Istituto Nazionale di
Geofisica e Vulcanologia

Geologic methane sources

Principles, measurements, misconceptions, global emission estimates

Giuseppe Etiope

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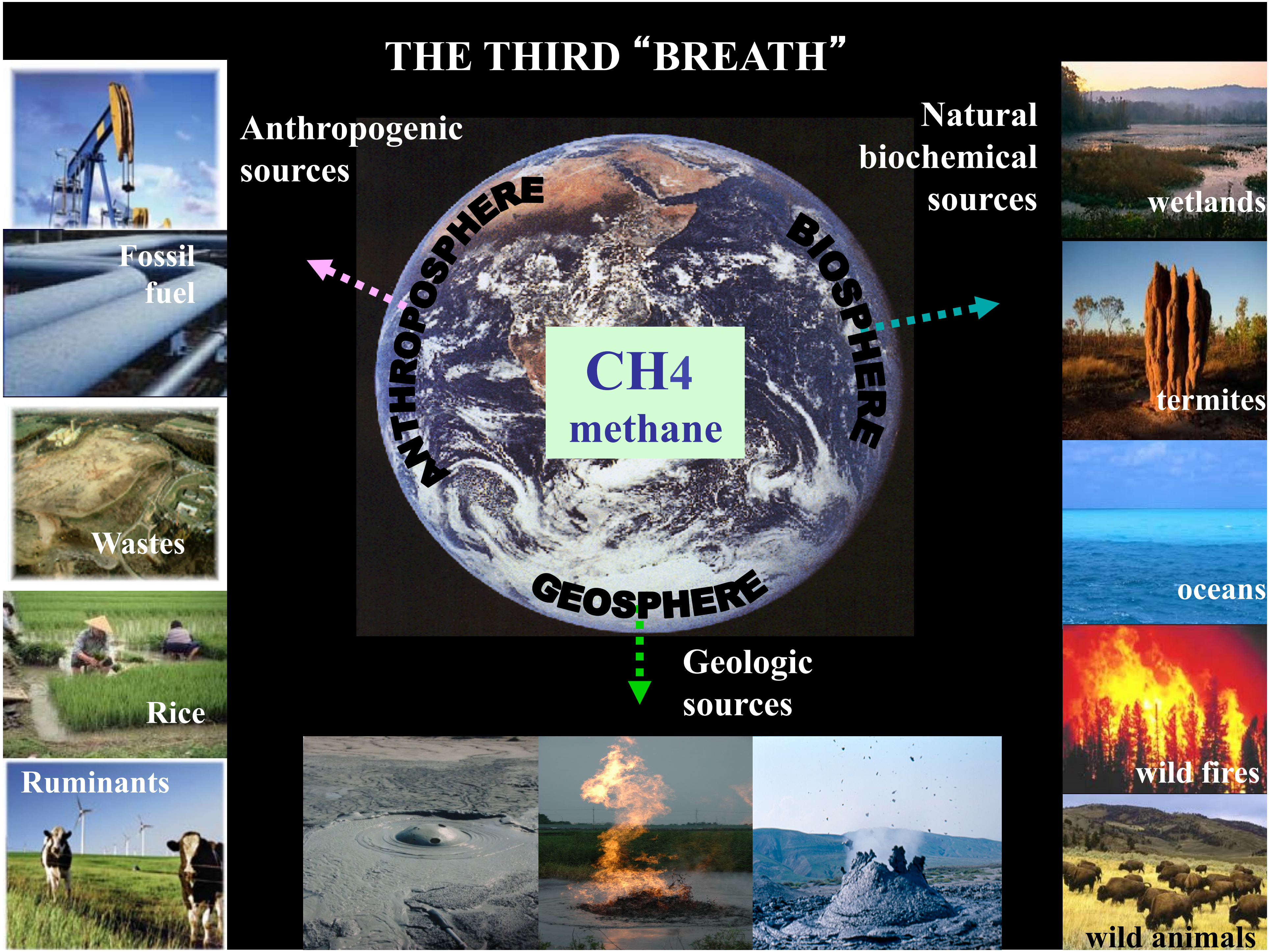
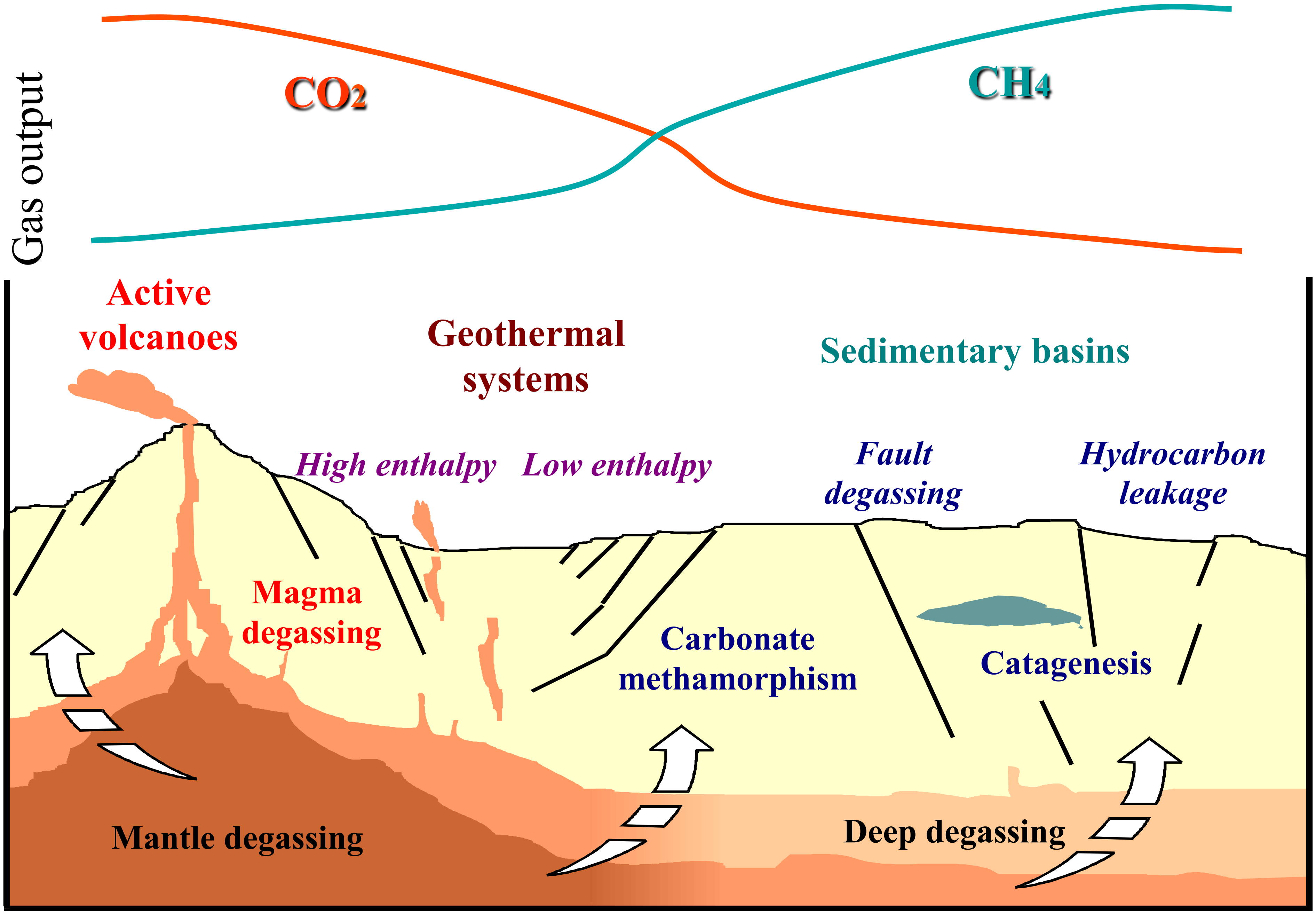
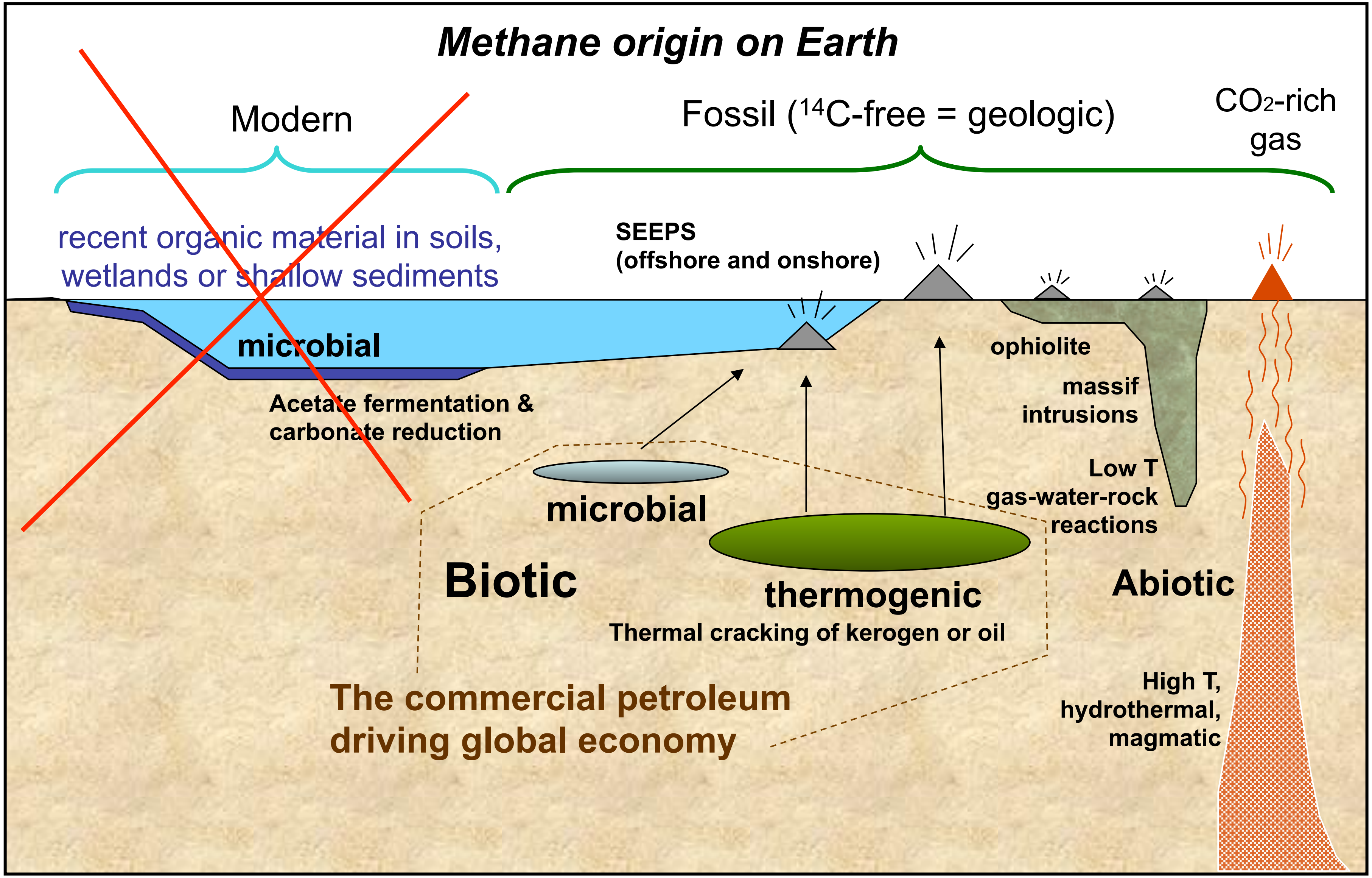

THE THIRD "BREATH"
Anthropogenic sources
Natural biochemical sources
ANTHROPOSPHERE
BIOSPHERE
GEOSPHERE
CH4
methane
Geologic sources
Fossil fuel
Wastes
Rice
Ruminants
wetlands
termites
oceans
wild fires
wild animals

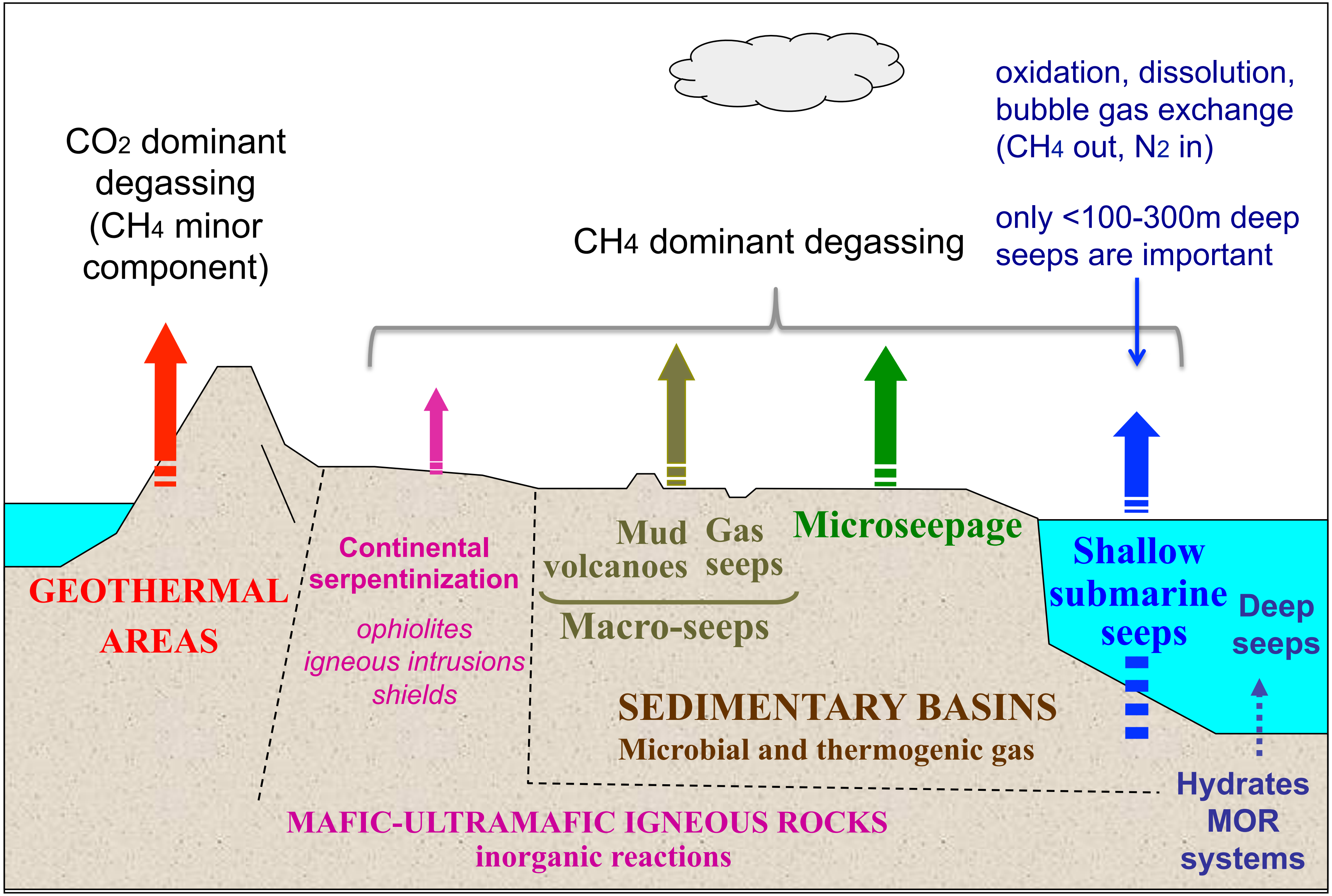
CO_2 and CH_4 DEGASSING FROM THE SOLID EARTH



