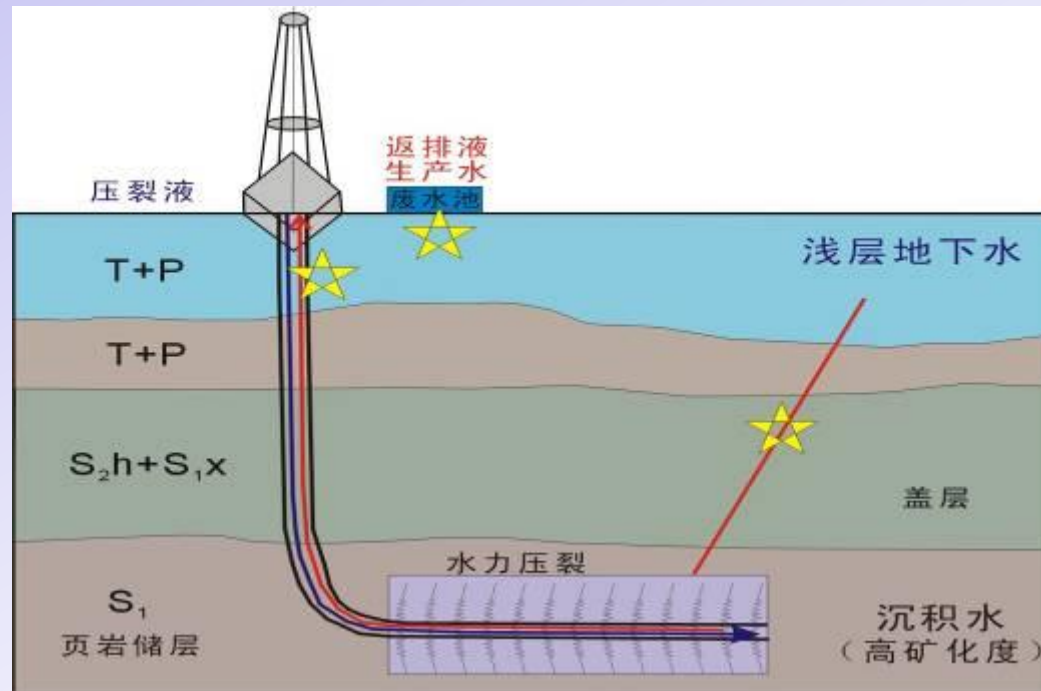
Zolfaghari et al., 2016
Paukert Vankeuren et al., 2017

