

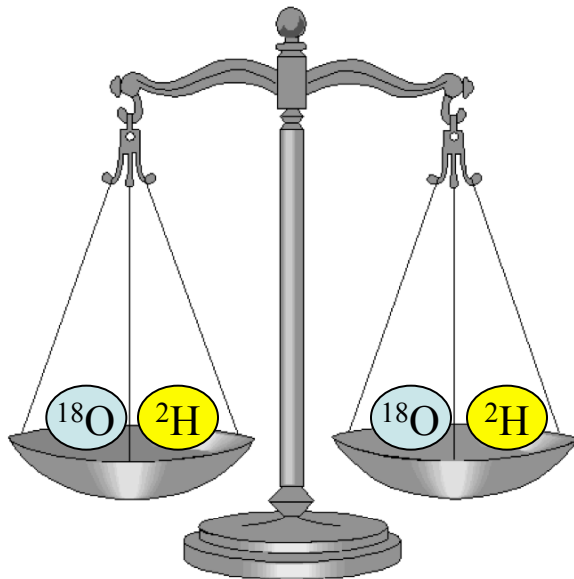
# Determining Geographic Origins of Foods Using Stable Isotope Ratios

James Ehleringer and Lesley Chesson

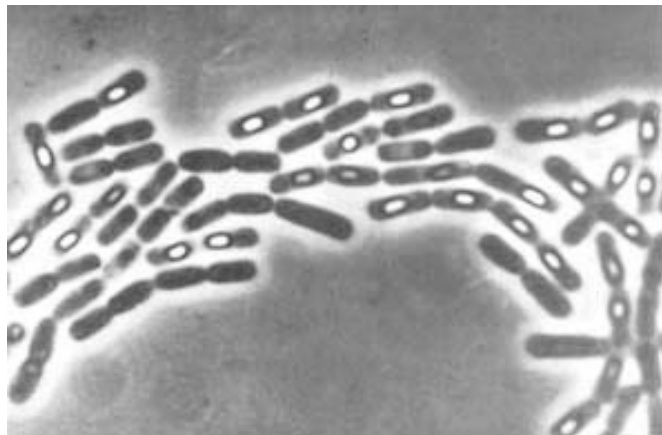
University of Utah & IsoForensics Inc.  
Salt Lake City, Utah



**IsoForensics**  
geo-location services



# Thesis: geographical information is recorded in organic matter



# There are multiple approaches to relate biology and geography

## Stable isotopes of light elements

- biological-environmental interactions
- hydrological- and climate-based variations

## Stable isotopes of heavy elements

- soil surface based variations

## Elemental composition

- soil surface based variations

## Genetic composition

- adaptive based variations
- breeding based variations

# Applications of isotope analyses at natural abundance levels

## Hydrogen and oxygen isotopes

- geography of water and water-based products
- geography of protein foods (e.g., meats, seeds)
- geography of carbohydrate foods (e.g., honey, flour)
- geography of lipid-based foods (e.g., oils, waxes)
- adulteration of beverages and juices

## Carbon isotopes

- food sources for animals (e.g., C3 versus C4)
- adulteration of foods, beverages, juices, spirits

## Nitrogen isotopes

- nitrogen sources for plants (i.e., fertilizers)
- food sources for animals

## Sulfur isotopes

- inland versus coastal geography
- sulfur sources for plants (i.e., fertilizers)
- food sources for animals



# Stable isotope analyses are increasingly useful in forensic studies



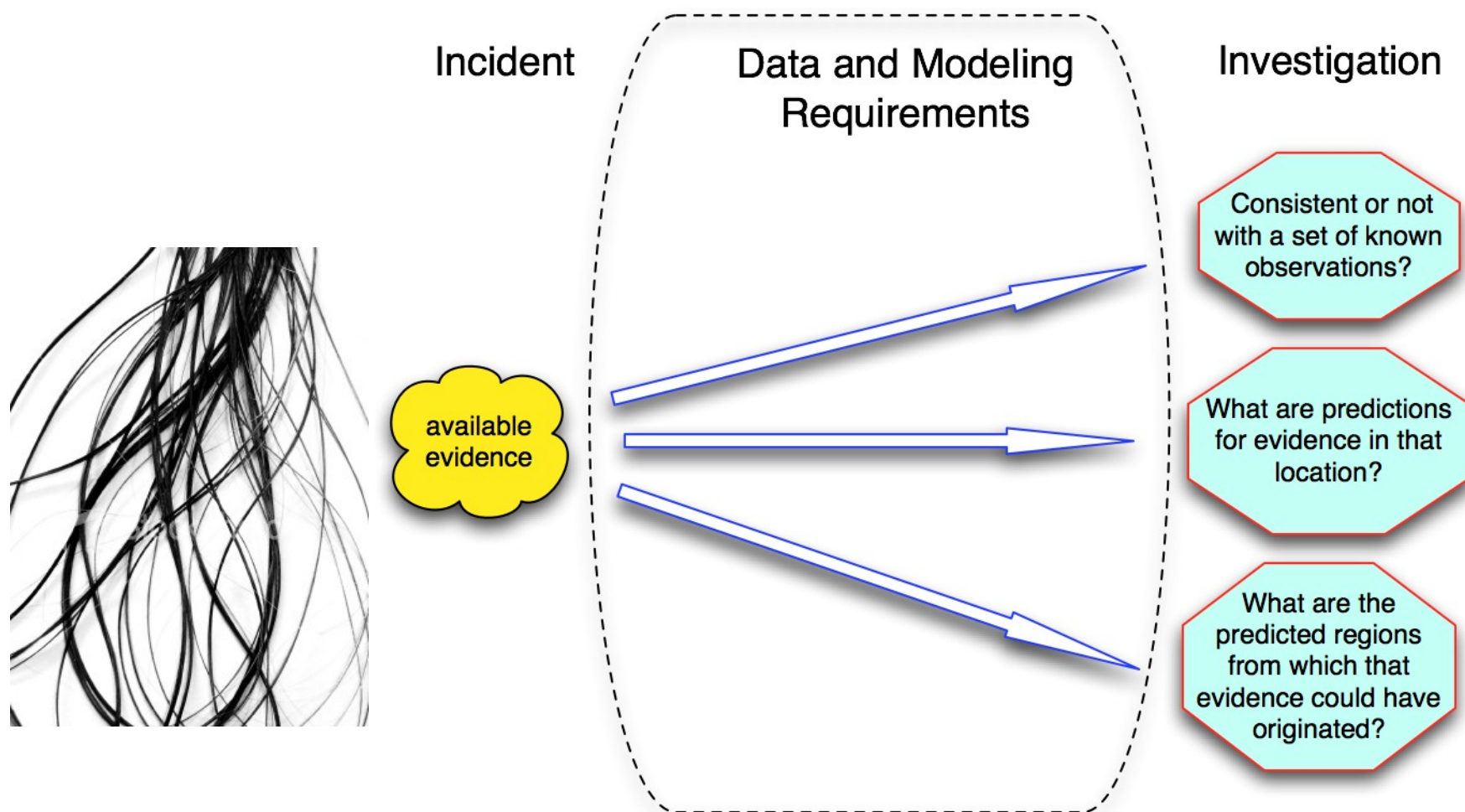
Floyd Landis is  
guilty of drug use

“Saltair Sally”