What is geologic methane



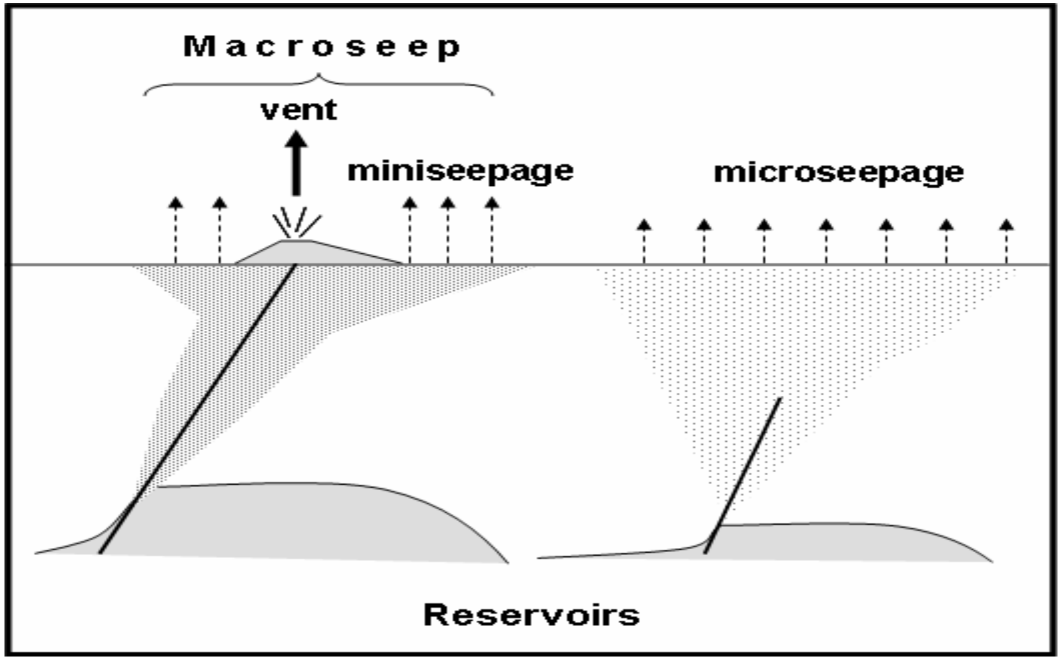
GEOLOGIC METHANE SOURCES

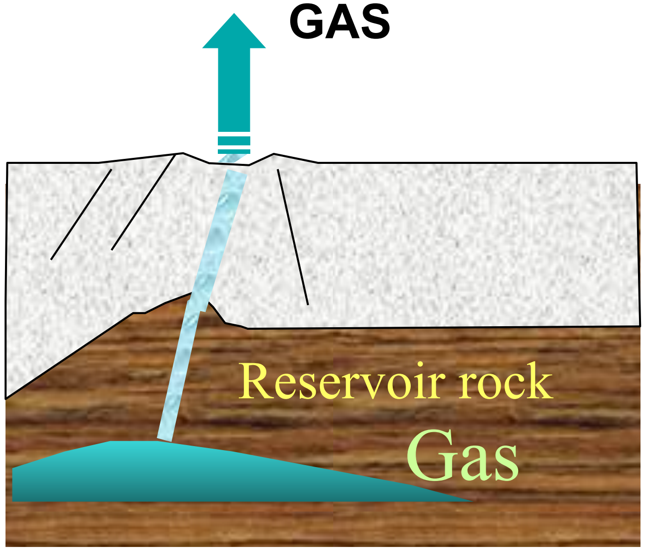


Seepage….a jungle of names

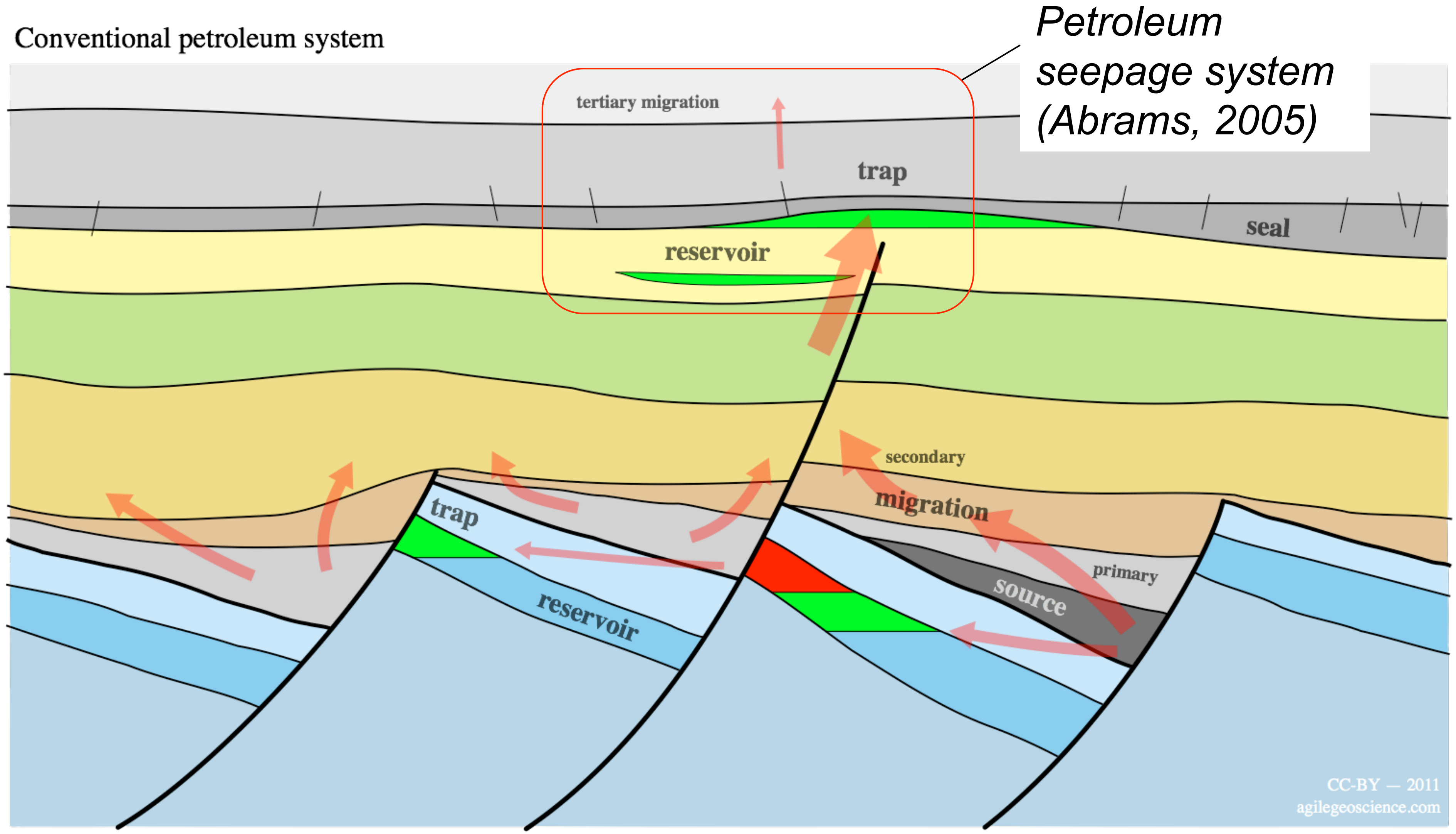
Macroseeps
- Focused flow
  - Gas seeps *(gas only)*
  - Oil seeps *(gas and oil)*
  - Water-gas seeps *(bubbling springs or aquifers)*
  - Mud volcanoes *(gas, water, oil and sediments)*
- Diffuse flow
  - Miniseepage (invisible)

Microseepage - Diffuse flow (invisible, independent from macroseeps)





TOTAL PETROLEUM SYSTEM



Gas seeps and “eternal” fires

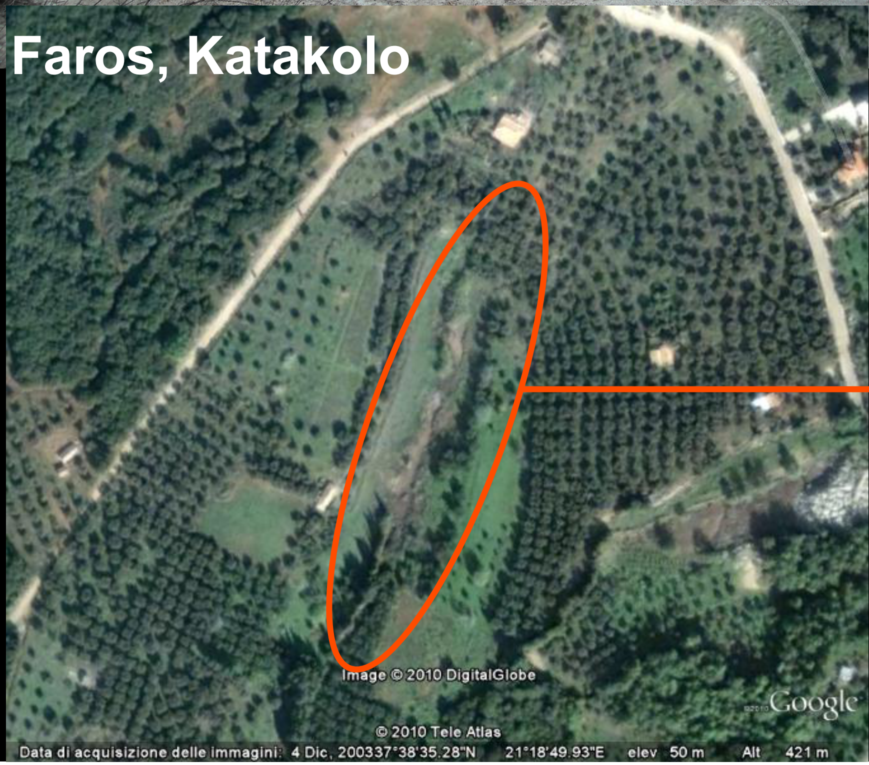

Eternal fires and Zoroastrianism
Azerbaijan (Yanardag)

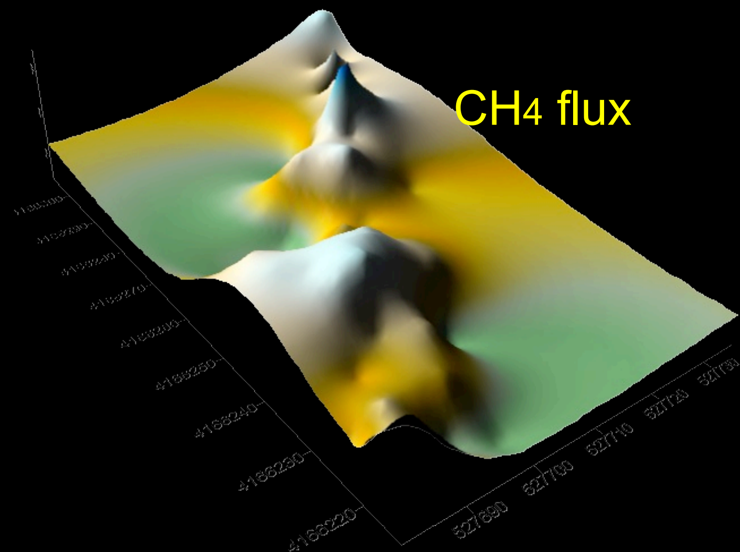

Baba Gurgur (Iraq) one of the largest oil fields in the world
A natural seep active since >4000 years; probably the “burning fiery furnace” into which Nebuchadnezzar cast the Jews (*Yergin, 1991*).
mentioned by Herodotus, Plutarch, and in the Old Testament’s Book of Daniel

MACRO-SEEPS

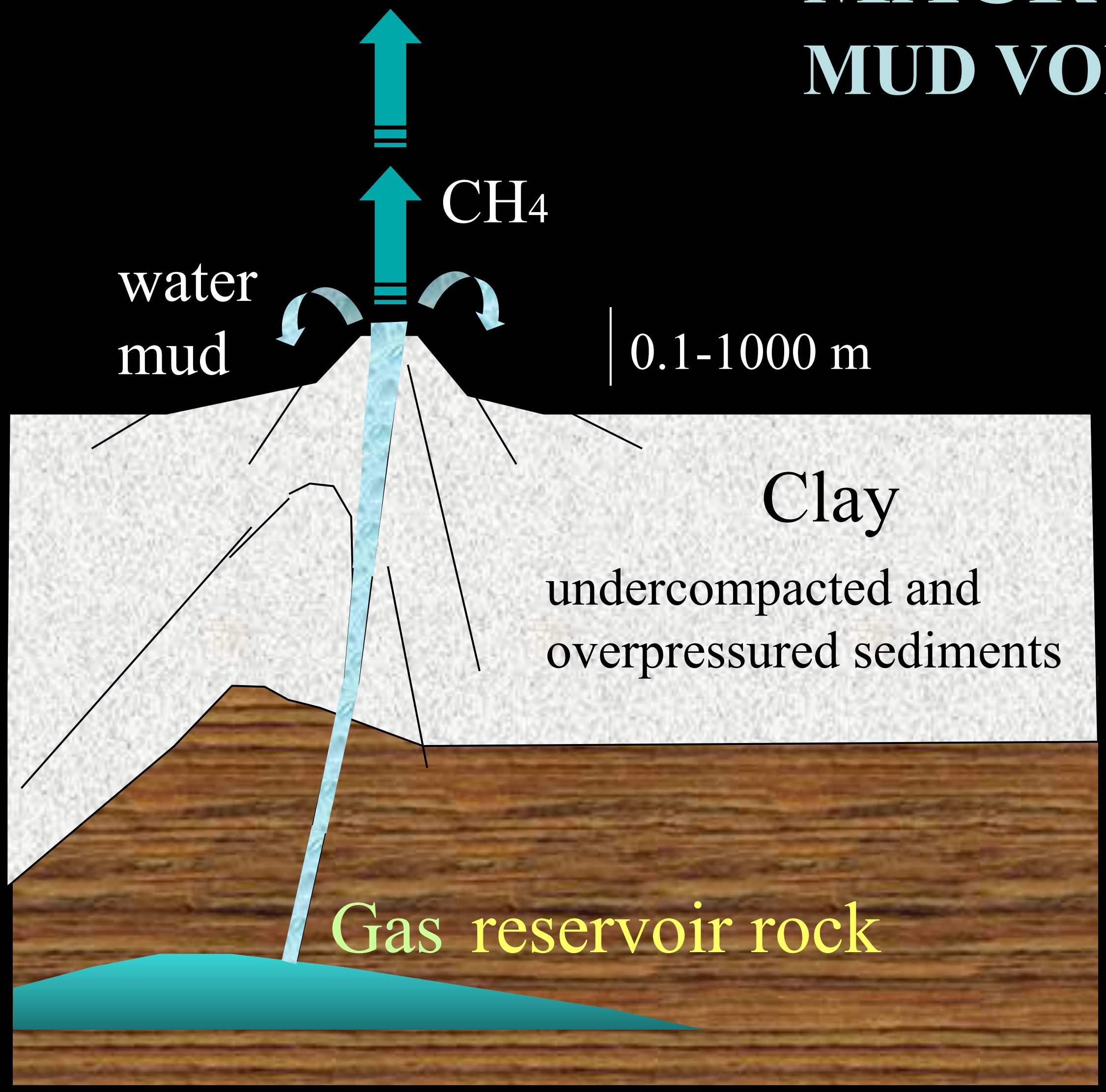
GAS-SEEPS

Greece





MACRO-SEEPS
MUD VOLCANO



Gas release to the atmosphere

Gas migration along faults

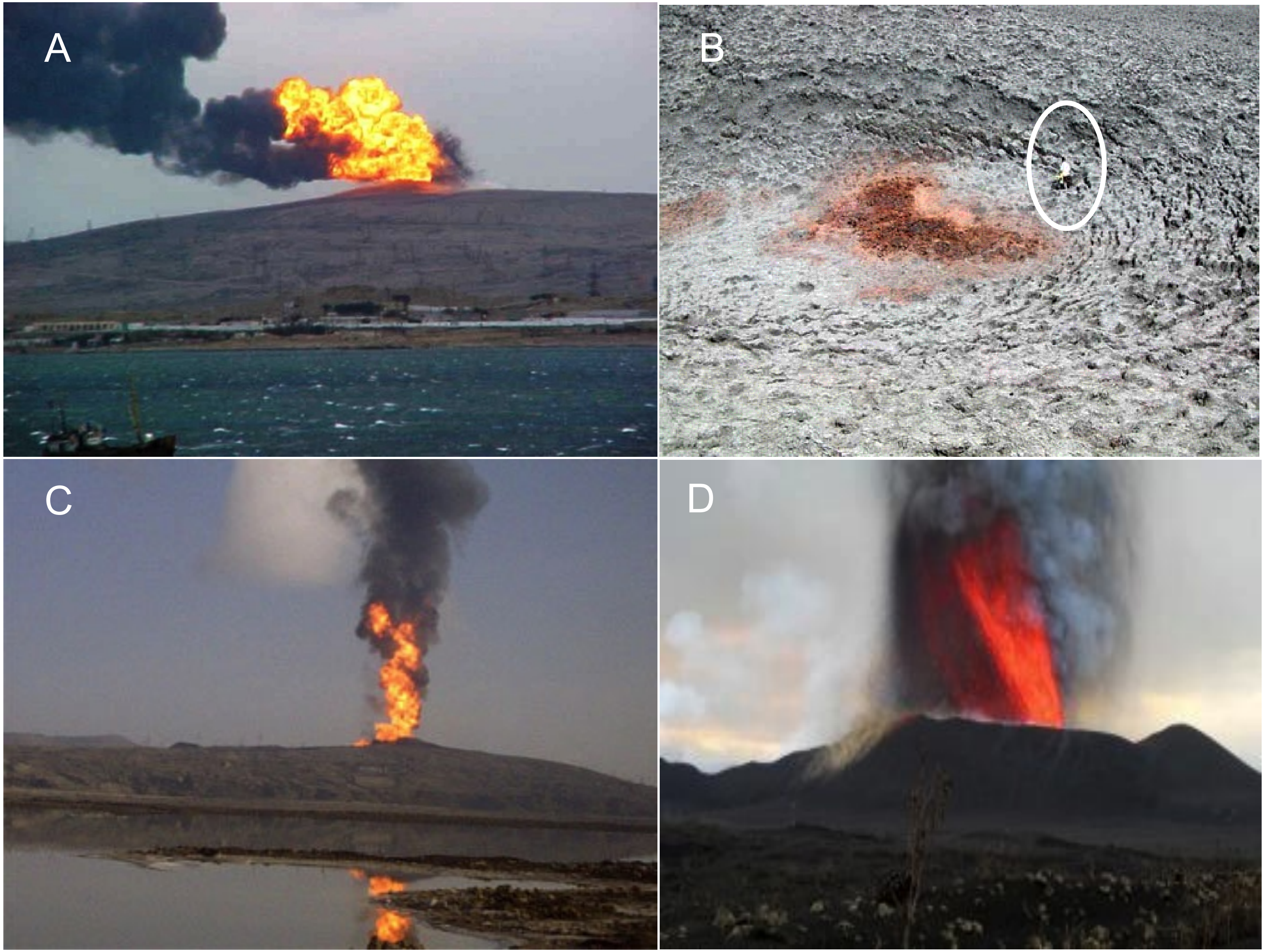
Pressurised gas from hydrocarbon reservoir