期待发生并改善储层



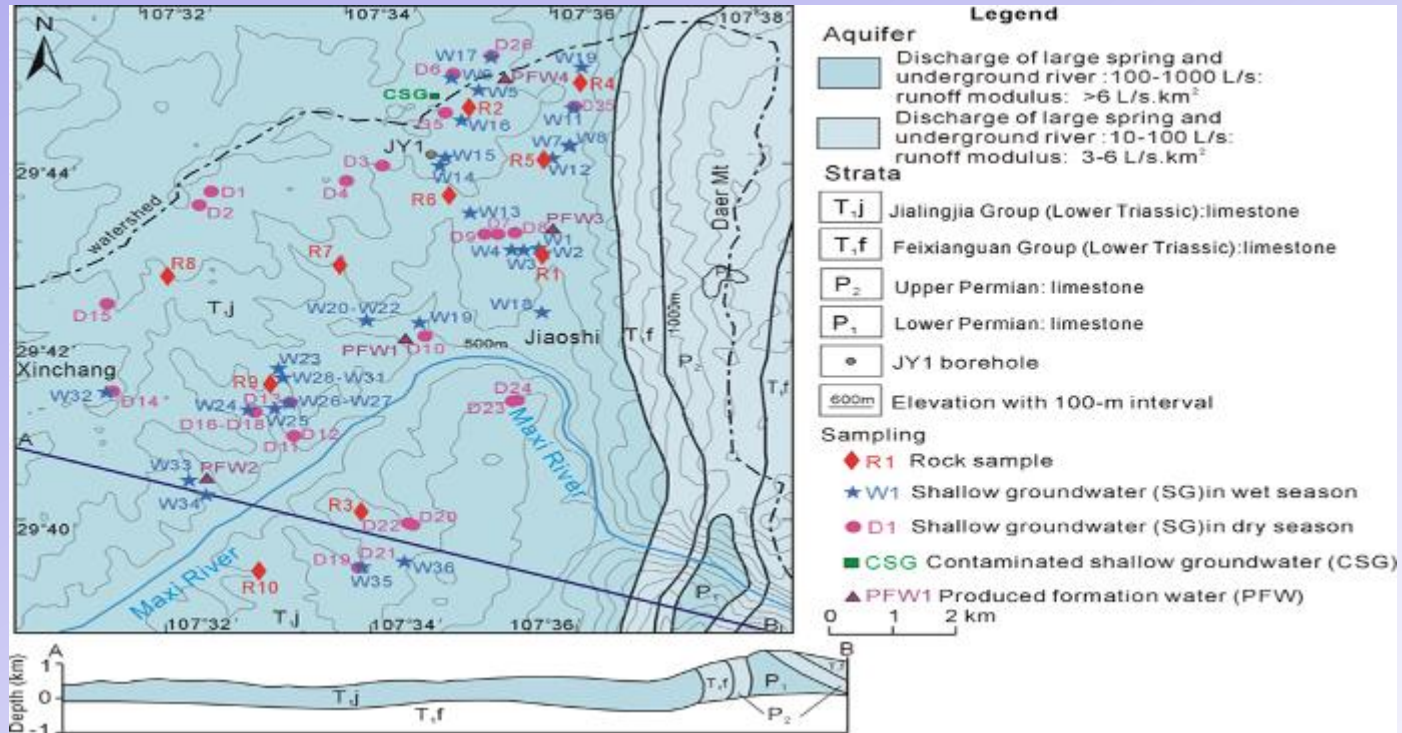
研究意义

原来认为是压裂液是重要污染源，目前看虽然压裂及返排过程中发生了众多地球化学反应，但返排液整体的水化学组成仍受控于高盐度地层水，因此**地层水的水化学、同位素及毒性研究对于页岩气开发的地下水水质监测及污染示踪具有重要意义**