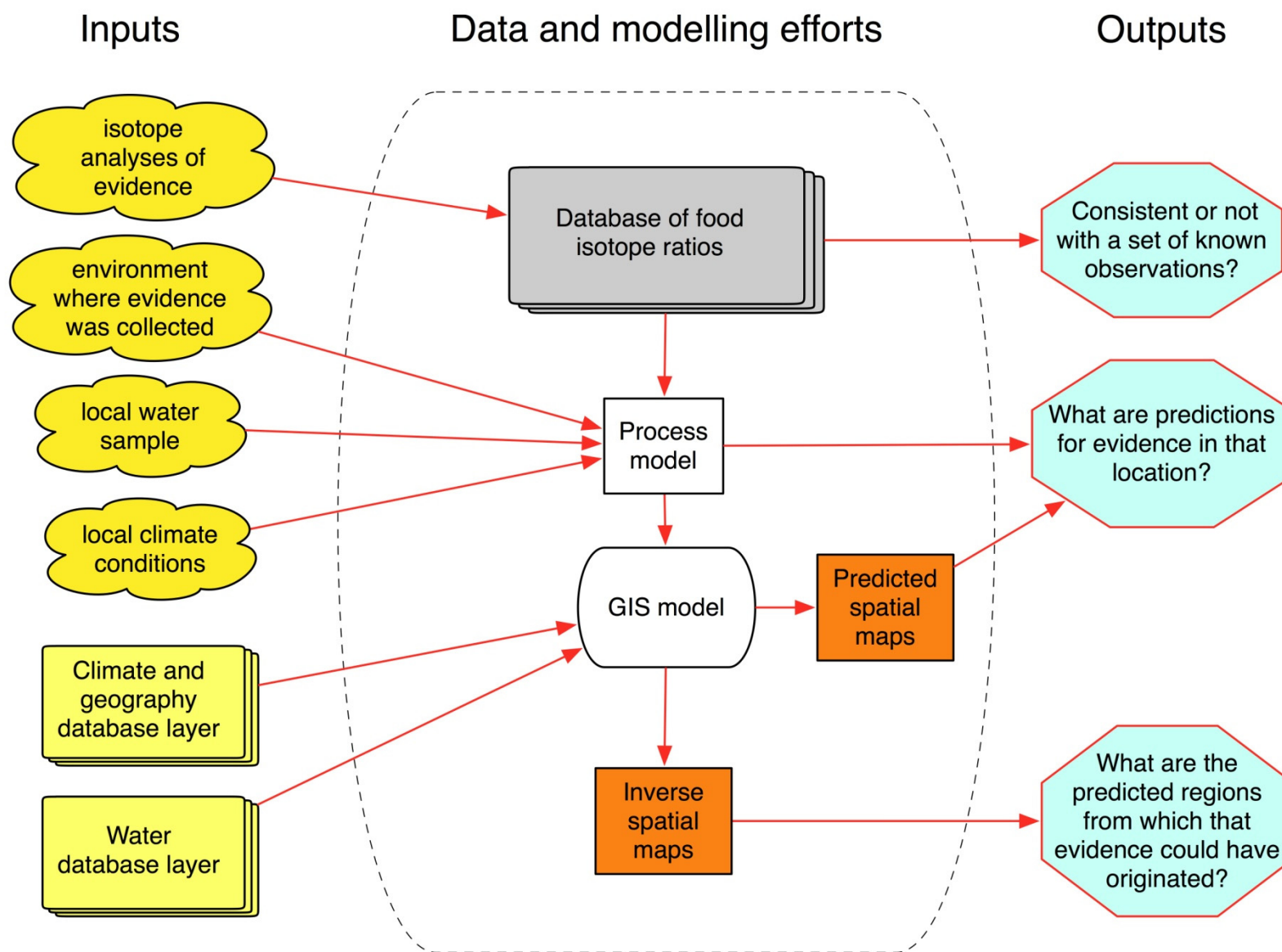


Pong Su, North Korea ship,  
found loaded with heroin

. . . where the objective is linking evidence to potential sources



# Our approach to addressing the regions-of-origin



# Stable isotopes are multiple forms of the same element

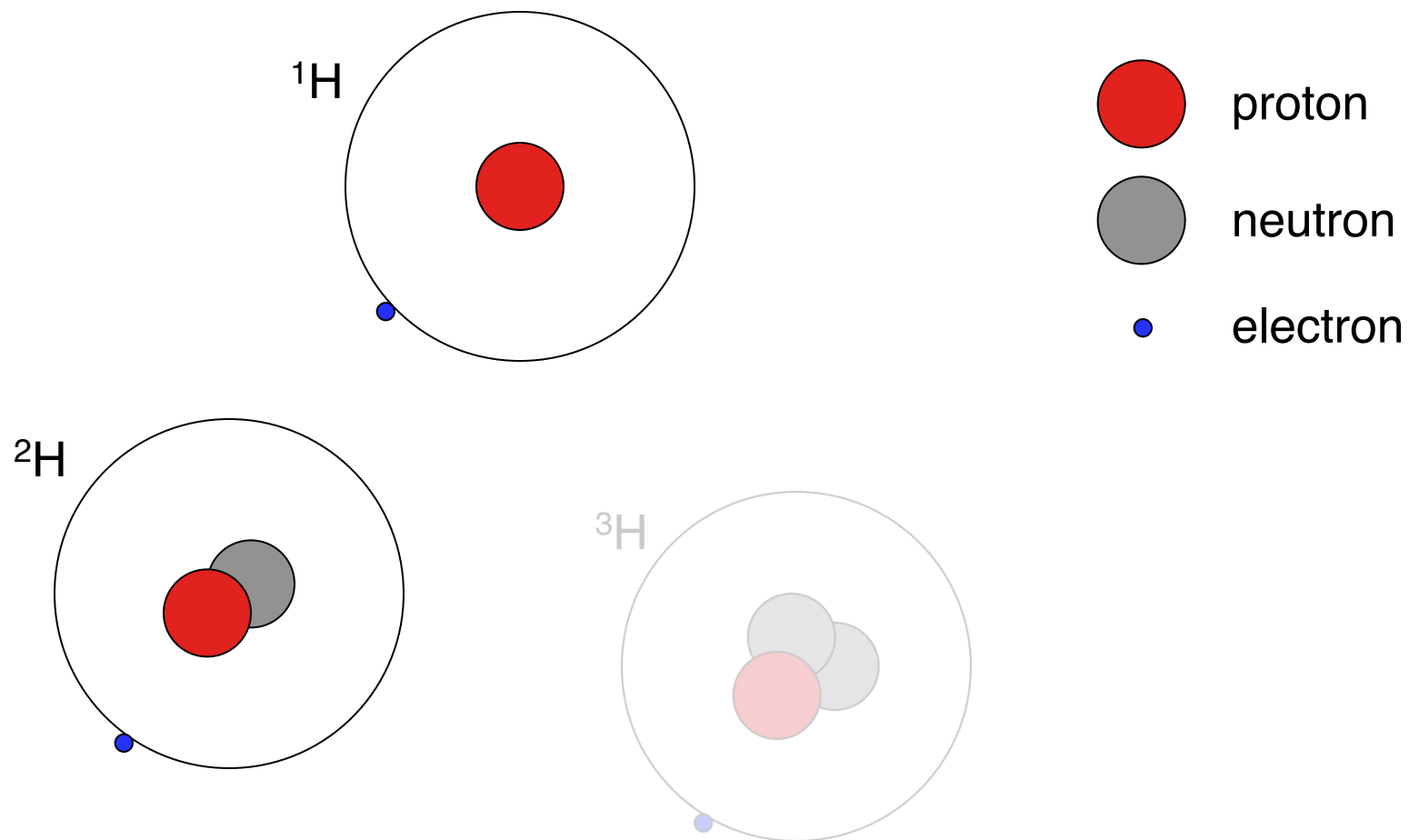
For example,

Hydrogen stable isotopes =  $^1\text{H}$  and  $^2\text{H}$

Carbon stable isotopes =  $^{12}\text{C}$  and  $^{13}\text{C}$

Nitrogen stable isotopes =  $^{14}\text{N}$  and  $^{15}\text{N}$

Heavier stable isotopes contain an additional 1-2 neutrons





## How do we express stable isotope abundances?

$$R = \frac{\text{heavy isotope}}{\text{light isotope}}$$

what do we mean by ratio

$$R = \frac{^{13}\text{C}}{^{12}\text{C}}$$

carbon as an example

the isotope ratio is typically presented in delta notation

$$\delta = \left( \frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) \bullet 1000 \text{ ‰}$$