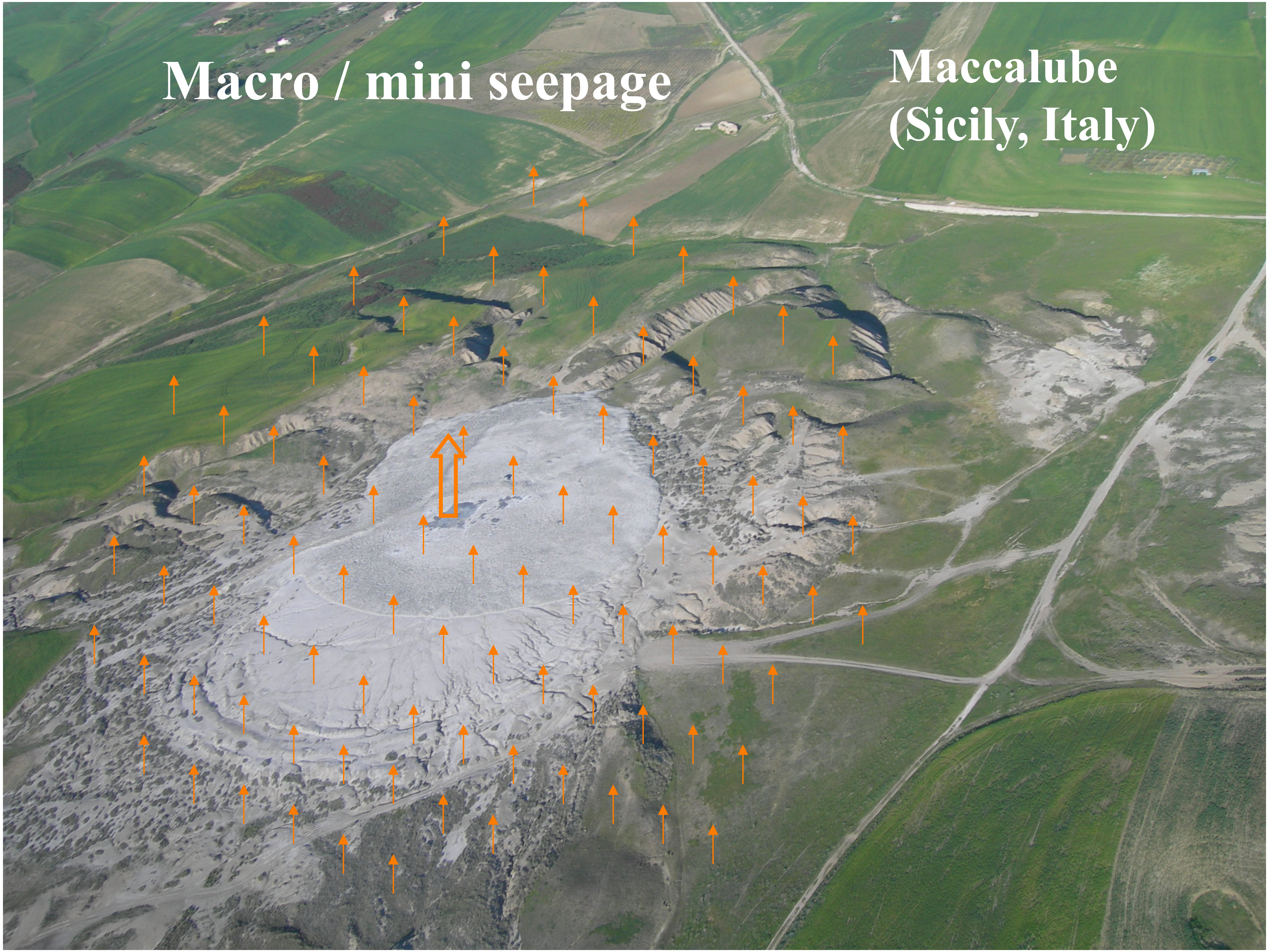
Mud volcanoes

Sedimentary volcanism, 3-phase system : gas-water-sediment

Mud volcanoes – Azerbaijan

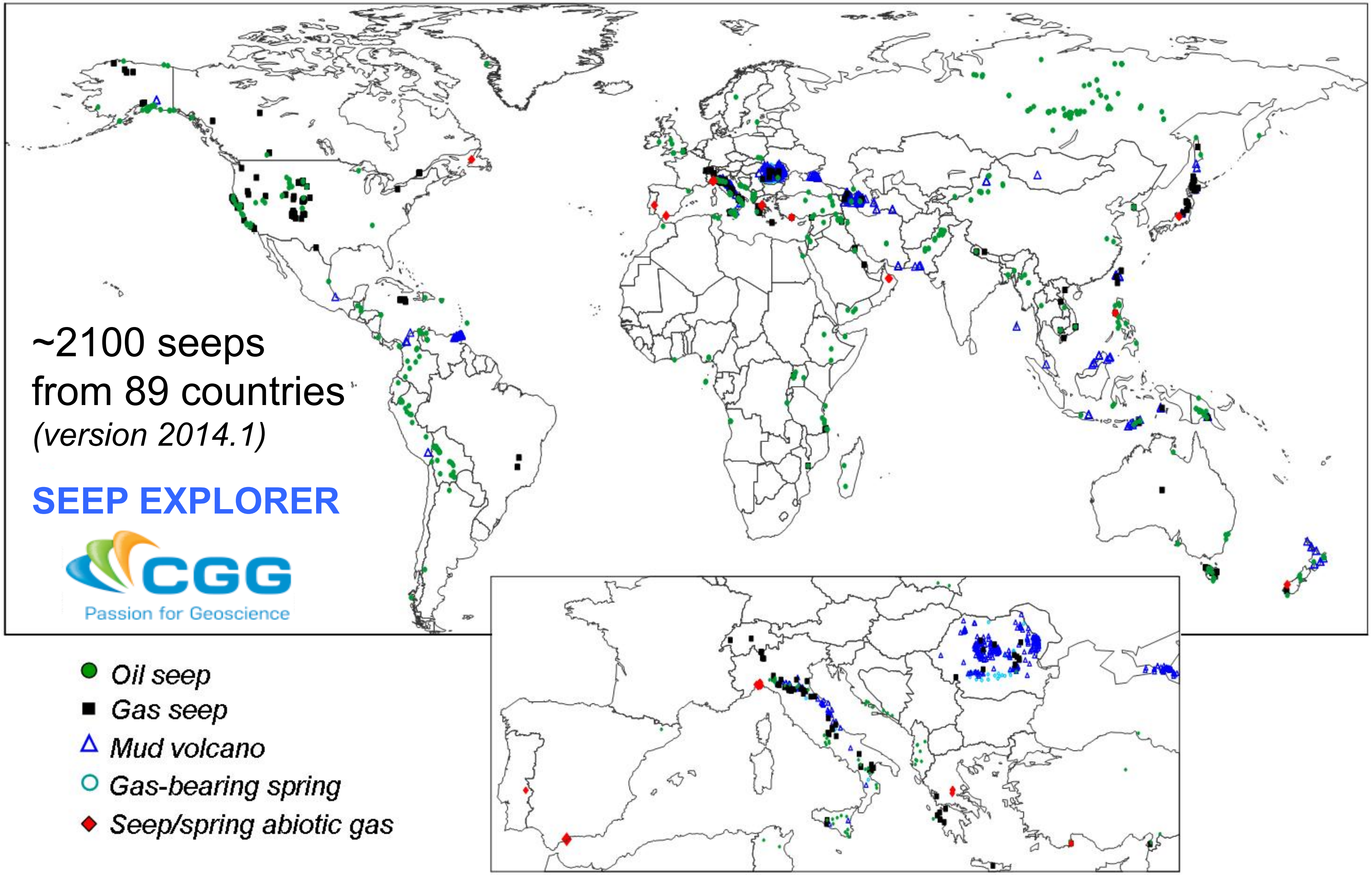
Mud volcano eruptions




Macro / mini seepage
Maccalube
(Sicily, Italy)

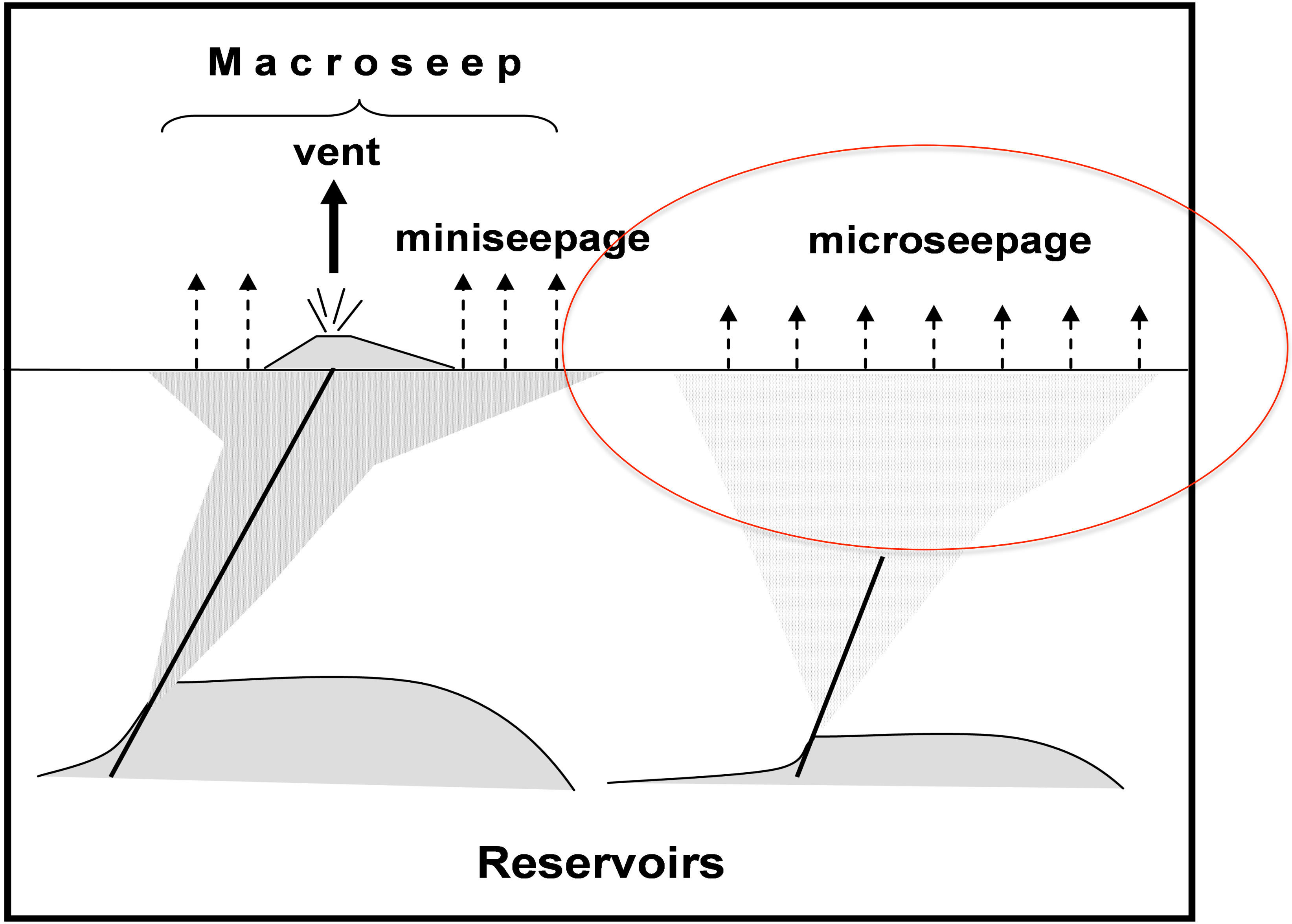
GLOBAL DISTRIBUTION OF SEEPS

(from GLOGOS - Global Onshore Gas-Oil seeps database)

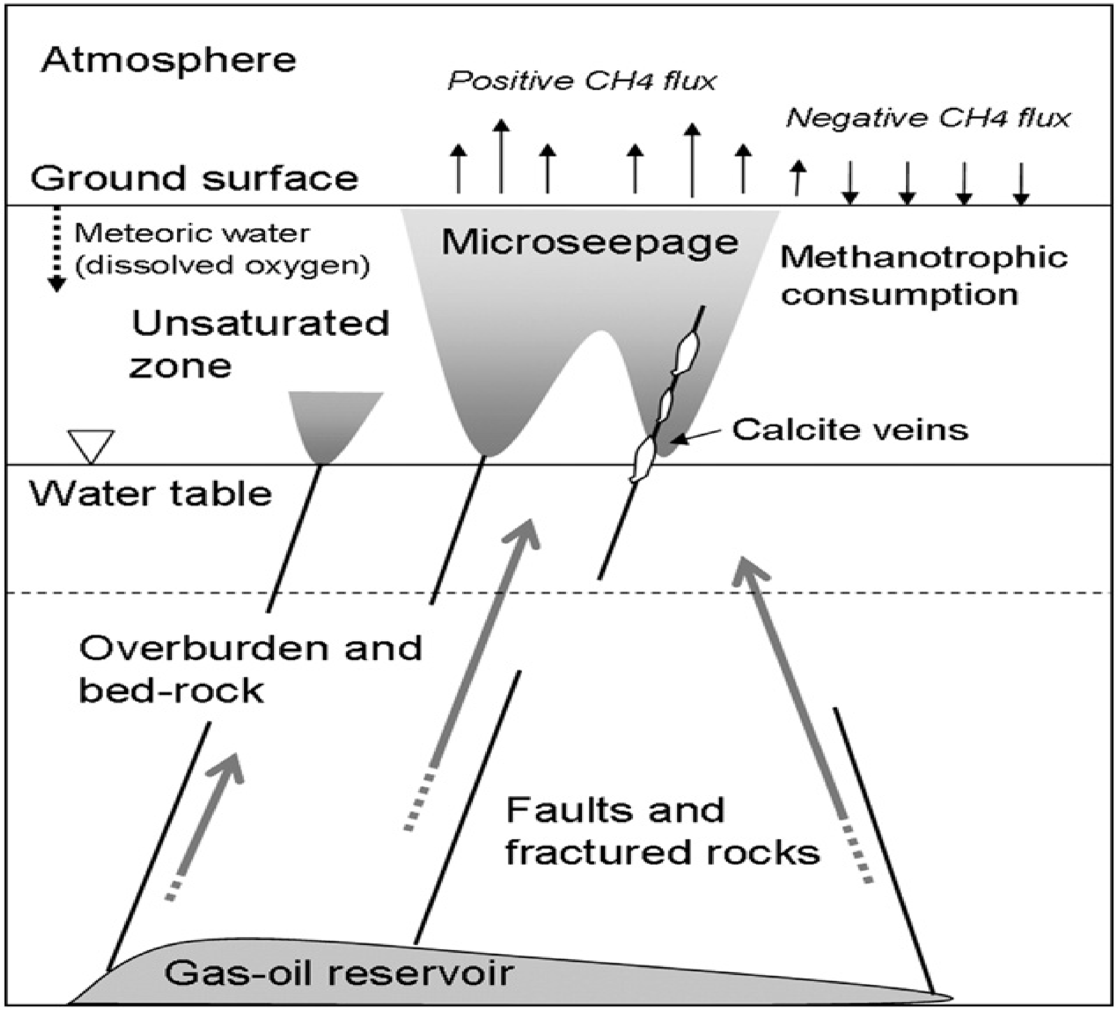


- Oil seep
- Gas seep
- Mud volcano
- Gas-bearing spring
- Seep/spring abiotic gas

Actual global number >10,000 ?