地下水污染示踪

(一) 系统地建立了中国首个大型页岩气田（焦石坝）地下水环境背景值



120组地下水数据

- 常量组分
- 微量组分
- 放射性
- 水中溶解气体
- 同位素

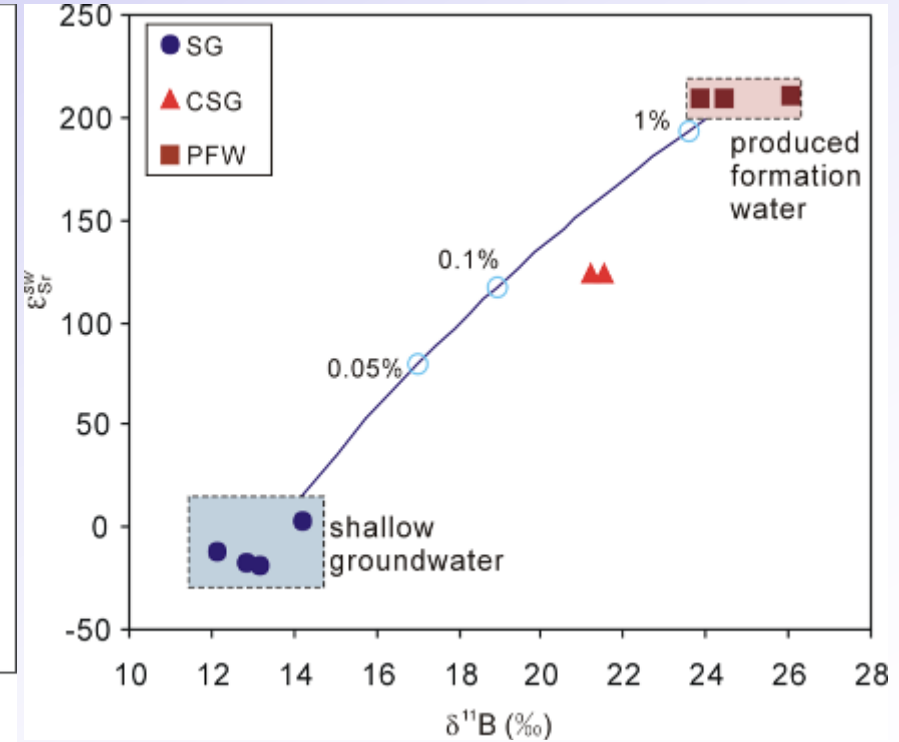
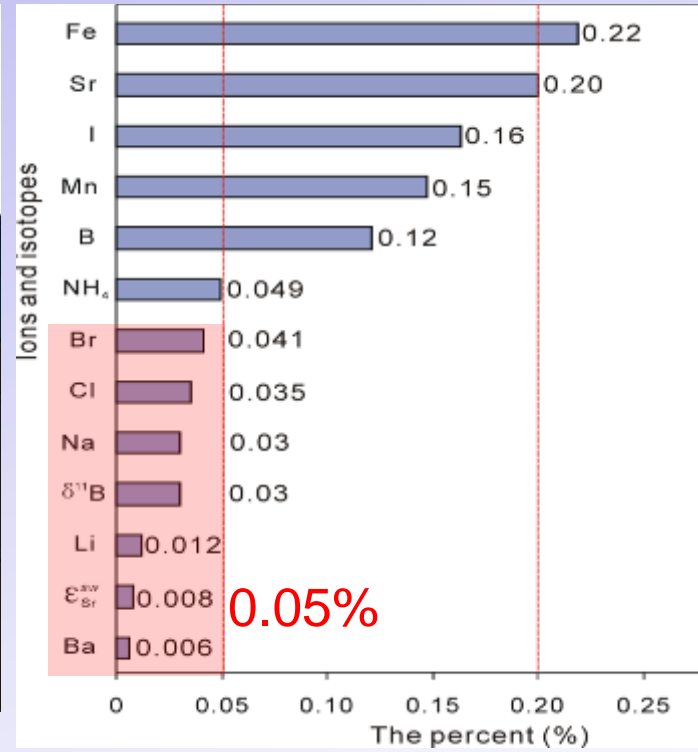
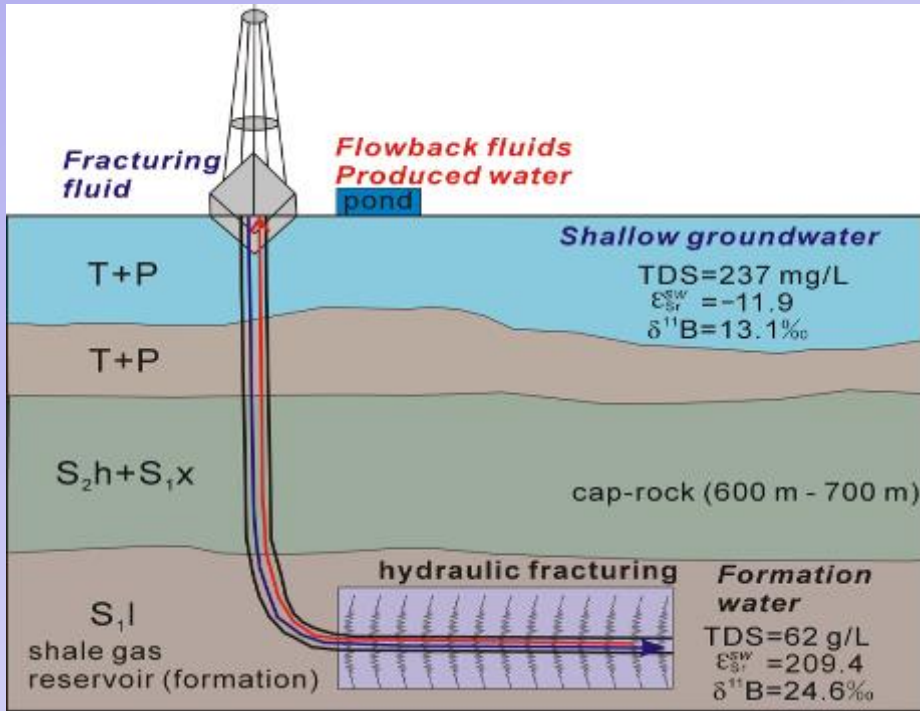
(Nicot et al., 2017a) or Eagle Ford Shale (Nicot et al., 2017b). Additional studies have been conducted in Canada (Rivard et al., 2017; Humez et al., 2016a, 2016b; Lavoie et al., 2014, 2016), Great Britain (Bell et al., 2016; Darling and Goody, 2006), Romania (Ionescu et al., 2017), China (Huang et al., 2017) and Australia (Currell et al., 2017; Atkins et al., 2015). Schloemer et al., 2018

Huang T, Pang Z, Tian J, Li Y, Yang S, Luo L (2017) Methane content and isotopic composition of shallow groundwater: implications for environmental monitoring related to shale gas exploitation. *JRNC* 312: 577-585