# An isotope ratio mass spectrometer measures isotope ratios

light gas  
elements

H<sub>2</sub>  
CO  
CO<sub>2</sub>  
N<sub>2</sub>  
SO<sub>2</sub>

sample accelerates  
along flight tube

magnet

detectors for  
different  
isotopes

sample enters here  
and is ionized

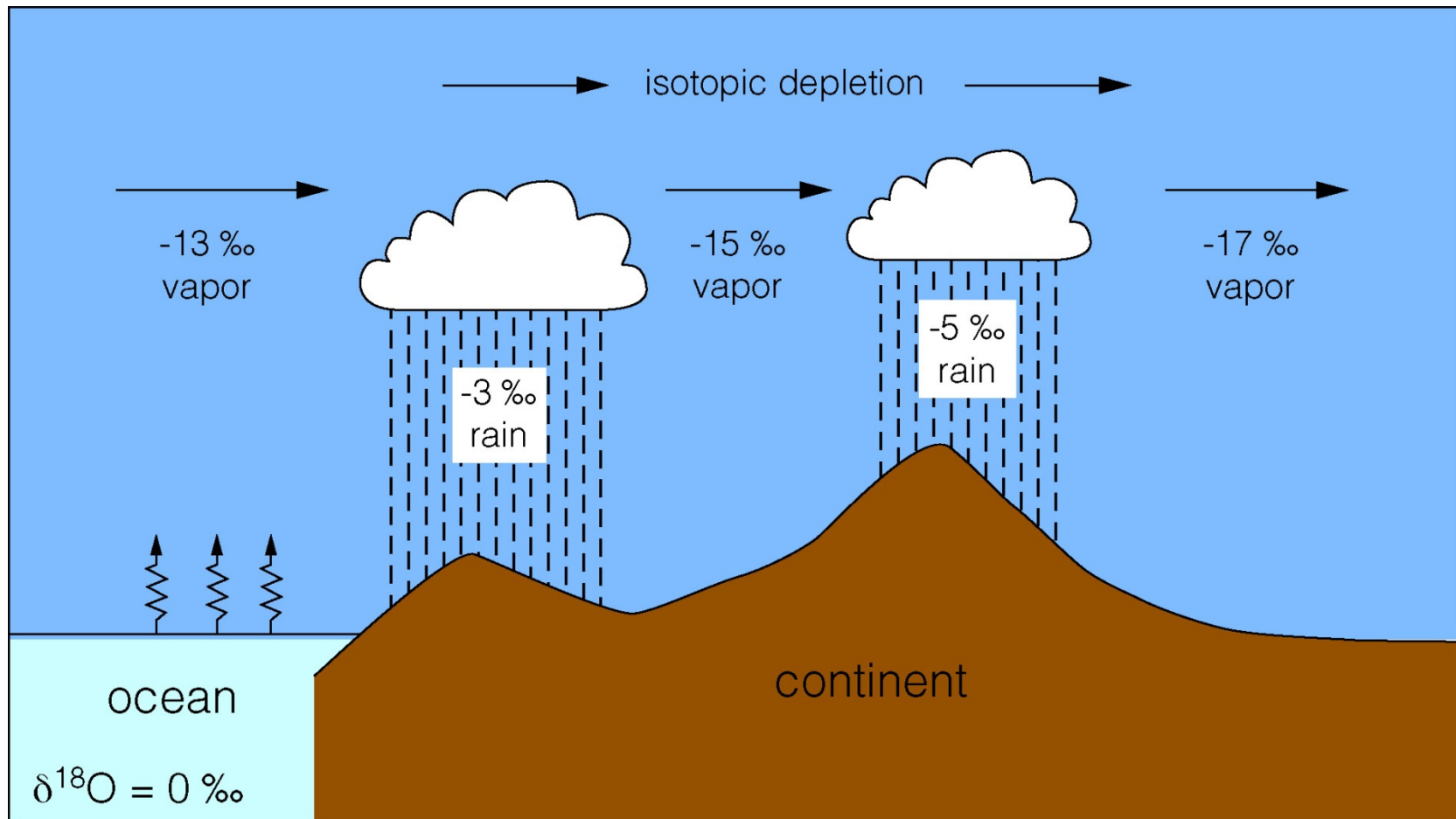


continuous flow linkage

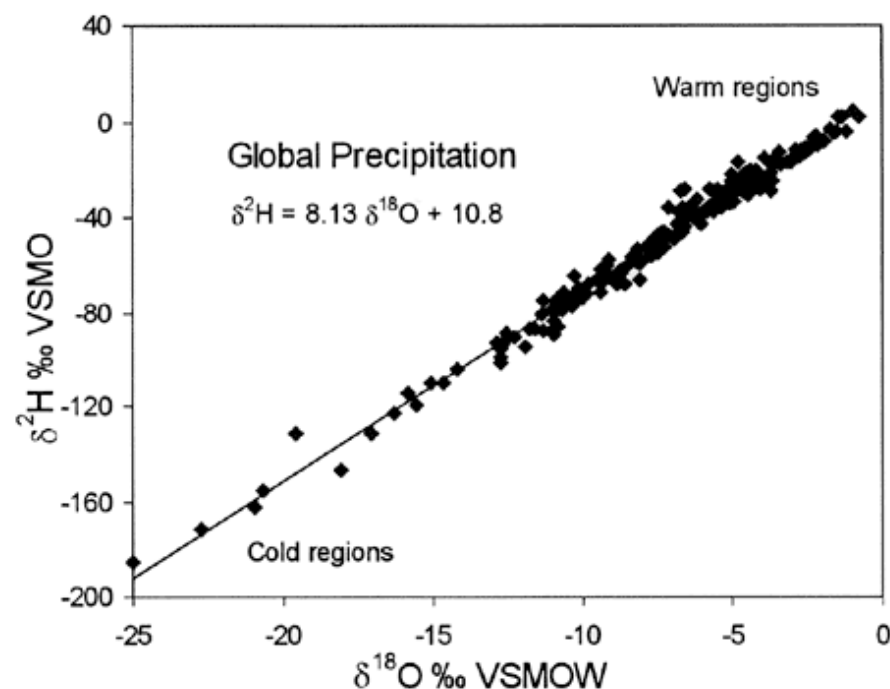
- elemental analyzer
- gas chromatography
- liquid chromatography
- laser ablation



$^2\text{H}$  and  $^{18}\text{O}$  isotopes preferentially fall out of precipitation first, leaving a residual cloud mass that is isotopically depleted



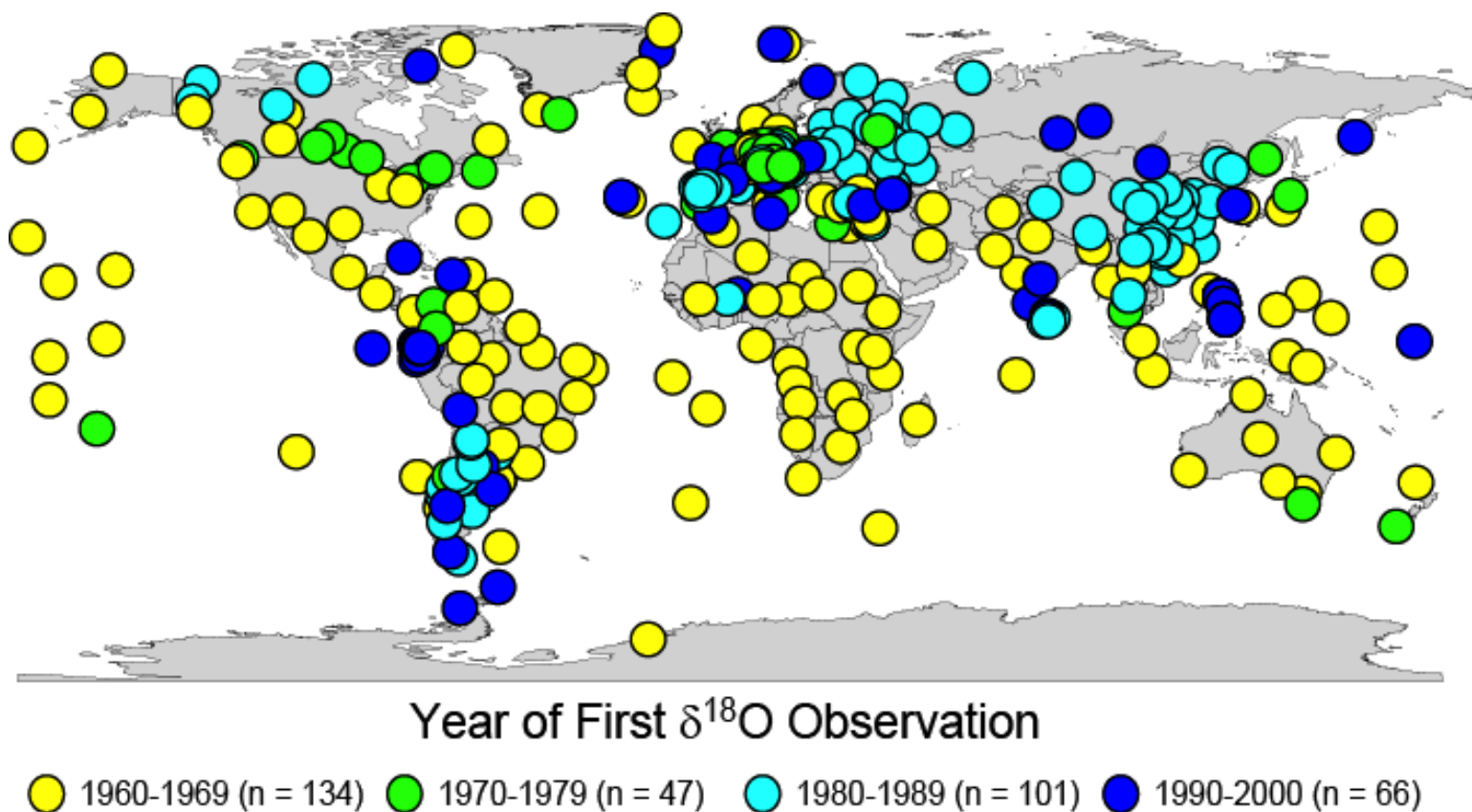
## H and O isotopes of rain follow a linear relationship



The slope of this line is 8 and is known as the meteoric water line; evaporated water has a slope of less than 8.