Macroseep
vent
miniseepage
microseepage
Reservoirs

Microseepage: the invisible emission



Global and Planetary Change 72 (2010) 265–274

Microseepage in drylands: Flux and implications in the global atmospheric source/sink budget of methane

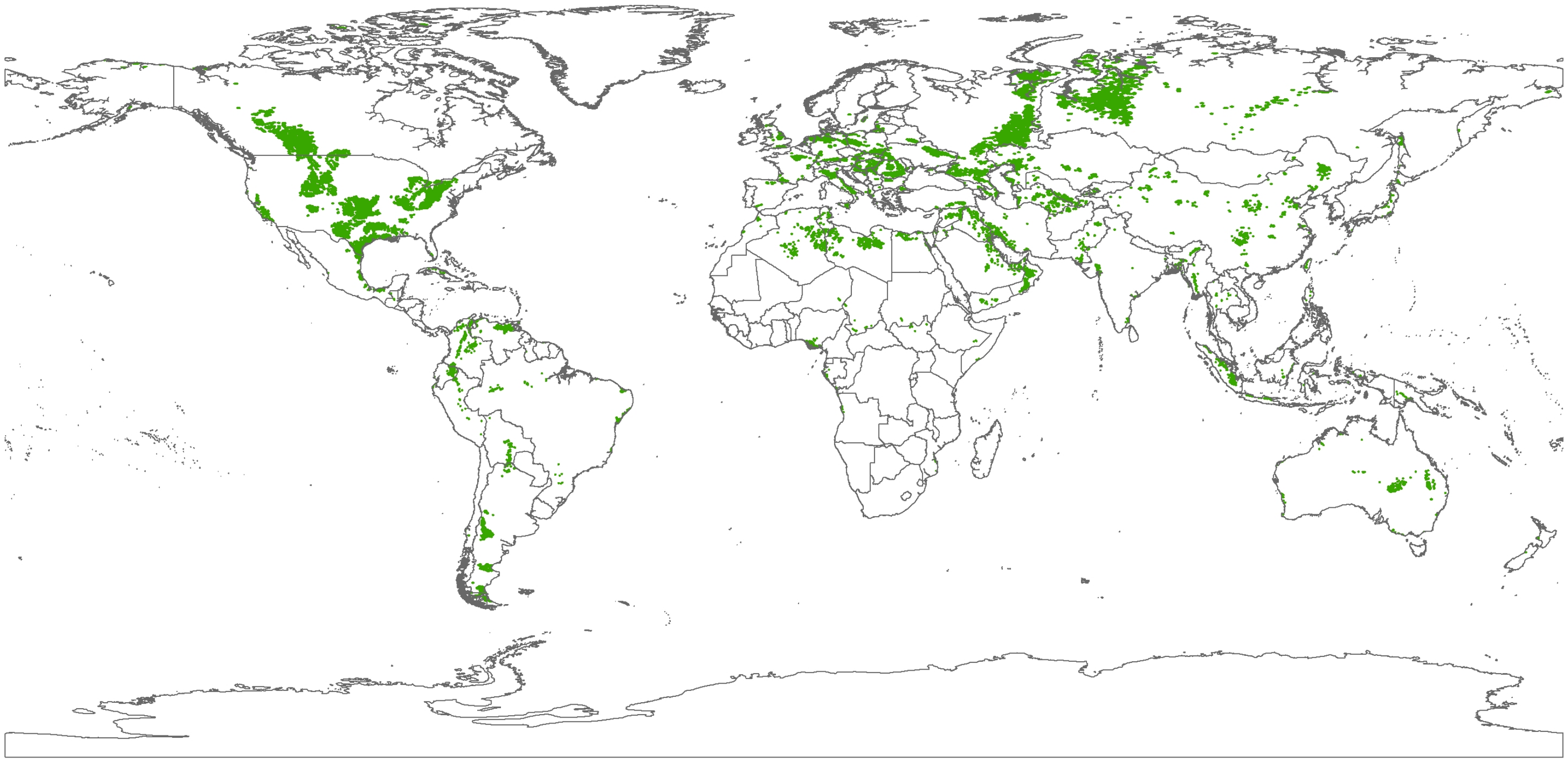
Giuseppe Etiope [a,b,*], Ronald W. Klusman [c]

Potential microseepage area (Level 1)

(Etiope, 2015)

Sedimentary basins with petroleum provinces

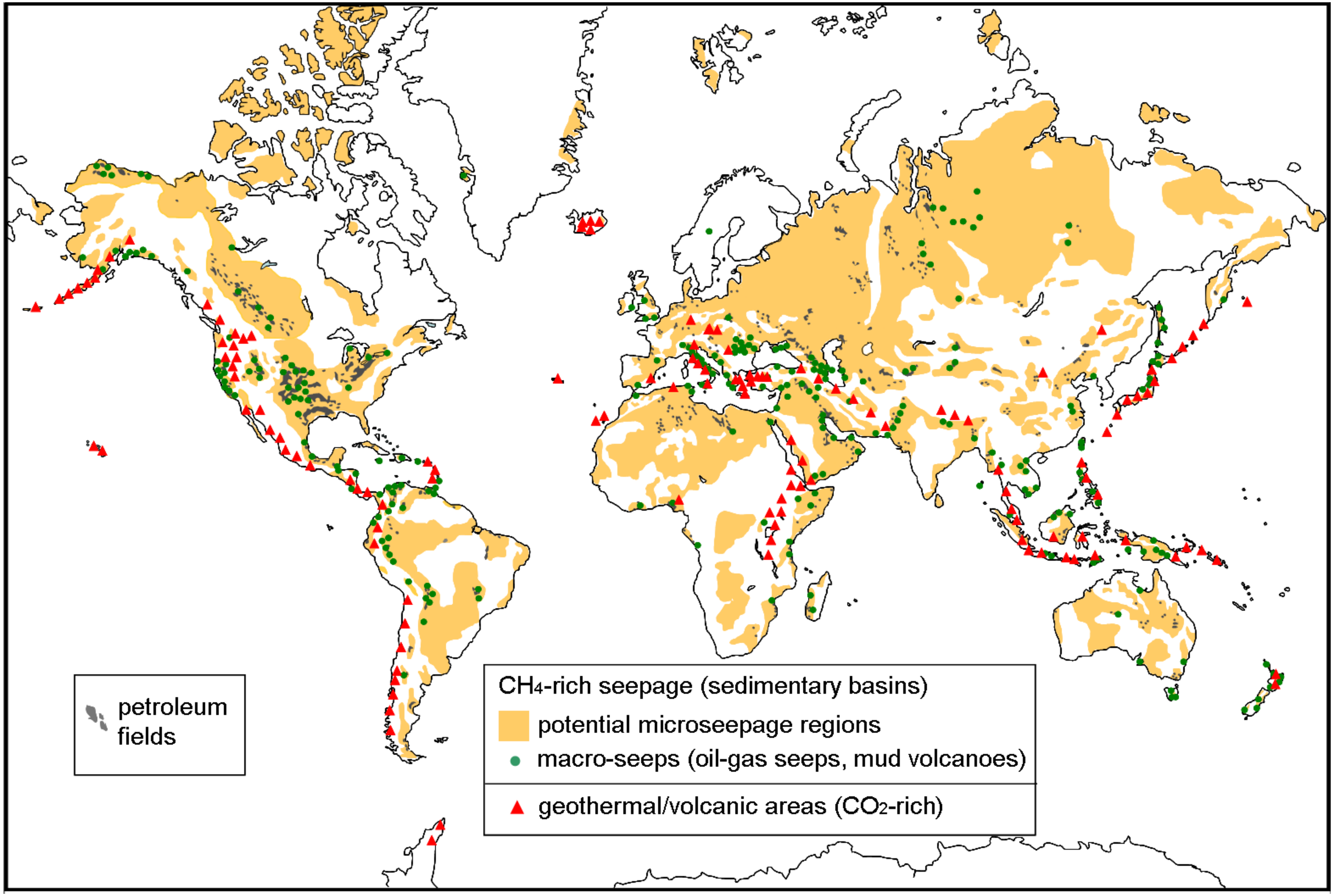
Potential microseepage area (Level 2)



~ 10 million km^2

Petroleum field areas

GLOBAL MAP OF GEOLOGIC METHANE EMISSION AREAS



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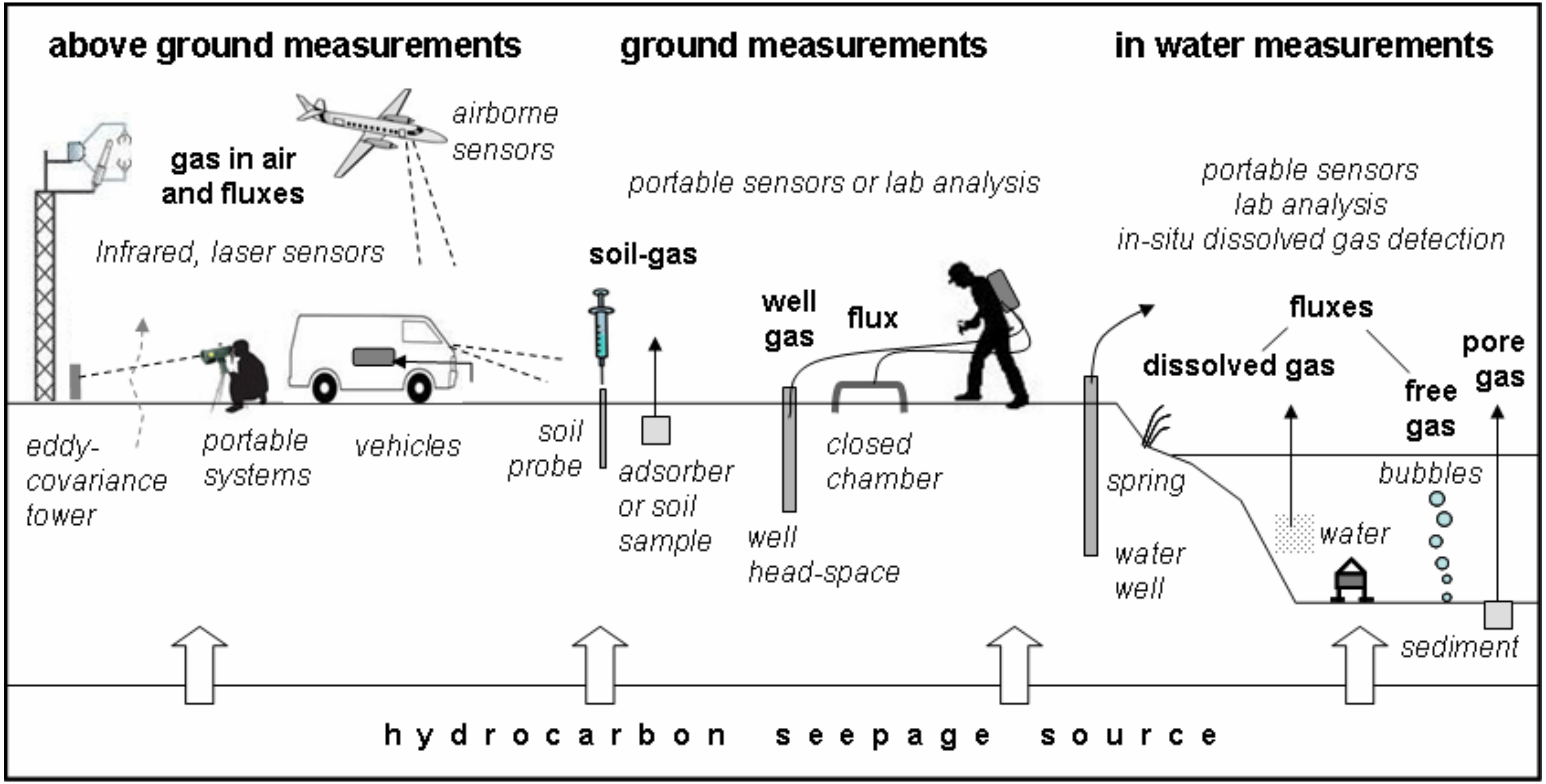
How to detect and measure gas seepage



CLOSED-CHAMBER METHOD FOR FLUX MEASUREMENT

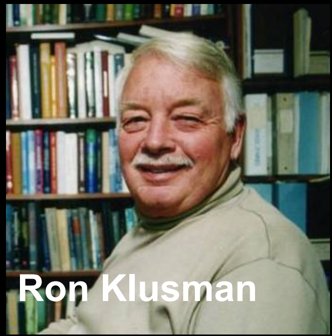
widely used for soil-respiration, gas fluxes from wetlands, rice paddies and permafrost

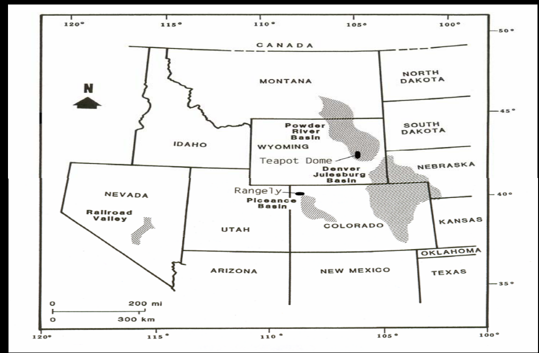
$$Q = \frac{V_{FC}}{A_{FC}} \cdot \frac{c_2 - c_1}{t_2 - t_1} \quad \left[\frac{mg}{m^2 * d}\right]$$

V_{FC} (m^3) chamber volume
A_{FC} (m^2) chamber area
c_1 - c_2 (mg/m^3) methane concentrations at times t_1 - t_2 (days).

Photo: Charlotte Sigsgaard

Closed-chambers used for seepage measurements



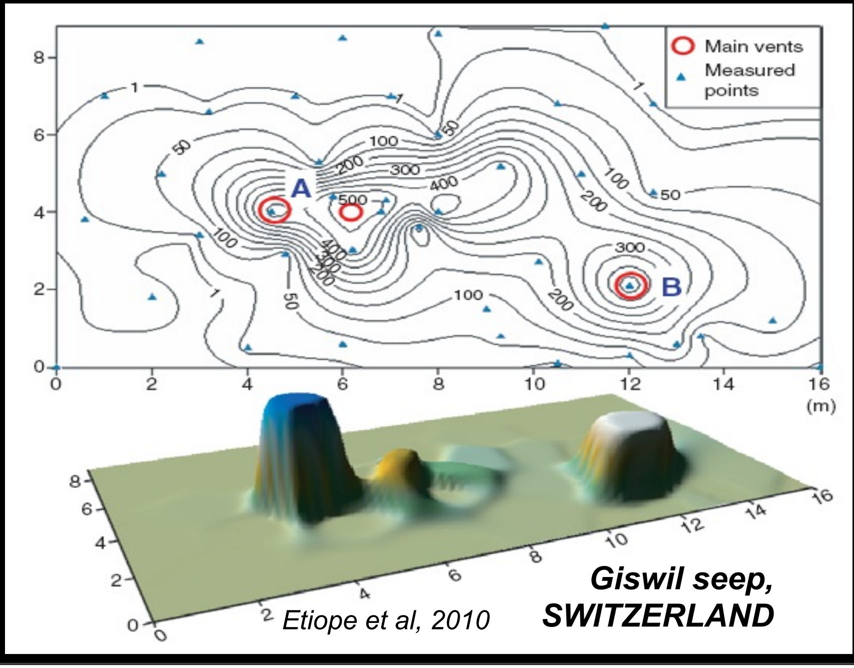


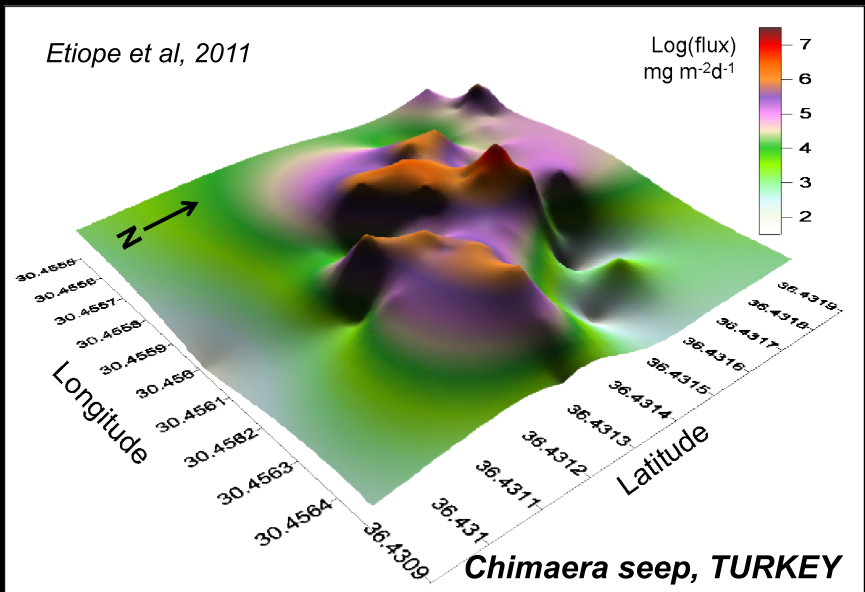
New York

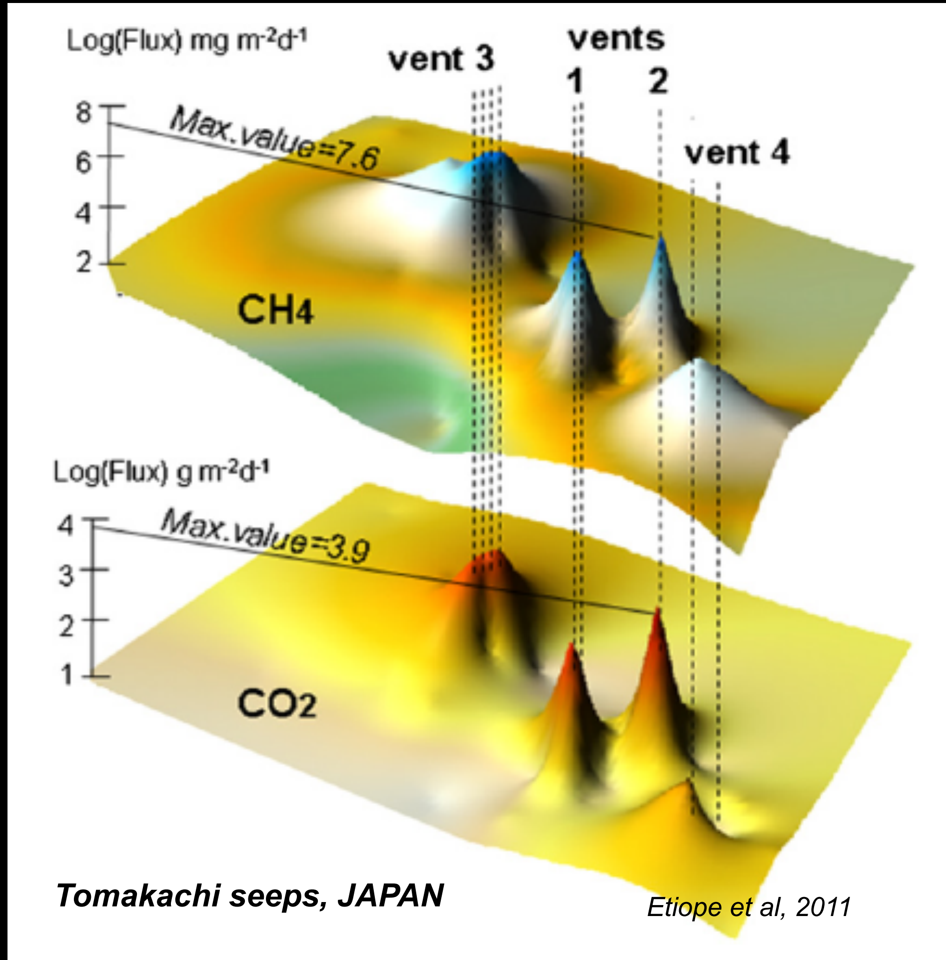
Italy

Last week at the
La Brea park, Los Angeles

Closed-chambers used for seepage measurements







New solutions for gas seepage investigation

based on a combination of a new generation of portable instruments for the measurement of a wide range of gases (methane, heavier hydrocarbons, CO_2, H_2, SO_2….)