地下水污染示踪

(二) 提出了地下水敏感性监测指标的**确定框架**

(1) 基于浅层地下水和地层水两个端元； (2) 对比分析； (3) 通用框架



Huang T, Pang Z, Li Z, Li Y, Hao Y (2020) A framework to determine sensitive inorganic monitoring indicators for tracing groundwater contamination by produced formation water from shale gas development in the Fuling Gasfield, SW China. *Journal of Hydrology* 581, 124403

地下水污染示踪

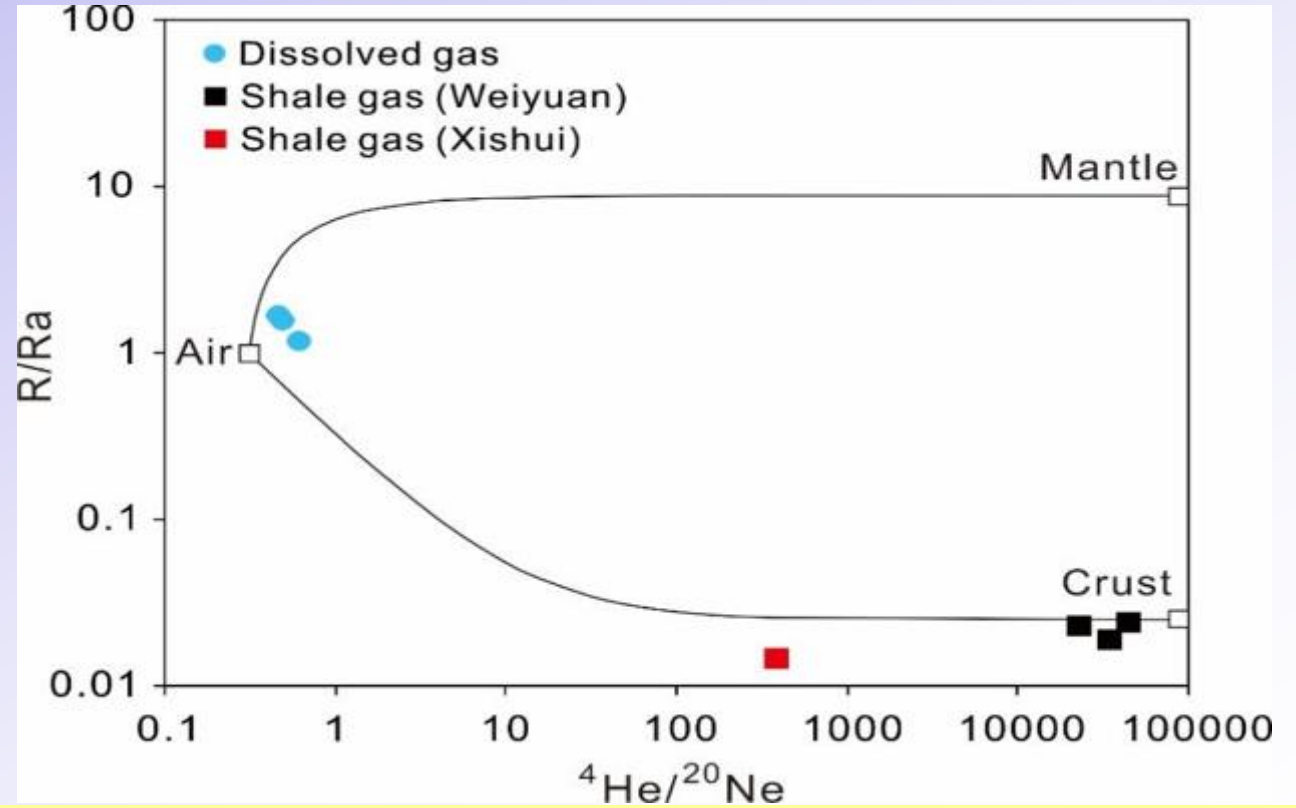
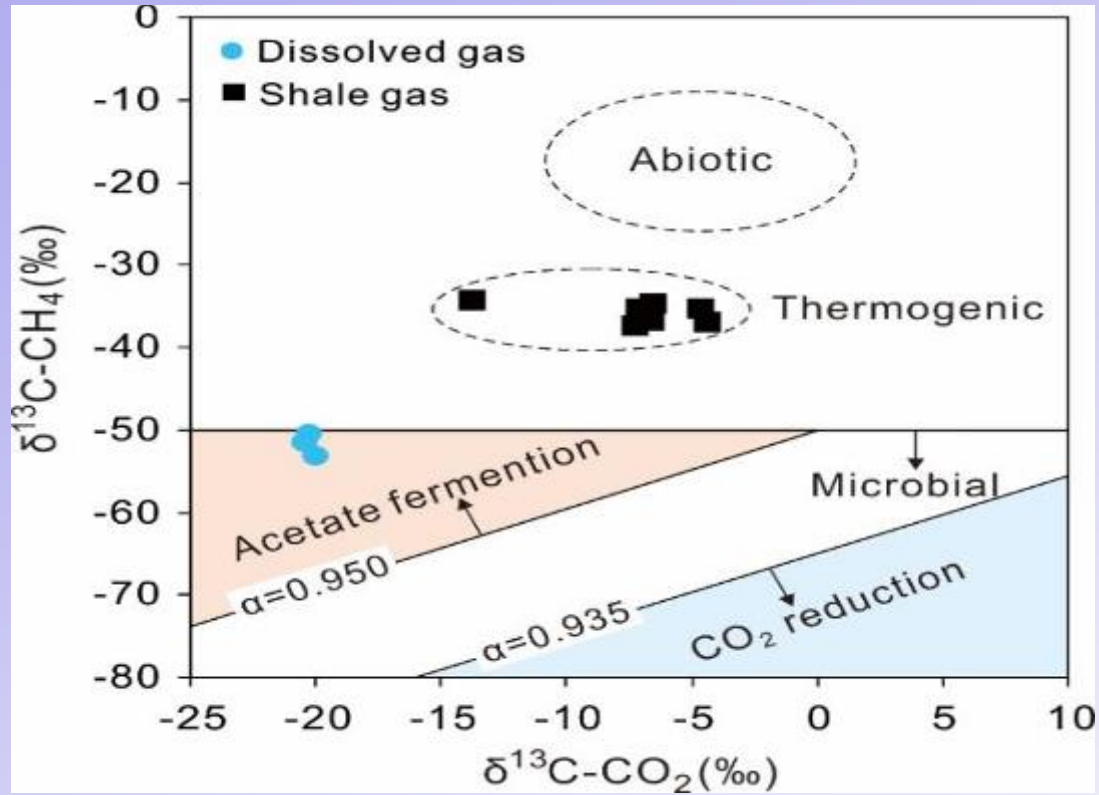
(二) 提出了地下水敏感性监测指标的**确定框架**