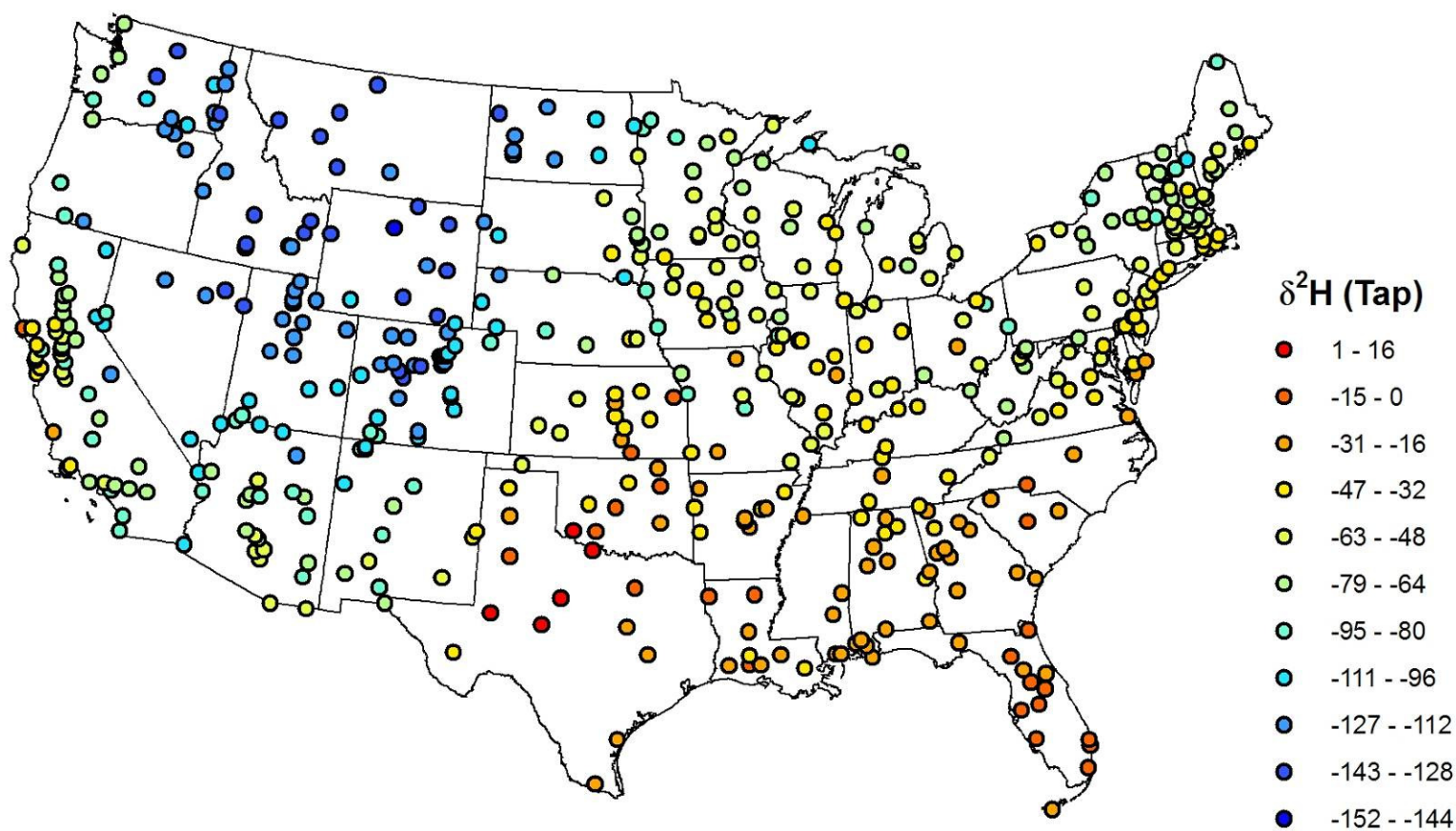
We determine patterns using observational data --- e.g., rainfall

- <http://isohis.iaea.org/>





We spatially integrate using observational data --- e.g., tapwater



Bowen et al., 2007

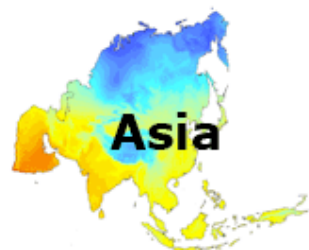
Maps are produced in GIS using isotope, latitude, and elevation



Waterisotopes.org

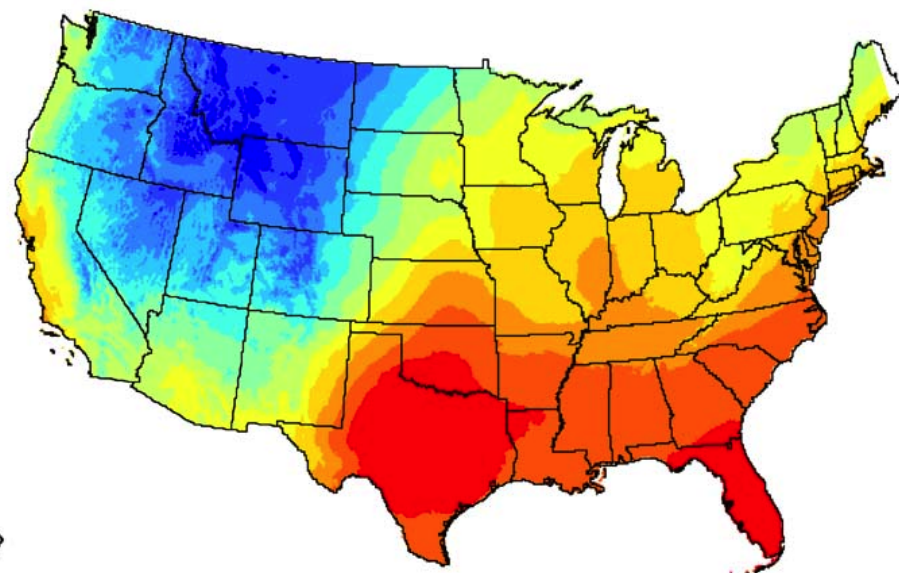


Gabe Bowen

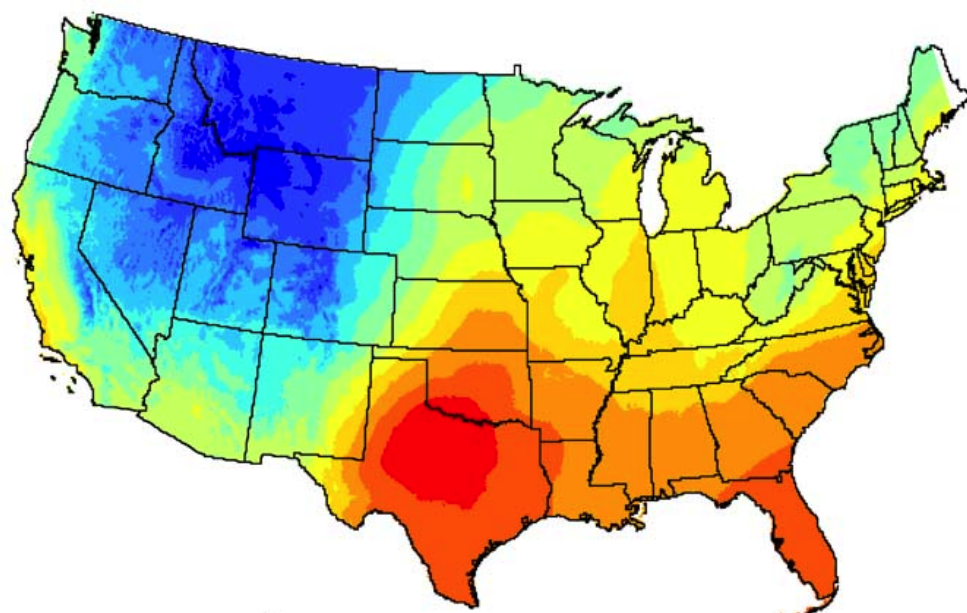
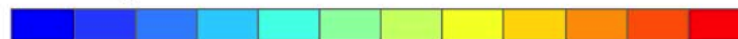


PURDUE  
UNIVERSITY

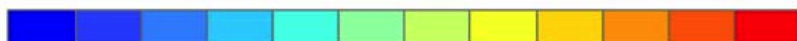
# Water isotopes reveal consistent predictable patterns