- Tunable Diode Laser Adsorption Spectrometry (TDLAS)
- Fourier Transform Infra-Red spectrometry (FTIR)
- Semiconductors and infrared sensors
- Closed-chamber systems for gas flux measurements

A LARGE METHANE FLUX DATABASE

Table 6.1 A summary of the main studies, with methane flux measurements from land-based natural gas seepage (hydrocarbon-rich emissions)

Country	Type of seepage	Reference
Azerbaijan	Gas seeps, mud volcanoes, miniseepage	Etiope et al. (2004), Kopf et al. (2009)
Australia	Gas seeps	Day et al. (2013)
China (PRC)	Microseepage	Tang et al. (2010)
Germany	Microseepage	Thielemann et al. (2000)
Greece	Gas seeps and miniseepage	Etiope et al. (2006, 2013a)
Italy	Gas seeps, mud volcanoes, miniseepage, microseepage	Etiope et al. (2002, 2007a), Etiope and Klusman (2010)
Japan	Mud volcanoes, miniseepage	Etiope et al. (2011a)
Romania	Gas seeps, mud volcanoes, miniseepage	Etiope et al. (2004), Baciu et al. (2008), Spulber et al. (2010), Frunzeti et al. (2012)
Switzerland	Gas seep and miniseepage	Etiope et al. (2010)
Taiwan (ROC)	Mud volcanoes, gas seeps, miniseepage	Yang et al. (2004), Chang et al. (2010), Chao et al. (2010), Hong et al. (2013)
Turkey	Gas seeps, miniseepage, microseepage	Etiope et al. (2011b)
USA—Alaska	Gas seeps	Walter Anthony et al. (2012)
USA—California	Gas seeps	Duffy et al. (2007)
USA—Colorado	Seeps Microseepage	LTE (2007), Klusman and Jakel (1998), Klusman et al. (2000)
USA—Nevada	Microseepage	Klusman et al. (2000)
USA—Wyoming	Microseepage	Klusman et al. (2000)
USA—New York	Gas seep, miniseepage	Etiope et al. (2013b)

A LARGE FLUX DATABASE – Emission factors are today well known

Thousands of flux data from USA, EUROPE, ASIA…

30 onshore MV from Italy, Romania, Azerbaijan, Japan, Taiwan

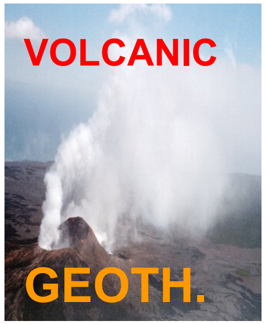
Typical EF 10^3 t km^{-2} y^{-1}

>80 gas seeps from 15 countries

Individual gas seeps (natural fires, bubbling pools) **10^0 to 10^3** t/y.

> 1000 data in different soil types and countries

Typically **10^1-10^2** mg m^{-2} d^{-1} (similar to wetlands),



Hundreds of direct or derived CH_4 fluxes in USA, Europe, Asia….

up to **10^2** t/y for individual vents

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Upscaling based on emission factor and activity

(EMEP/EEA 2009 Guidelines)

EEA Technical report | No 9/2009

EMEP/EEA air pollutant emission inventory guidebook 2009

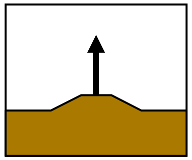
Technical guidance to prepare national emission inventories

ISSN 1725-2237

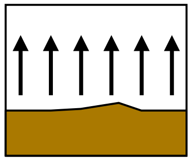
TOT. EMISSION = EMISS. FACTOR X

SEEPING AREAS

NUMBER OF SEEPS



Point sources (vents): ton/year



Area sources (with microseepage): t km^{-2} y^{-1} or mg m^{-2} d^{-1}

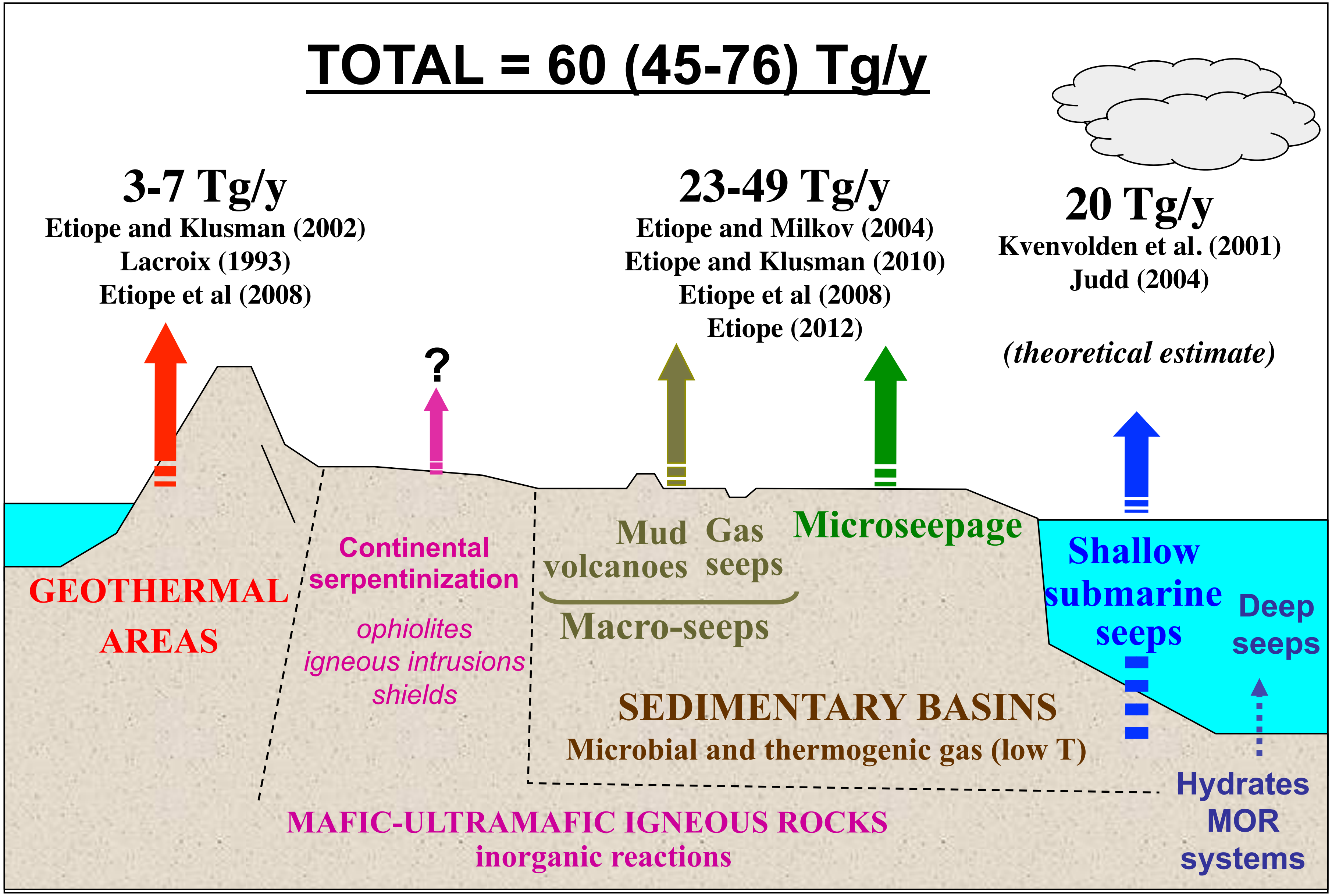
Same up-scaling procedures applied to
wetlands, rice fields, soil CO_2 respiration, soil CH_4 consumption…..

GLOBAL EMISSION ESTIMATES

Table 6.3 Global emissions of methane from geological sources

Type of seepage	($Tg\ yr^{-1}$)	References
Gas seeps	**3–4**	Etiope et al. (2008)
Mud volcanoes	5–10	Etiope and Klusman (2002)
	10.3–12.6	Dimitrov (2002)
	6–9	Etiope and Milkov (2004)
	10–20	Etiope et al. (2011a)
Microseepage	>7	Klusman et al. (1998)
	10–25	Etiope and Klusman (2010)
Marine seepage	18–48	Hornafius et al. (1999)
	10–30 (**20**)	Kvenvolden et al. (2001)
Geothermal/ volcanic areas	1.7–9.4	Lacroix (1993)
	2.5–6.3	*Volcanoes not included;* Etiope and Klusman (2002)
	<1	*Only volcanoes*; Etiope et al. (2008)
	2.2–7.3	This work (*only volcanoes would be around* 0.015 Tg/year)
Total	13–36	*Microseepage not included*; Judd (2004)
	30–70	Etiope and Klusman (2002)
	45	*Lowest microseepage estimate considered;* Kvenvolden and Rogers (2005)
	42–64	*Best estimate;* Etiope et al. (2008)
	30–80	*Extended range and top-down verification;* Etiope et al. (2008)
	45–76 (60)	Etiope (2012)

GEOLOGIC METHANE SOURCES



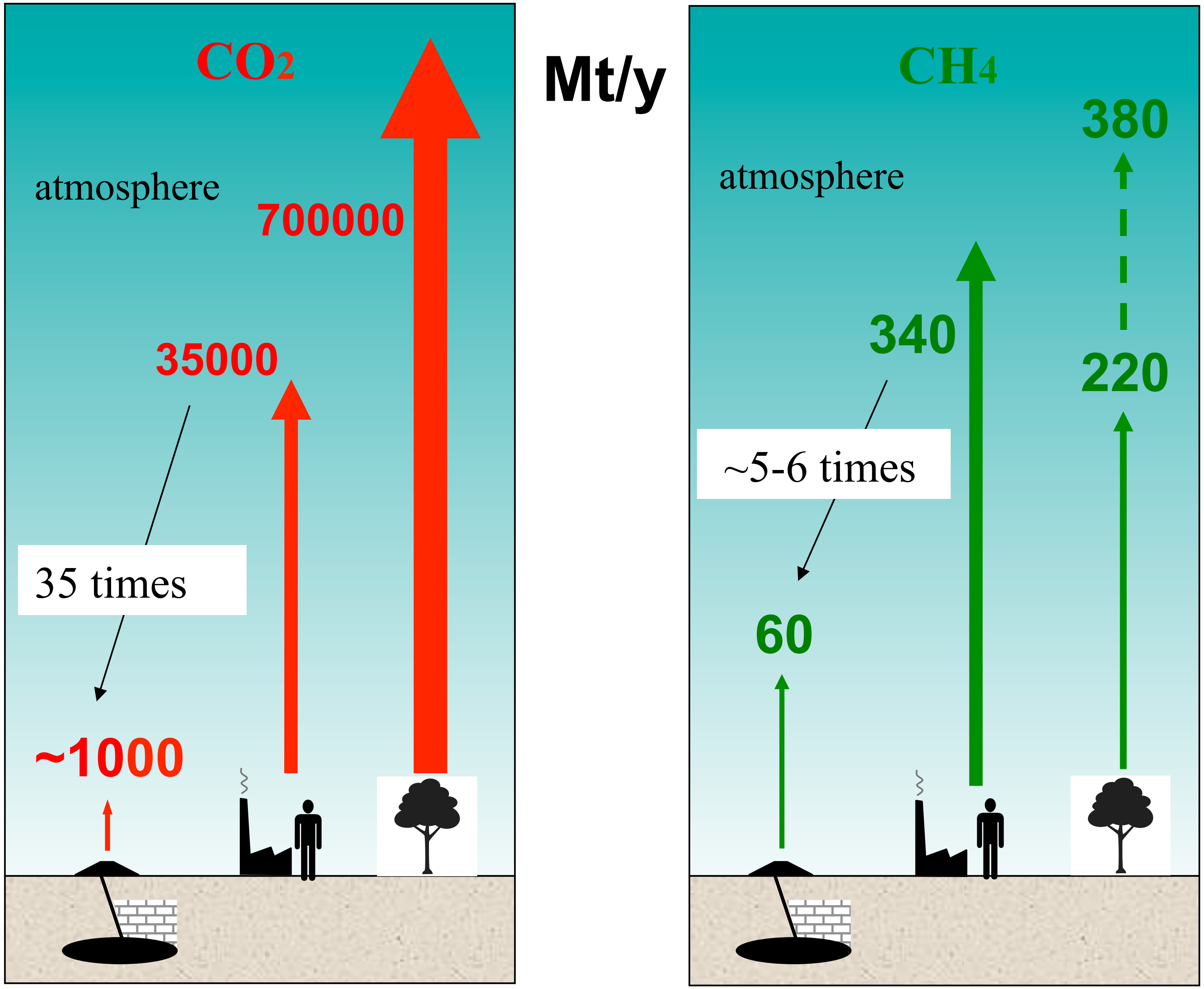
Geological CH_4

2^{nd} natural CH_4 source

≈10% of total CH_4 source

wastes

CO_2 and CH_4
geo-sources vs. man-made vs. natural



U N C E R T A I N T I E S

Tot. emission

=

Emission factor **x** **Area (or N. point sources)**

- MV eruptions

- Temporal variability (decadal, centuries…) due to endogenic factors (changes of pressure gradients, tectonics...)

limited knowledge for

- mud volcanoes
- submarine seeps (only theoretically estimated)
- microseepage area

Uncertainty is comparable or better than that of other natural sources (EMEP/EEA Guidebook 2009)

TOP-DOWN EMISSION ESTIMATES

Based on ^{14}C-free, fossil CH_4 in atmosphere

(*Etiope et al 2008; Lassey et al. 2007*)

^{14}C-free 30% of tot. emiss. (580 Tg/y ?) = 175 Tg/y

geo-CH_4 = 175 – fugitive (100?) = 75 Tg/y

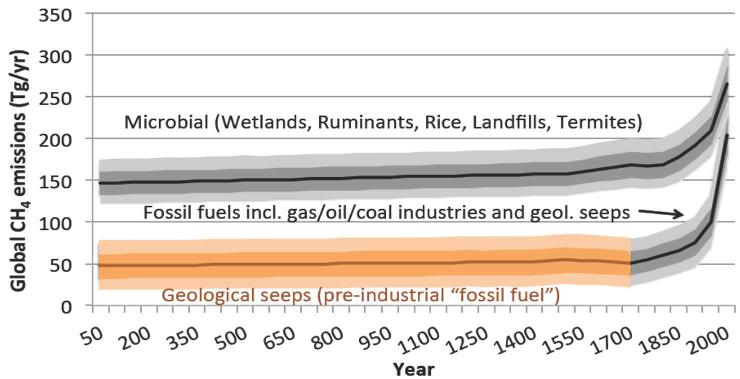
Atmospheric box models

Schwietzke et al (2016)

modeling based on two independent datasets:
(a) CH_4 and isotopic data from ice-core records
(b) current day atmospheric CH_4 and isotopic data

(a) suggests geo-CH4 : 30-70 (50) Tg/y

(b) suggests total fossil fuel (fugitive + geo): 200 Tg/y
If fugitive=150 Tg/y, geo-CH4= 50 Tg/y
If fugitive=100 Tg/y, geo-CH4=100 Tg/y



Overall geo-CH_4 emissions could be in the range 30-100 Tg/y.