基于环境背景值数据库及地下水敏感性监测指标，开发了《**焦石坝区块页岩气开发地下水污染识别软件**》，并被当地环境管理部门成功应用



地下水污染示踪

(三) 引入惰性气体帮助识别气体污染



Huang T, Pang Z, Tian J, Li Y, Yang S, Luo L (2017) Methane content and isotopic composition of shallow groundwater: implications for environmental monitoring related to shale gas exploitation. *JRNC* 312: 577-585

Li Z, Huang T, Ma B, Long Y, Zhang F, Tian J, Li Y, Pang Z (2020) Baseline groundwater quality before shale gas development in Xishui, Southwest China: Analyses of hydrochemistry and multiple environmental isotopes (^2H , ^{18}O , ^{13}C , $^{87}\text{Sr}/^{86}\text{Sr}$, ^{11}B , and noble gas isotopes). *Water* 12, 1741

相关论文

Geophysical Research Letters



RESEARCH LETTER

10.1029/2020GL090420

Key Points:

- This study provided novel and detailed insights into the water-rock interactions following hydraulic fracturing
- Cation exchange, adsorption/desorption, and barite precipitation were observed both in the laboratory and field studies
- The oxidation of pyrite was not

Identification of Geochemical Processes During Hydraulic Fracturing of a Shale Gas Reservoir: A Controlled Field and Laboratory Water-Rock Interaction Experiment

Tianming Huang^{1,2,3} , Zhenbin Li^{1,3}, Bernhard Mayer² , Michael Nightingale², Xiao Li^{1,3}, Guanfang Li^{1,3}, Yin Long^{1,3}, and Zhonghe Pang^{1,3} 

¹Key Laboratory of Shale Gas and Geoengineering, Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing, China, ²Department of Geoscience, University of Calgary, Calgary, Canada, ³College of Earth and Planetary Sciences, University of Chinese Academy of Sciences, Beijing, China