Predicted Tap,  $\delta^2\text{H}$

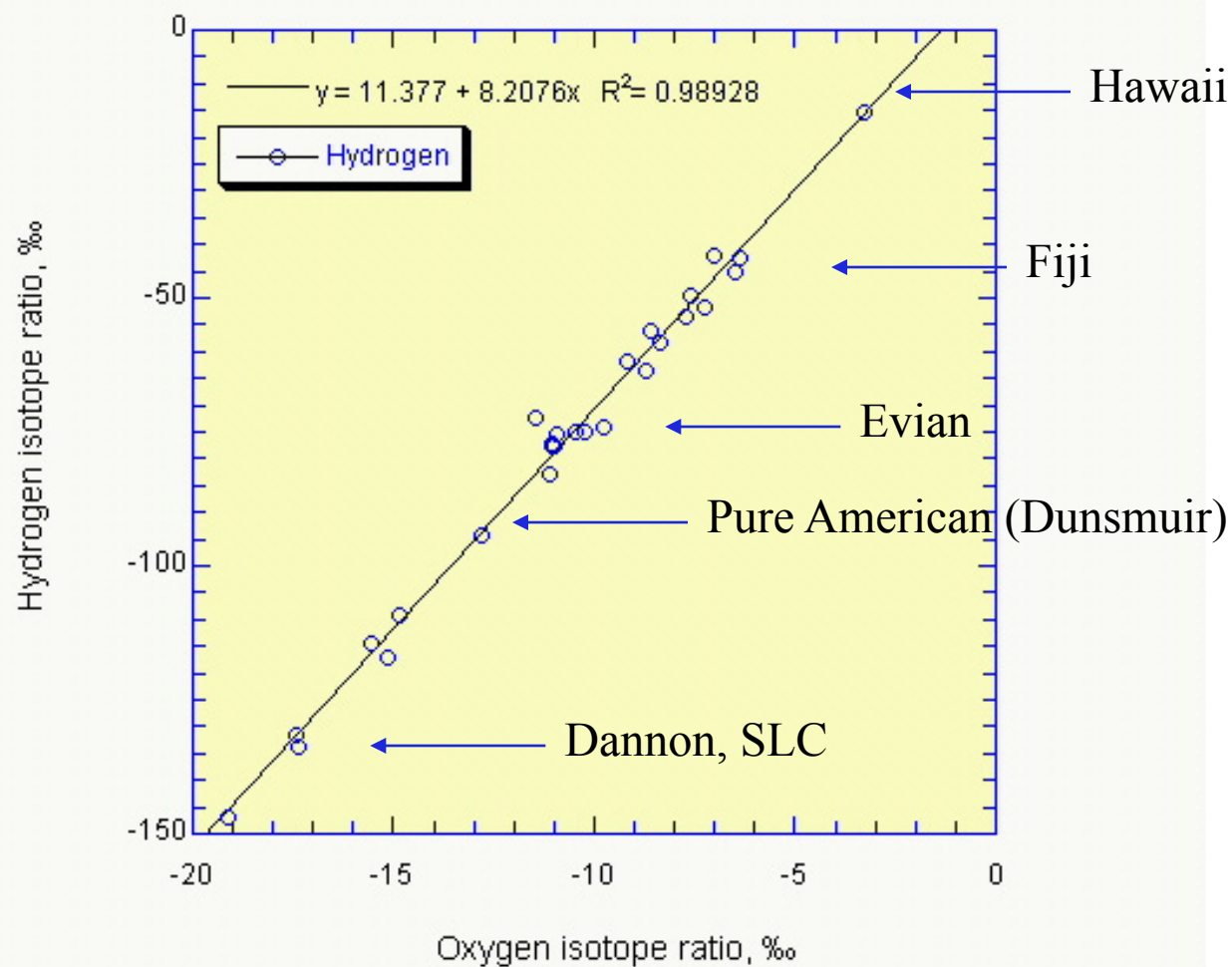


Predicted Tap,  $\delta^{18}\text{O}$



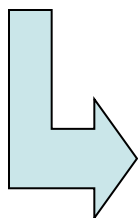
Bowen et al., 2007

## Application: verifying the origins of bottled waters



# An organism's water isotopes reflect the water source geo-environment

environment water



organism water  
(blood, leaf water, etc.)

A series of fractionation events occur along the sequence from the water in a geographical region to the water in the organism. Included are evaporation, metabolism, and respiration factors (can be modeled).

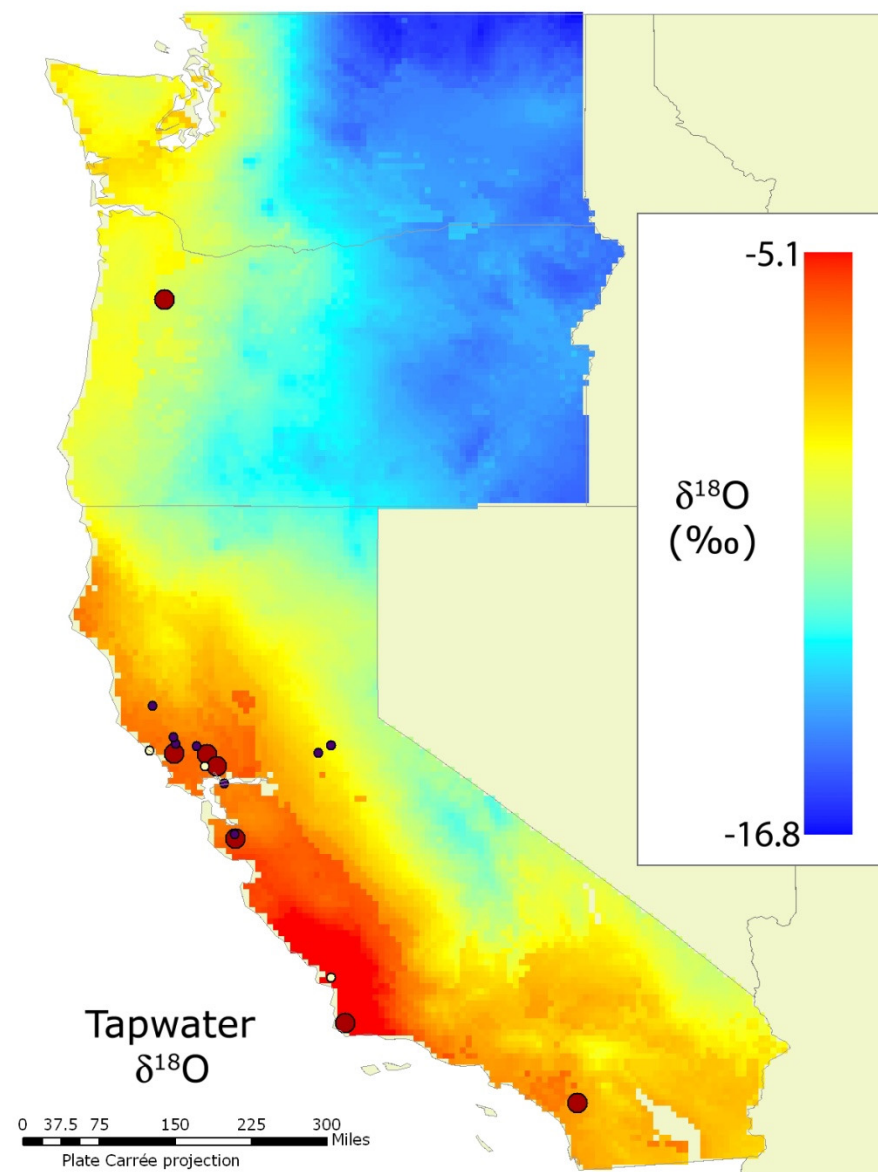
blood  
leaf water  
juices  
beverages  
wine  
fruit