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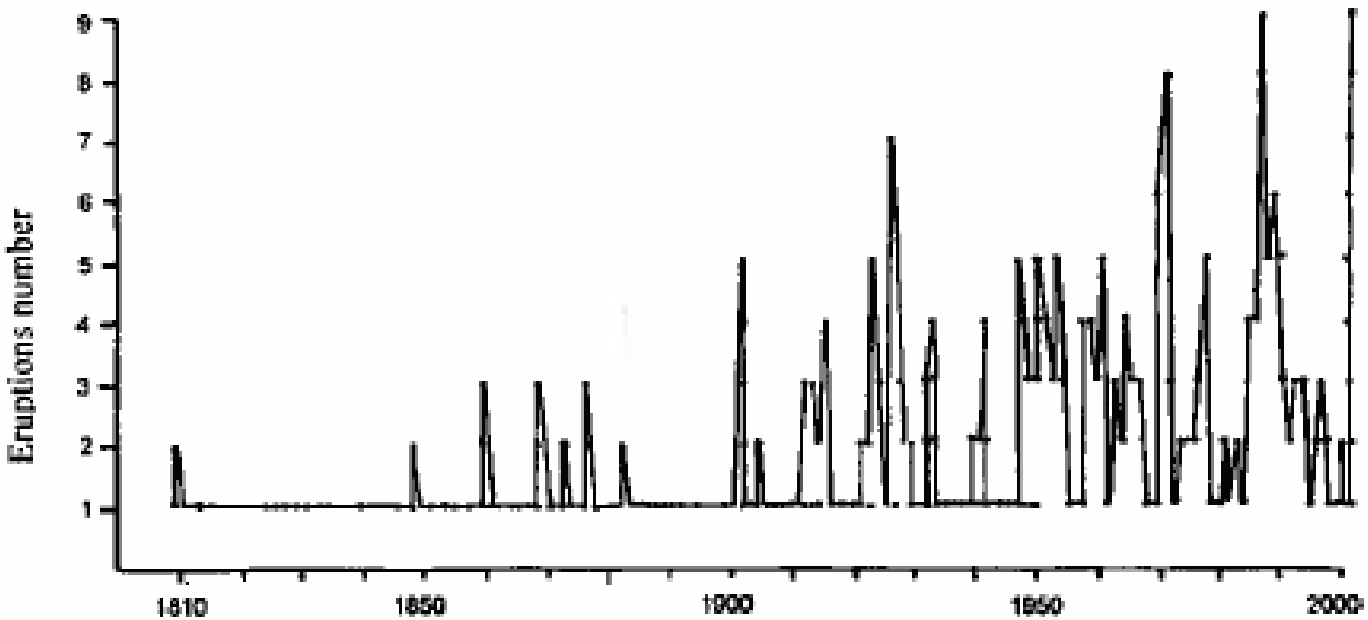
How to consider GEO-CH_4 emission for inverse studies?

Two misconceptions (wrong paradigm)

- Geo-CH_4 emissions are constant over time
- Geo-CH_4 is always ^{13}C enriched

Mud volcano eruptions over time



Aliyev et al (2002)

Fig. 13. Frequency of mud volcano eruptions of Azerbaijan.

Geo-CH_4 emissions are NOT constant over time

geologic CH_4 flux (advective process) depends on

k subsurface permeability

ΔP gas pressure gradient

$$F = Cv$$

$$v = k\frac{\Delta P}{\mu Z}$$

Fracturing

tectonics (long term)

seismicity (short term)

Fluid charge/discharge

deep gas migration pulses

natural gas extraction

hydrostatic (aquifer) variations

$$P_g > P_w + P_c$$

Enhanced degassing related to seismicity

It is well known that gas migration, seepage and, in particular, eruptions of mud volcanoes, can be stimulated by earthquakes, i.e. by the **passage of seismic waves or by coseismic changes in crustal stress and permeability** (e.g. Mellors et al., 2007; Manga et al., 2009; Mazzini and Etiope, 2017).

Reports of correlations between earthquakes and mud volcanoes eruptions are widespread

Examples:

Changes in Yellowstone geysers eruption behaviour after the 2002 M 7.9 Alaskan Denali earthquake (Husen et al., 2004)

Nirano MV (Italy) emissions after June 2013 M4.7 (Lupi et al., 2016)

Eruption of LUSI mud-gas flow (Indonesia) in March 2006, 2 days after 6.3 M earthquake (Mazzini et al, 2009)

Formation of the Kevachy MV island few hours after the September 2013 M7.7 Makran coast (Avouac et al., 2014)

Many eruptions of CH_4-rich mud volcanoes in Azerbaijan….

Examples of decadal changes of seismicity

Tectonophysics, 57 (1979)

Global variation of seismic activity

KIYOO MOGI

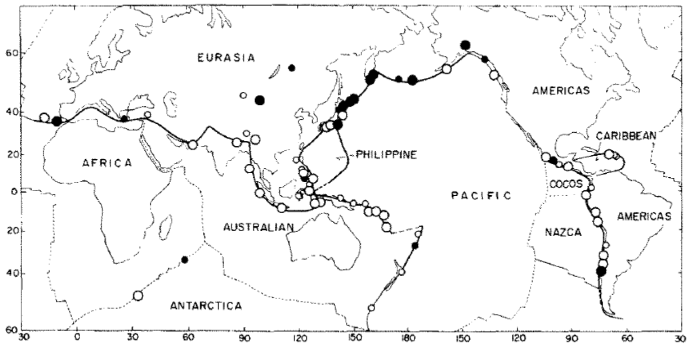


Fig. 2. Locations of great shallow earthquakes ($M \geq 7.8$, focal depth < 100 km) during the period from 1931 to 1970. Open circles: 1931—1950; solid circles 1951—1970. (after Mogi, 1974)

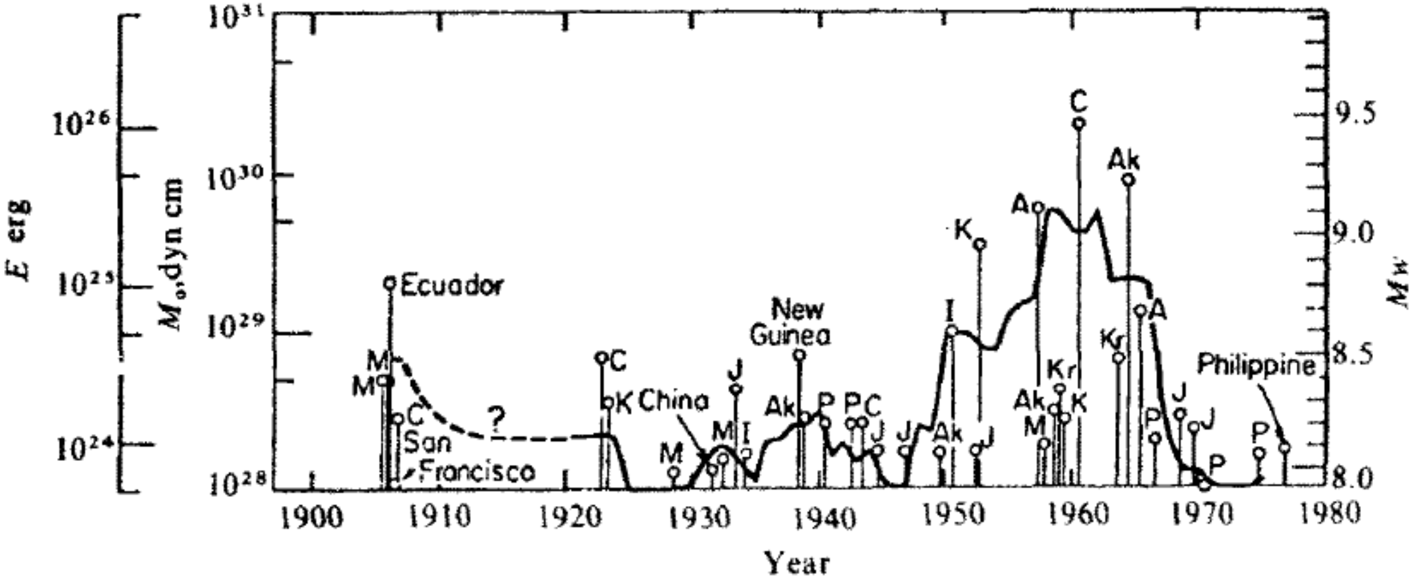


Fig. 1. The seismic moment M_0, seismic energy E and magnitude M_w of great shallow earthquakes. The vertical bars are for individual earthquakes. The solid curve shows the annual average of seismic energy obtained by taking an unlagged 5-year running average. (after Kanamori, 1978).

Geo-CH_4 emissions are NOT constant over time

geological CH_4 flux (advective process) depends on

k subsurface permeability

ΔP gas pressure gradient

$$F = Cv$$

$$v = k\frac{\Delta P}{\mu Z}$$

Fracturing

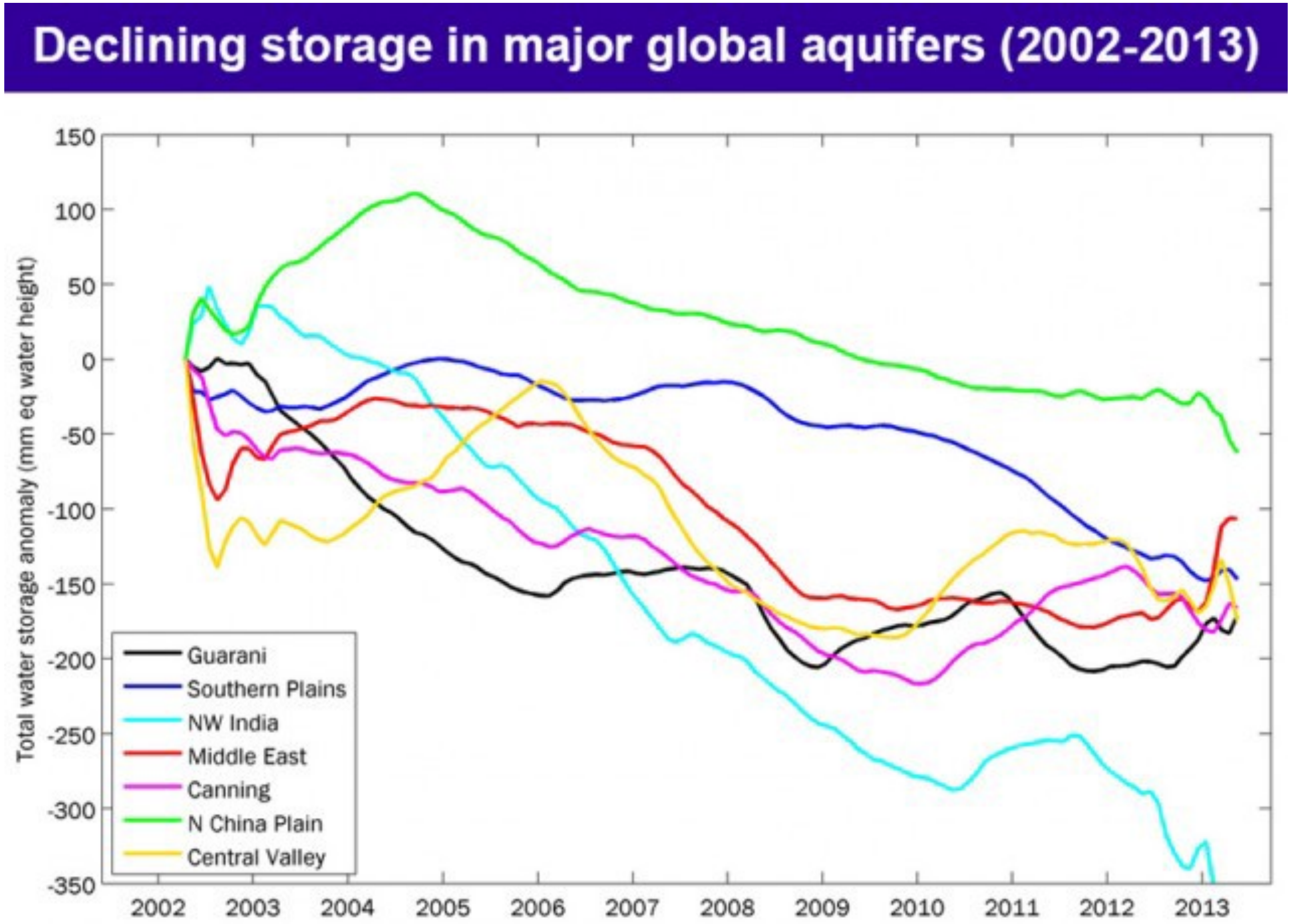
tectonics (long term)
seismicity (short term)

Fluid charge/discharge

deep gas migration pulses
natural gas extraction
hydrostatic (aquifer) variations

$$P_{\mathrm{g}} > \boxed{P_{\mathrm{w}}} + P_{\mathrm{c}}$$

Examples of decadal changes of hydrostatic pressure (groundwater depletion)

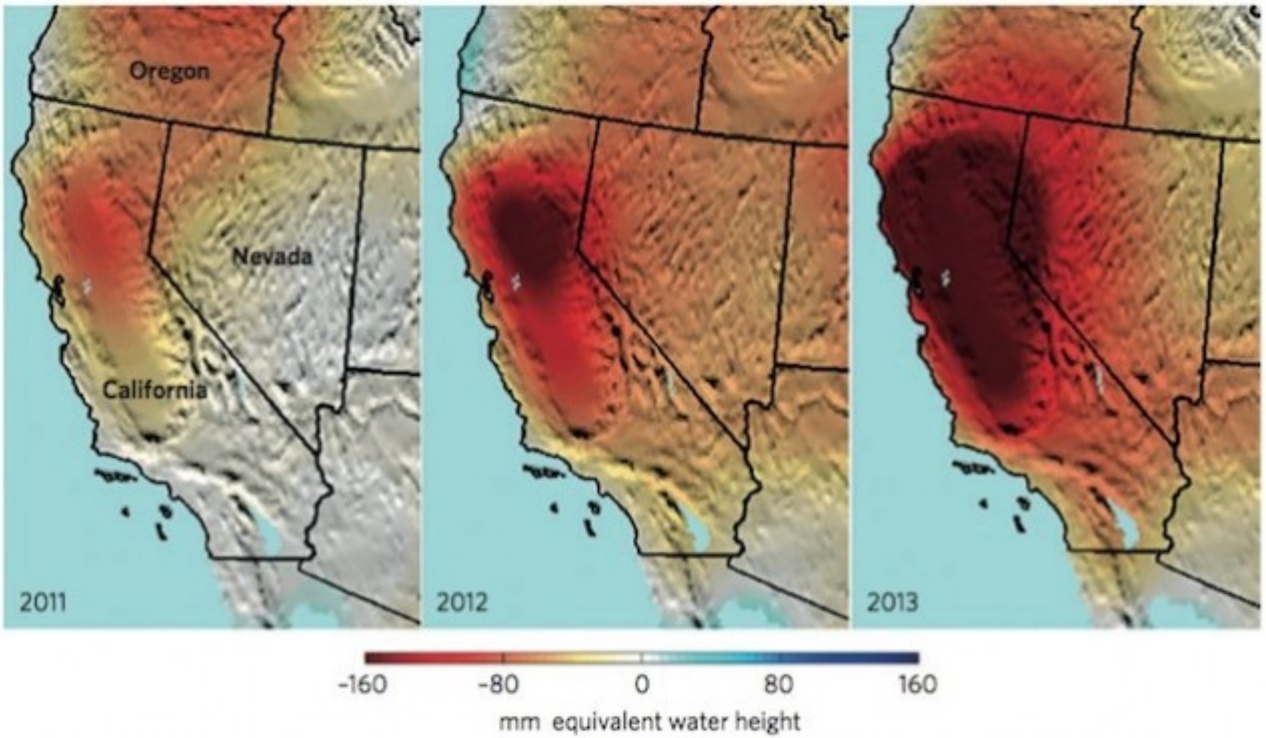


Famiglietti (2014; The global groundwater crisis. Nature Climate Change 4)

Examples of decadal changes of hydrostatic pressure (groundwater depletion)

higher hydrostatic pressure
=
lower gas flux

lower hydrostatic pressure
=
higher gas flux



Famiglietti (2014; The global groundwater crisis. Nature Climate Change 4)

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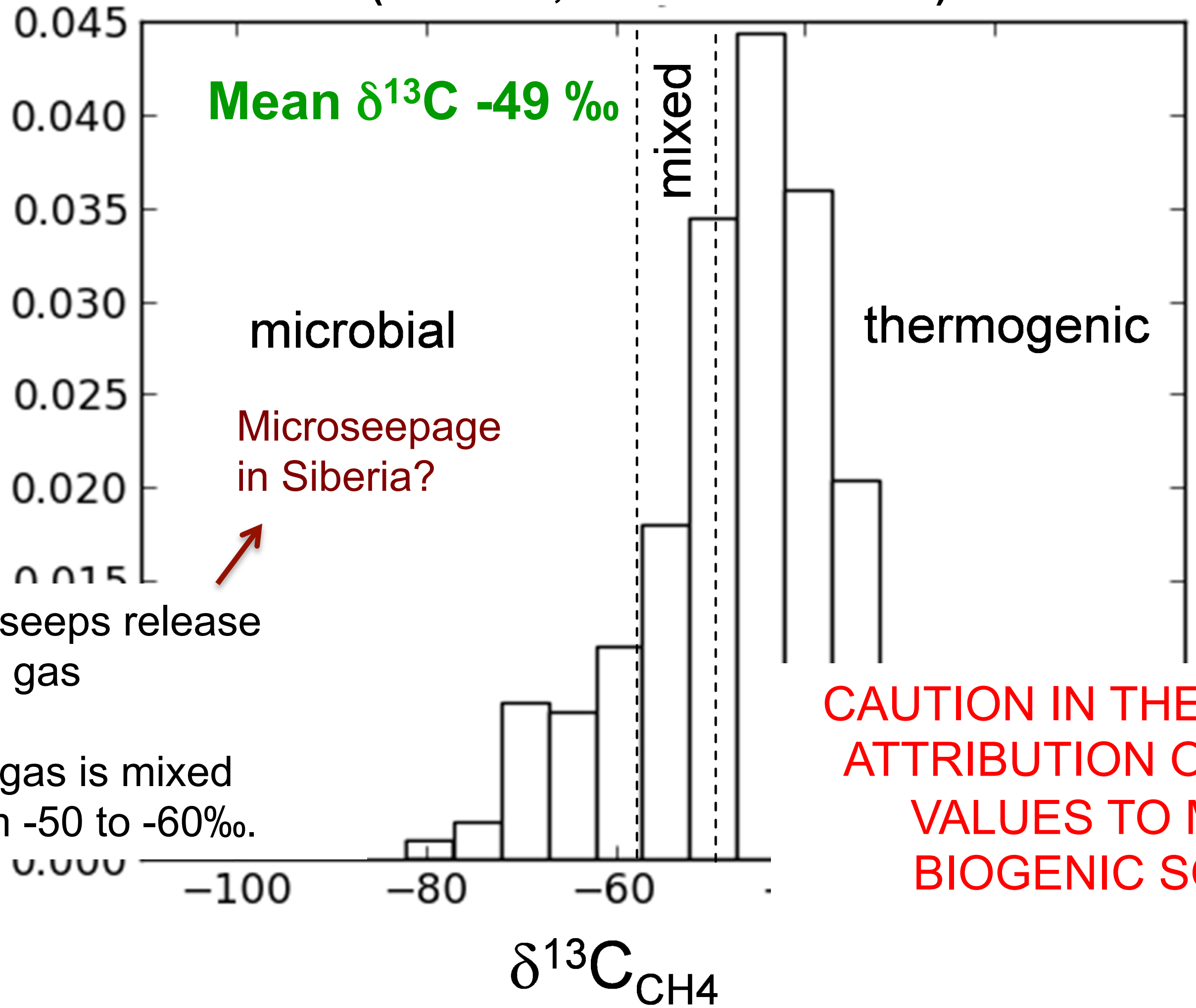
GEOLOGIC SOURCES ARE NOT ALWAYS ^{13}C-ENRICHED

GLOBAL STATISTICS OF SEEPING METHANE ORIGIN
(629 data, GLOGOS data-set)



≈10% of seeps release microbial gas

≈30% of gas is mixed
δ^{13}C from -50 to -60‰.