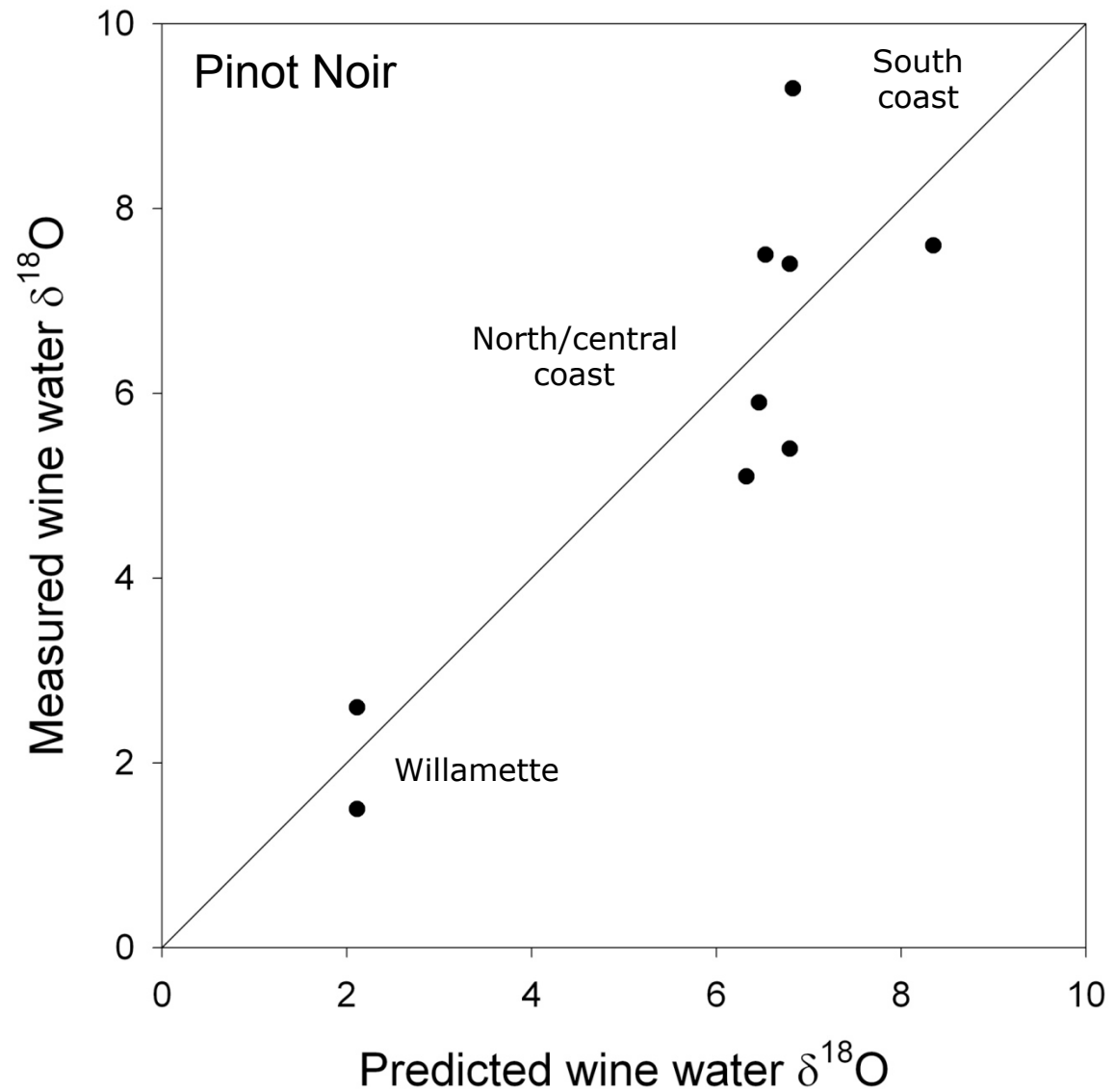
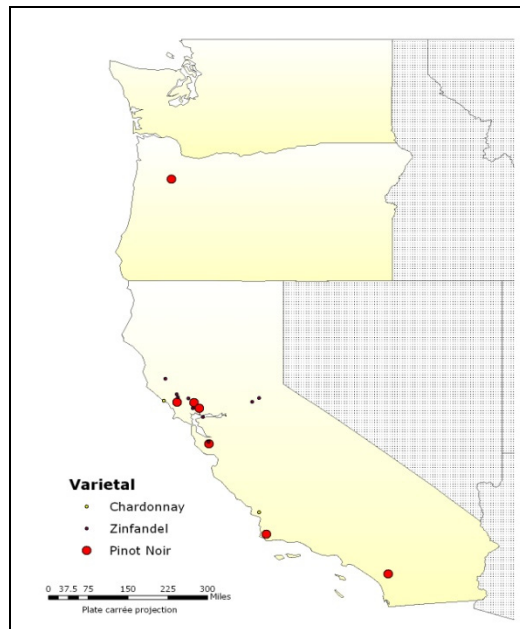


## Application: predicting the origins of wine



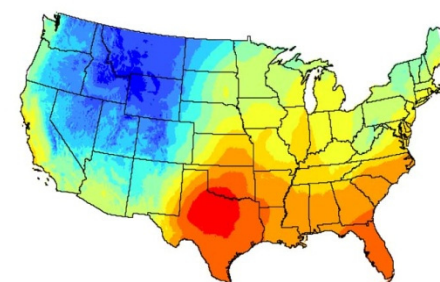
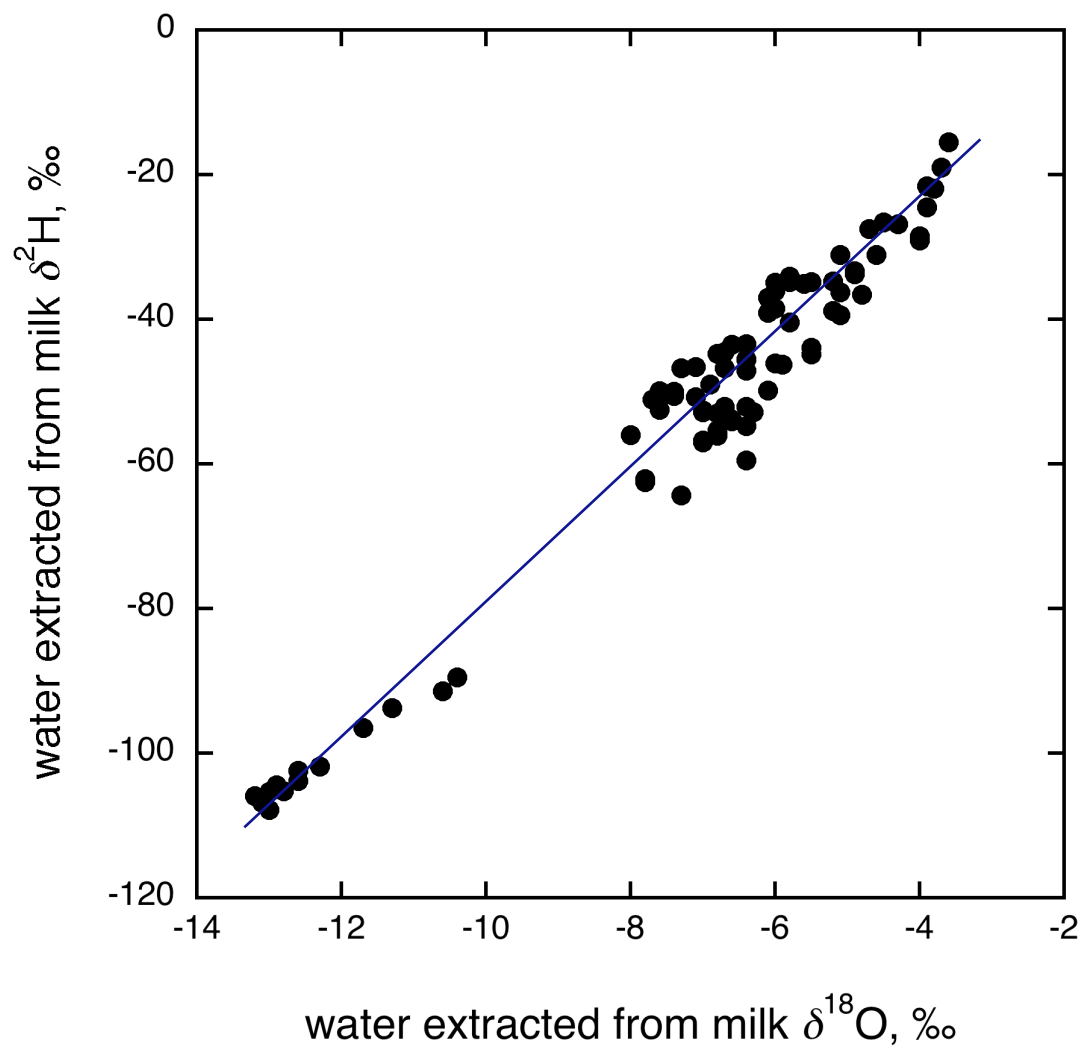
West et al. (2007)

# Stable isotopes predict origins of wine varieties



West et al. (2007)

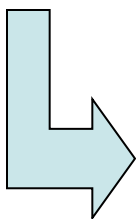
# Water in milk across the USA exhibits geographic variation



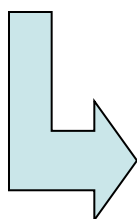
Chesson et al., unpublished (see poster)

# Organic H and O isotopes record water environment

environment water



organism water  
(blood, leaf water, etc.)

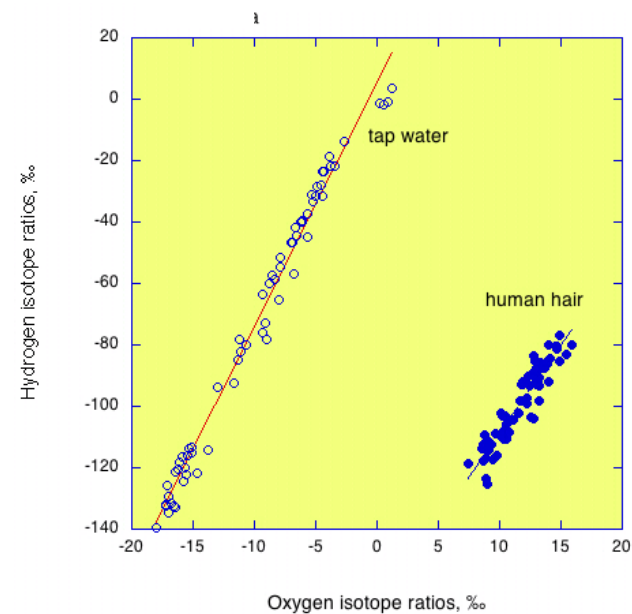
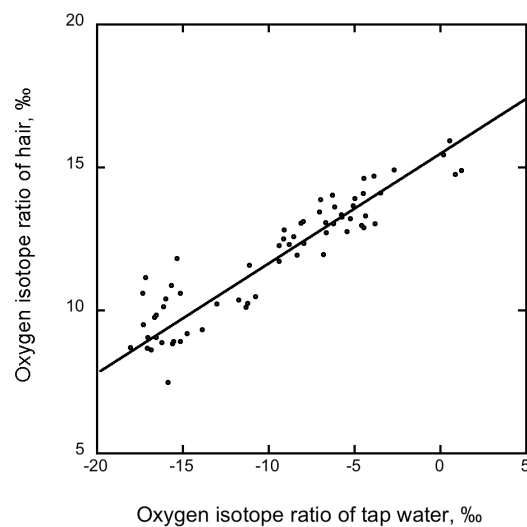
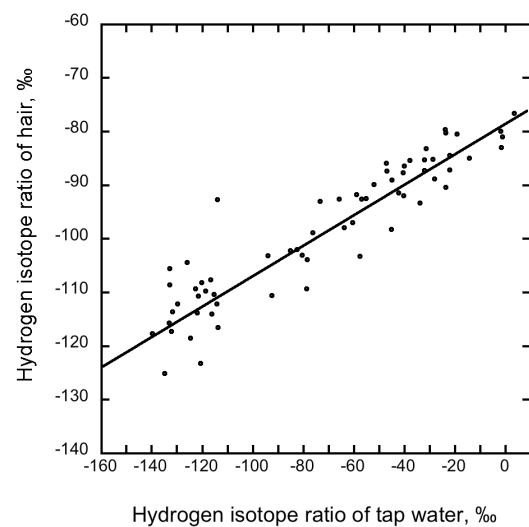
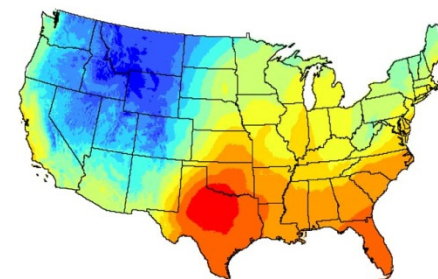


plant or animal  
organic matter

A series of fractionation events occur along the sequence from the water in a geographical region to the organic matter of the organism.

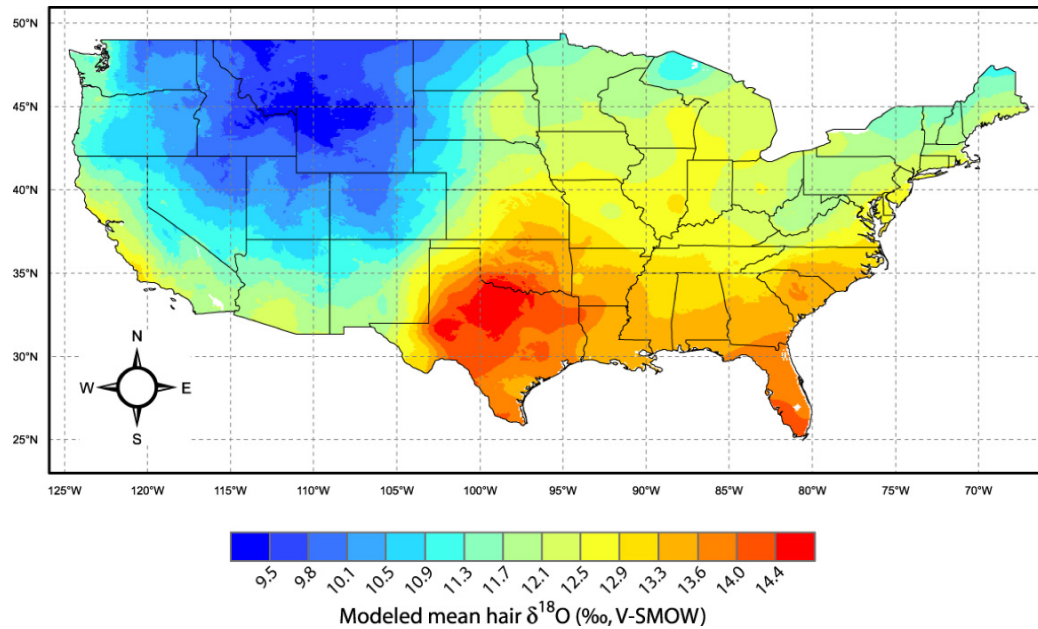


# Hydrogen and oxygen isotope ratios of keratin protein are correlated with tap water

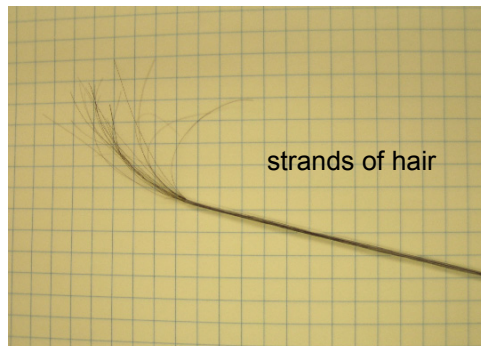


Ehleringer et al. (2008)

# Application: O isotopes in hair reveal geographic movement



“Saltair Sally”



strands of hair



travel histories



Ehleringer et al. (2009)

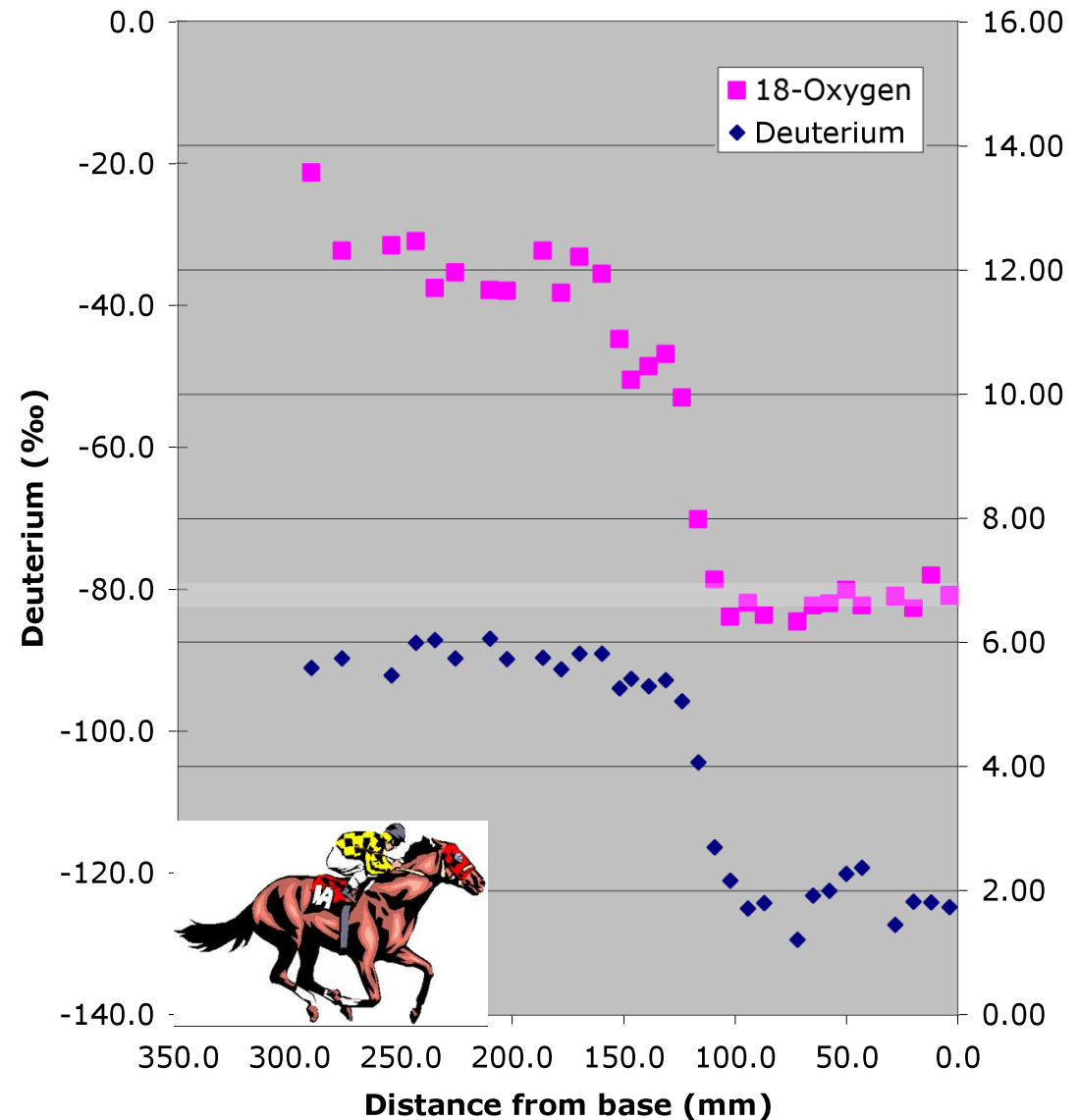
# Application: O isotopes in hair reveal geographic movement