CAUTION IN THE EXCLUSIVE ATTRIBUTION OF LOW δ^{13}C VALUES TO MODERN BIOGENIC SOURCES

Geologic microbial methane sources (with "wetland type" $\delta^{13}C$)

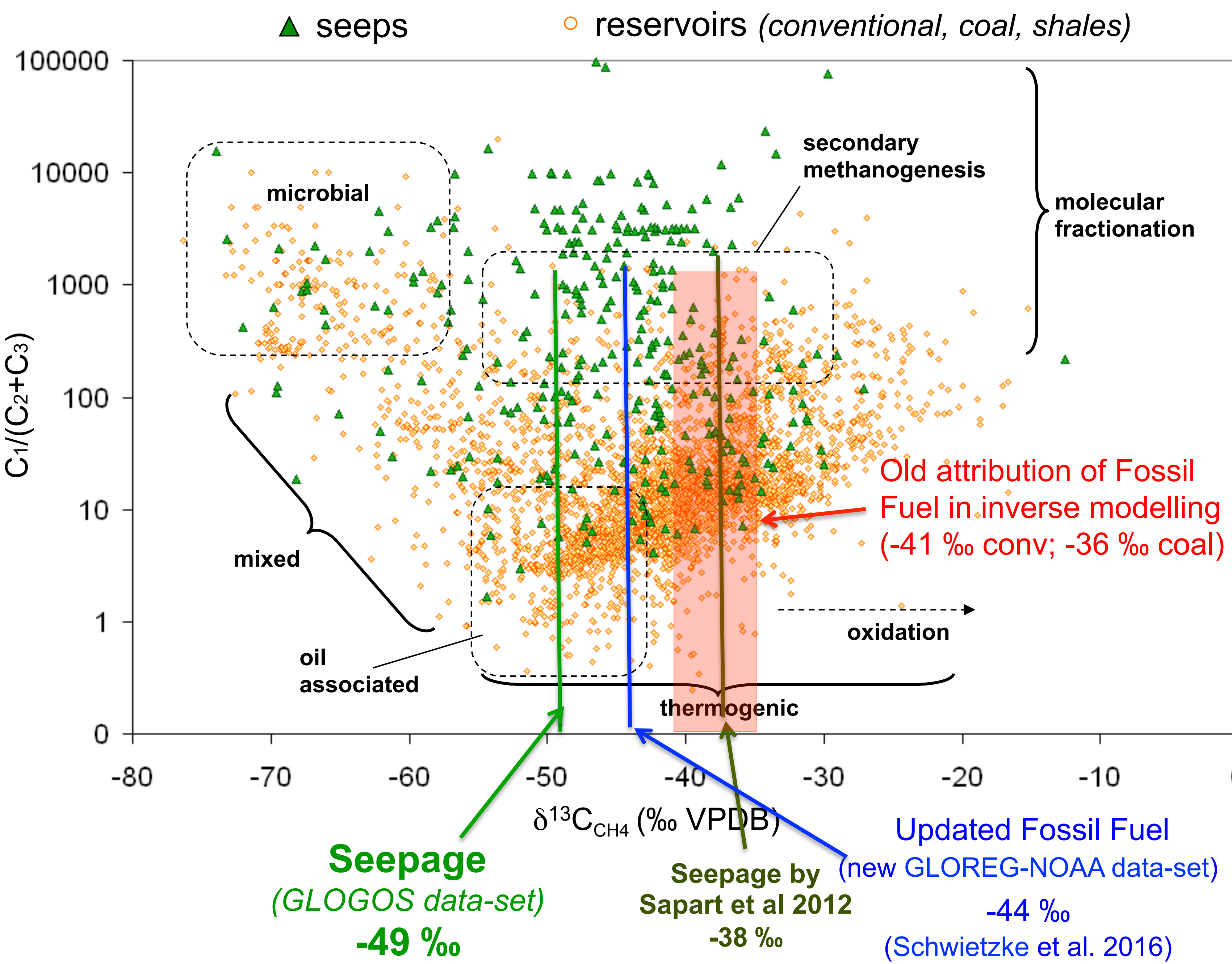
$\delta^{13}C < -60$ ‰

Geologic microbial methane sources (with “wetland type” $\delta^{13}C$)

$\delta^{13}C < -60$ ‰

Transylvania seeps

Porsugel mud volcano (Turkmenistan)



seeps
reservoirs (conventional, coal, shales)
100000
10000
1000
100
10
1
0
C1/(C2+C3)
-80
-70
-60
-50
-40
-30
-20
-10
0
δ13CCH4 (‰ VPDB)
microbial
secondary methanogenesis
molecular fractionation
mixed
oil associated
thermogenic
oxidation
Old attribution of Fossil Fuel in inverse modelling (-41 ‰ conv; -36 ‰ coal)
Seepage
(GLOGOS data-set)
-49 ‰
Seepage by Sapart et al 2012
-38 ‰
Updated Fossil Fuel
(new GLOREG-NOAA data-set)
-44 ‰
(Schwietzke et al. 2016)

1. Geologic methane sources: definitions, classifications (what and where)

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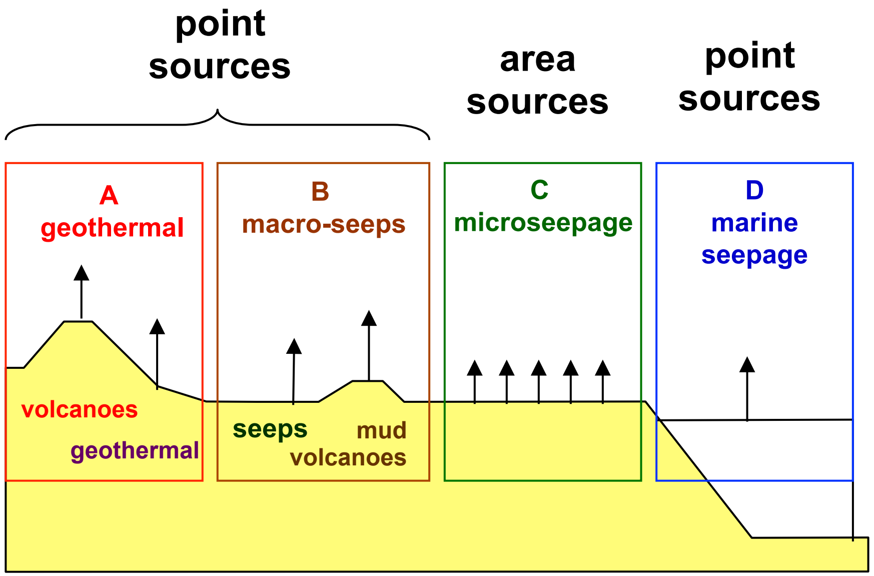
6. Geo-emissions in the official inventories (and the fake gas hydrate story)

7. Abiotic methane: a newly discovered geo-source

Improving emission estimates: revision of emission factors and activity (2017-2018)



Revision and development of data-sets for each category of geo-sources

1. **Emission factors:** - accurate analysis of seep size and intensity (gas, seeps, mud volcanoes)
   - new microseepage flux data

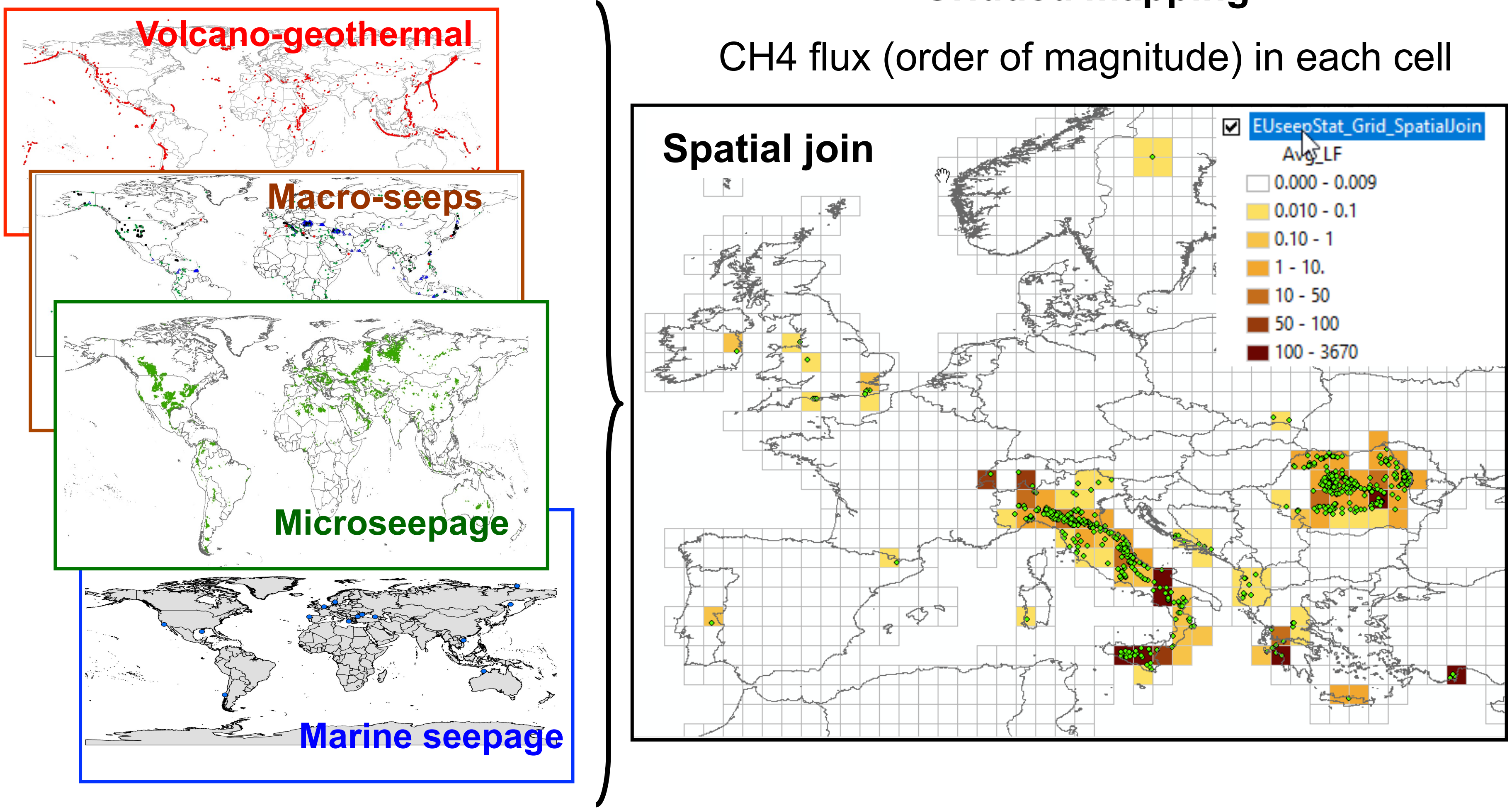
2. **Activity** (number of seeps and seepage area); developing accurate microseepage maps

3. **Gridded maps** for inverse studies → NASA-IDS project (2017-2019) coordinated by Stefan Schwietzke (CIRES-Colorado Un-NOAA)

Process-level investigation of revised global methane budget based on in situ and remote sensing of atmospheric composition and the land surface

Gridded mapping

CH4 flux (order of magnitude) in each cell



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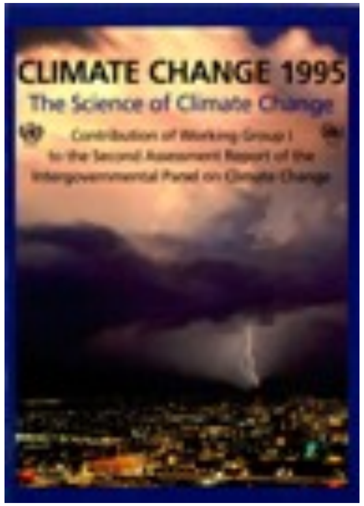
Evolution of the IPCC Assessment Reports

1990 AR1

1995 AR2



2001 AR3

2007 AR4

2013 AR5

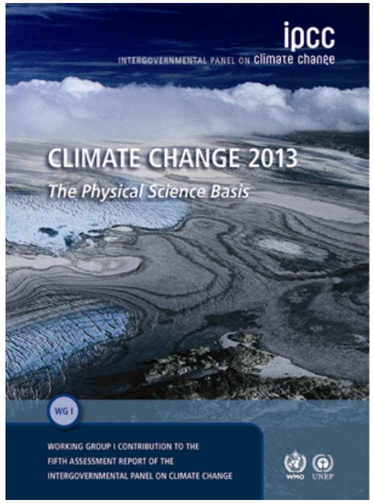


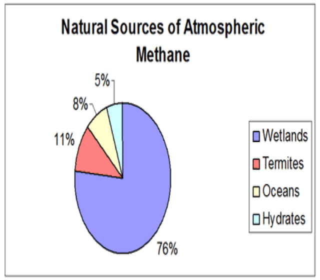
Table 1 2 Estimated Sources and Sinks of Methane

Source	Annual Release (Tg CH_4)
Natural Wetlands (bogs, swamps, tundra, etc)	115
Rice Paddies	110
Enteric Fermentation (animals)	80
Gas Drilling, venting, transmission	45
Biomass Burning	40
Termites	40
Landfills	40
Coal Mining	35
Oceans	10
Freshwaters	5
CH_4 Hydrate Destabilization	5

NO GEOLOGICAL SOURCES

NO DISTINCTION OF SOURCES

NO GEOLOGICAL SOURCES



NO GEOLOGICAL SOURCES

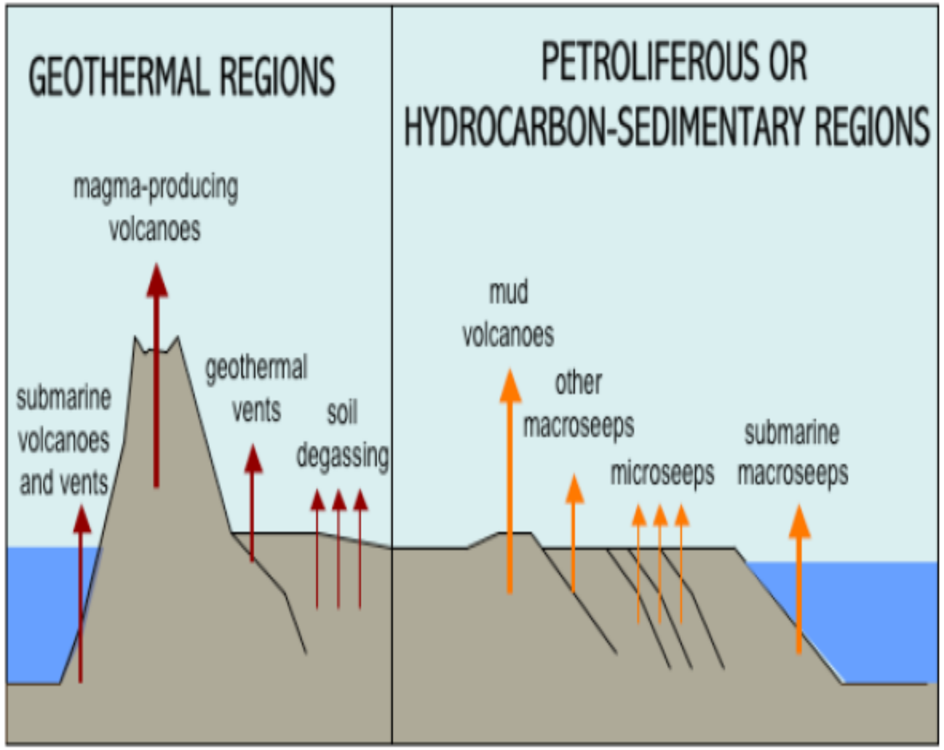
GEOLOGICAL ADDED, BUT ONLY OLD/ODD NUMBERS FROM MARINE SEEPAGE

RECENT GEOLOGICAL VALUES ENDORSED

EPA United States Environmental Protection Agency
Office of Atmospheric Programs (6207J)
Washington, DC 20460
EPA 430-R-10-001
April 2010

Methane and Nitrous Oxide Emissions From Natural Sources

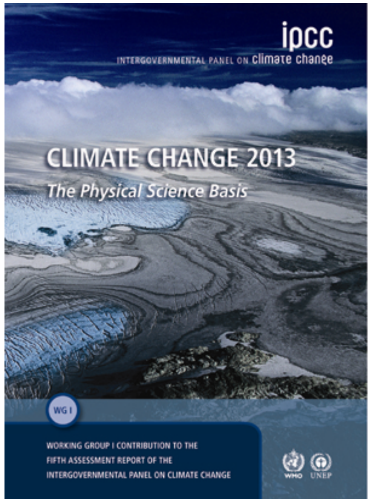
Chapter 8. Terrestrial and Marine Geologic Sources



Source	Methane ($Tg\ CH_4$/year)		
	Emissions Estimate[a]	Range[b]	$\delta^{13}C$ (‰)[c]
Wetlands	**170.3**		
- Northern/bogs	42.7	24–72	-62
- Tropical/swamps	127.6	81–206	-58.9
Upland soils and riparian areas	-30	Not available	
Oceans, estuaries, and rivers	9.1	2.3–15.6	-58
Permafrost	0.5	0–1	
Lakes	30	10–50	-53.8
Gas hydrates		2–9[e]	-62.5
Terrestrial and marine geologic sources		42-64	-41.8
Wildfires		2–5	-25
Vegetation		Not a source or 20–60	Not available
Terrestrial arthropods[f]	20	2–22	-63
Wild animals	8	2–15	-60.5

[e] The emission estimates for gas hydrates correspond to the flux of methane to the ocean, most of which is likely to be oxidized in the ocean water column.

2013
AR5



GAS HYDRATES….

FICTIONAL NUMBERS

Ciais et al (2013)

?

GAS-HYDRATES MISQUOTATION and SPECULATION

IPCC (2013) : 6 (2-9) Tg y^{-1}

IPCC (2007) 4-5

Dickens (2003)
1 page comment of a book –
NO NUMBERS

IPCC (2001) 5-10

Lelieveld et al. (1998) 5-15
permafrost+hydrates+other geologic sources

Fung et al. (1991)

EPA (1993) 5
only continental hydrates (not oceanic)

Cicerone & Oremland (1988)
5

IPCC (1992) 5

Kvenvolden (1991) 3-4

Nobody has ever measured or experimentally estimated the emissions

FICTIONAL NUMBERS, NO EXPERIMENTAL DATA!

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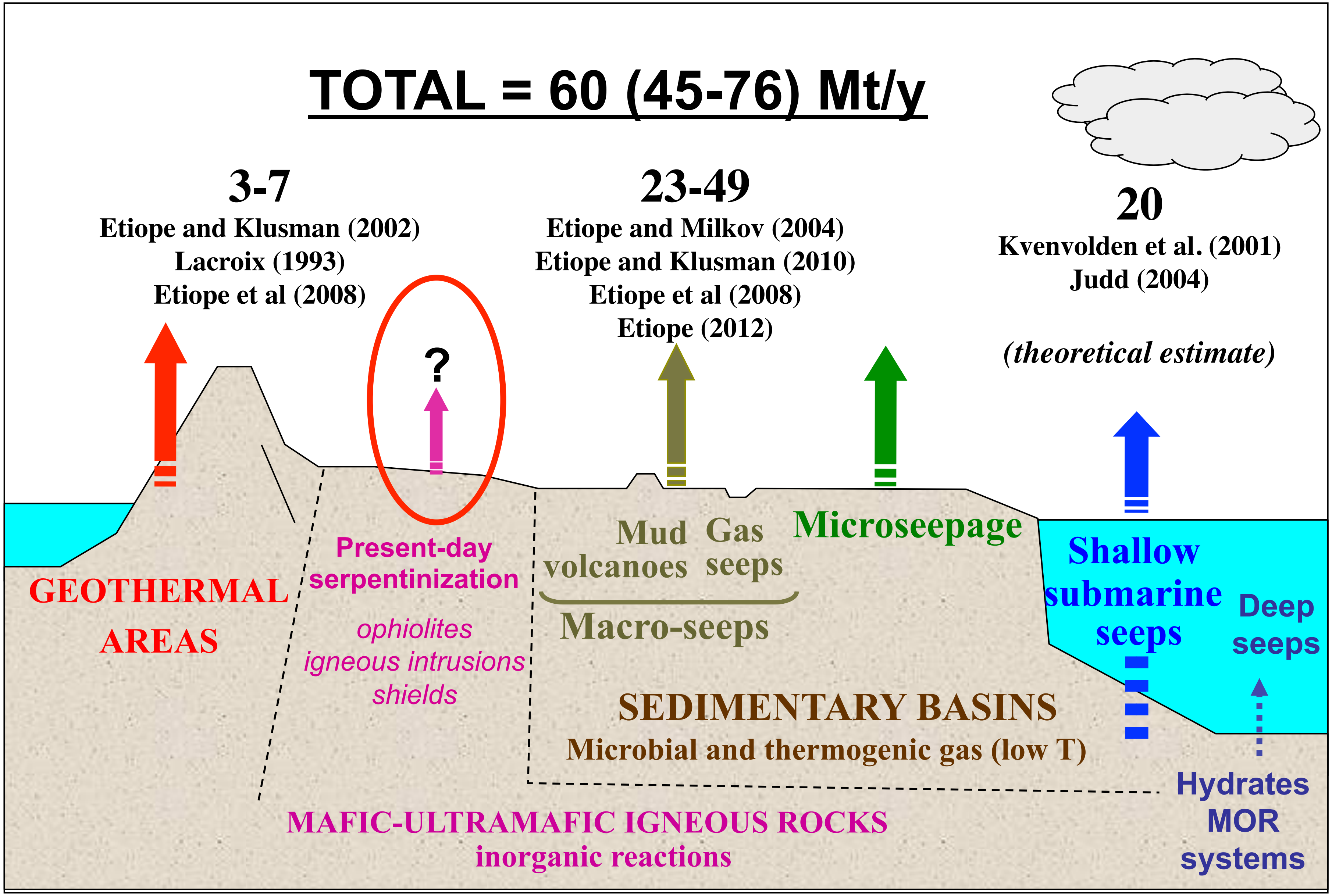
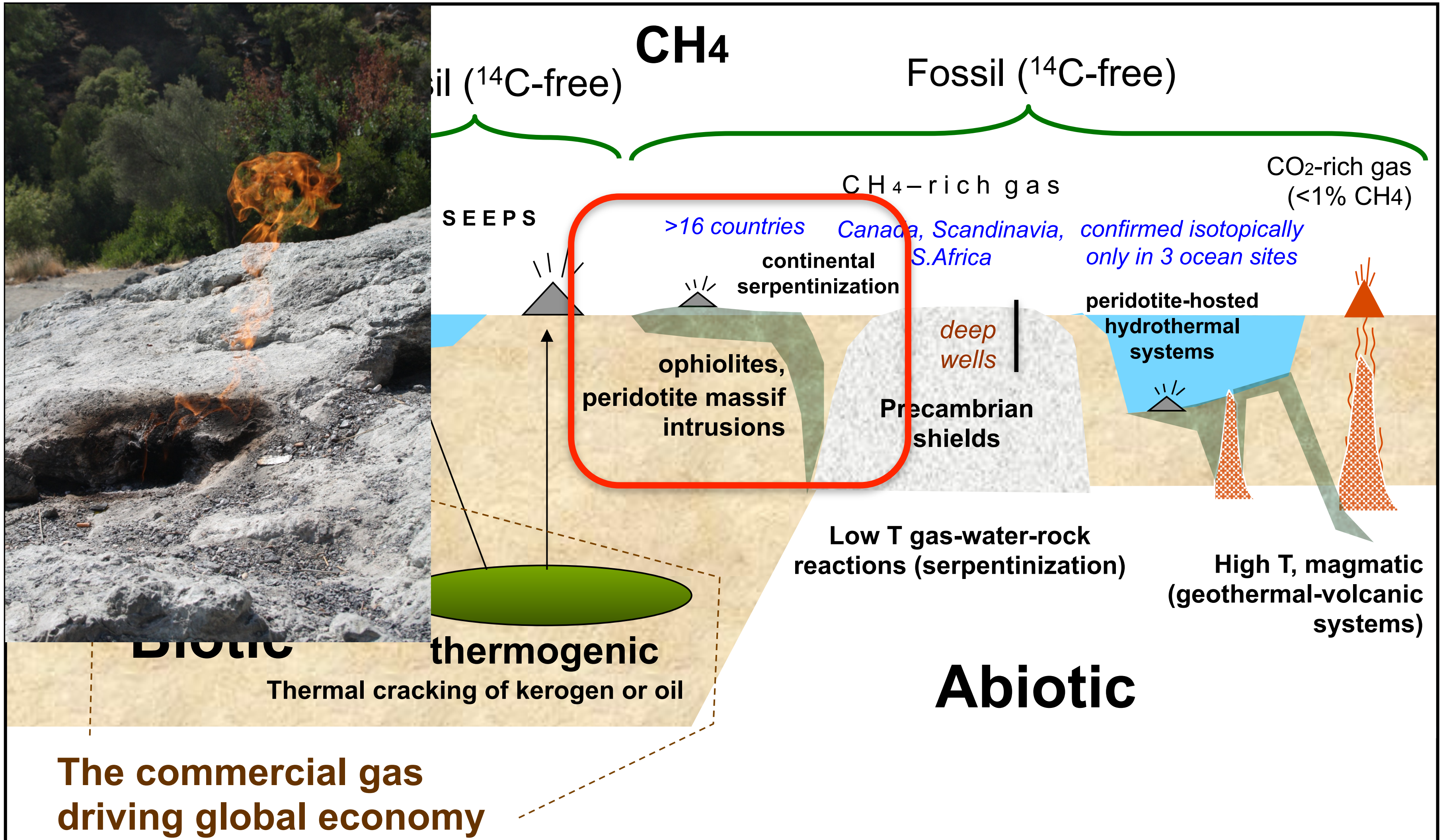
GEOLOGIC METHANE SOURCES



Methane origin on Earth



the largest abiotic CH_4 amounts observed in surface seeps and boreholes are produced in the crust, not in the mantle

Abiotic methane…not so rare

Reviews of Geophysics, 51 / 2013

ABIOTIC METHANE ON EARTH

Giuseppe Etiope[1,2] and Barbara Sherwood Lollar[3]

Elements
An International Magazine of Mineralogy, Geochemistry, and Petrology

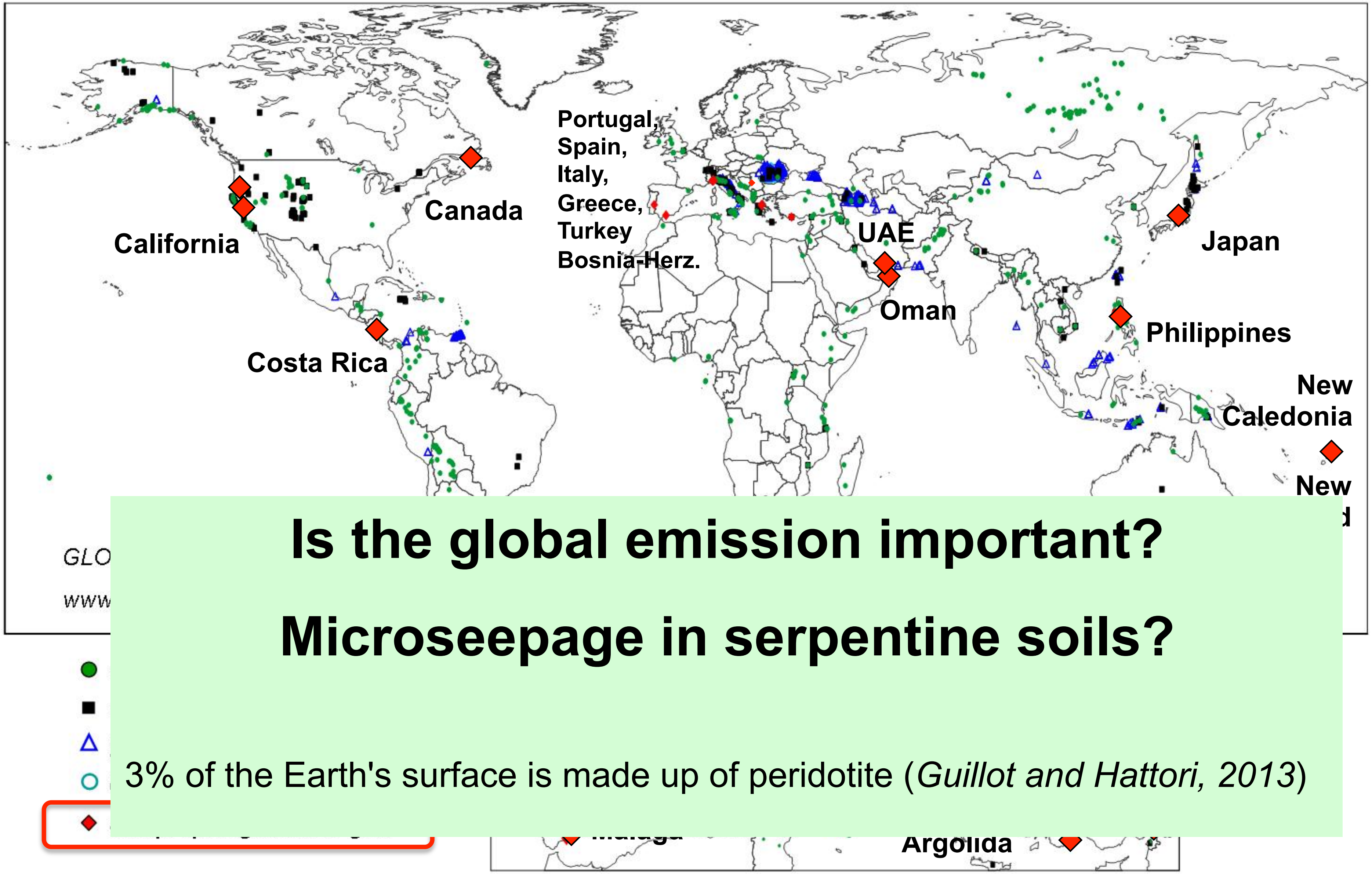
Abiotic Gas:
Atypical, But Not Rare

Giuseppe Etiope[1,2] and Martin Schoell[3]

Table 7.1 A list of land-based serpentinisation sites where methane seepage (or transport by hyperalkaline waters) has been documented (updated December, 2014)

Country	Site	Setting	References
Methane C–H isotopes determined			
Greece	Othrys Mt. (Archani, Ekkara)	Ophiolite	Etiope et al. (2013b)
Italy	Voltri-Genova (e.g., Acquasanta)	Ophiolite	Boschetti et al. (2013)
Japan	Hakuba-Happo	Orogenic massif	Suda et al. (2014)
New Zealand	Poison Bay	Ophiolite	Lyon et al. (1990)
New Zealand	Red Hills, Dun Mountain	Ophiolite	Pawson et al. (2014)
Oman	Semail (e.g., Al Khaoud, Nizwa)	Ophiolite	Fritz et al. (1992), Boulart et al. (2013)
Philippines	Zambales (Los Fuegos Eternos)	Ophiolite	Abrajano et al. (1988)
Portugal	Cabeço de Vide	Igneous intrusion	Etiope et al. (2013c)
Spain	Ronda peridotites	Orogenic massif	Etiope et al. (2014)
Turkey	Chimaera	Ophiolite	Etiope et al. (2011b)
Turkey	Amik Basin (Kurtbagi, Tahtakopru)	Ophiolite	Yuce et al. (2014)
U.A.Emirates	Al Farfar	Ophiolite	Etiope et al. (2015)
Incomplete or missing C–H isotope analyses			
Canada	Tablelands	Ophiolite	Szponar et al. (2013)
Costa Rica	Santa Elena	Ophiolite	Sanchez-Murillo et al. (2014)
New Caledonia	Prony Bay Fiordland	Ophiolite	Monnin et al. (2014)
Norway	Leka	Ophiolite	Okland et al. (2012)
Philippines	Zambales (Manleluag)	Ophiolite	Meyer-Dombard et al. (2013)
Philippines	Palawan (Brooke's Point)	Ophiolite	Meyer-Dombard et al. (2013)
Serbia	Zlatibor	Ophiolite	Milenic et al. (2009)
US—California	The Cedars	Ophiolite	Morrill et al. (2013)

SEEPAGE OF ABIOTIC METHANE



Is the global emission important?

Microseepage in serpentine soils?

3% of the Earth's surface is made up of peridotite (*Guillot and Hattori, 2013*)

TAKE HOME MESSAGE

Geo-CH_4: **2nd most important CH_4 natural source (~10%)**
(2nd source in absolute in pre-anthropogenic times)

Bottom-up estimates
- **confirmed by inverse modelling**
- **updates and improvements in progress**
- **more geo-marine flux data are needed**

Not constant over time (decadal changes possible)

Not always ^{13}C-enriched
(partially overlapping with wetlands)

Abiotic CH_4 emission unknown (to be added)

Thanks