Consider the movement of an animal based on the stable isotope analysis of a single hair

History:

Jan - Nov 2002, Virginia

Nov - May 2003, Utah



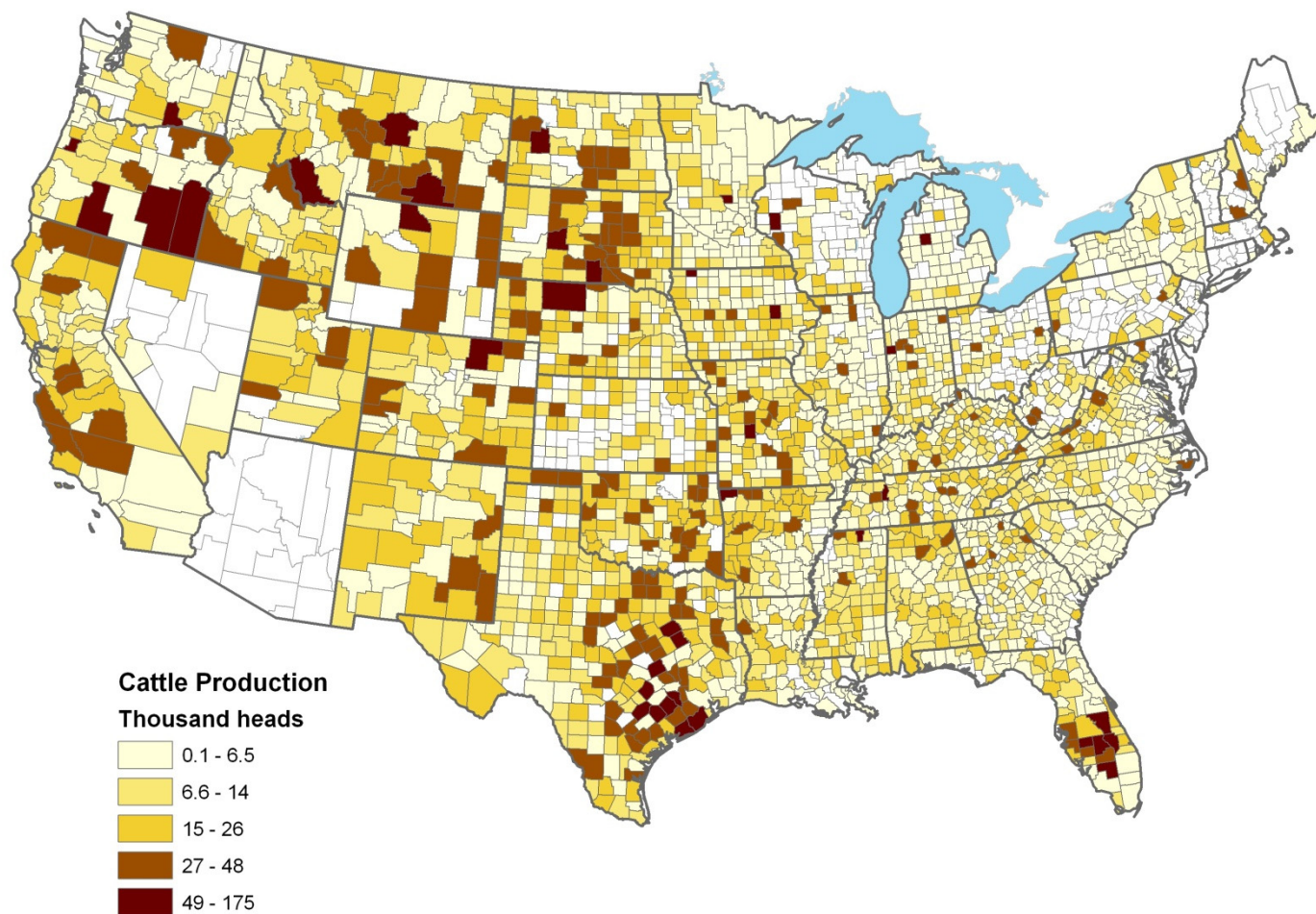
(unpublished data)

The same approach can be applied to food proteins



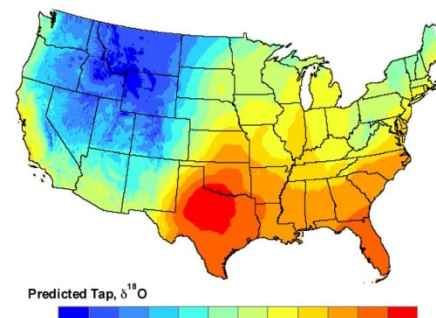
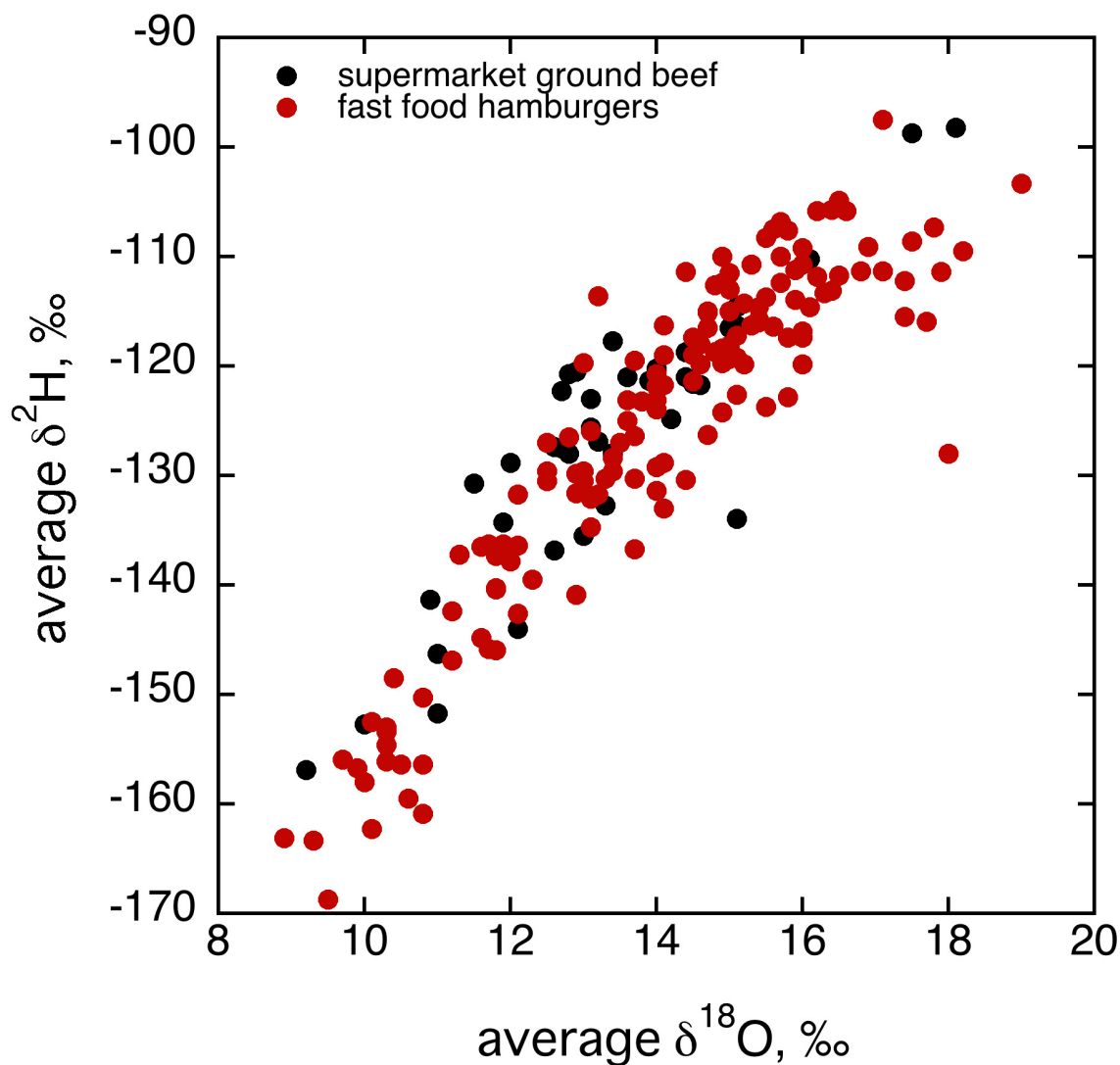


# Cattle are concentrated in isotopically distinct regions



Data from 2002 (USDA National Agricultural Statistics Service)

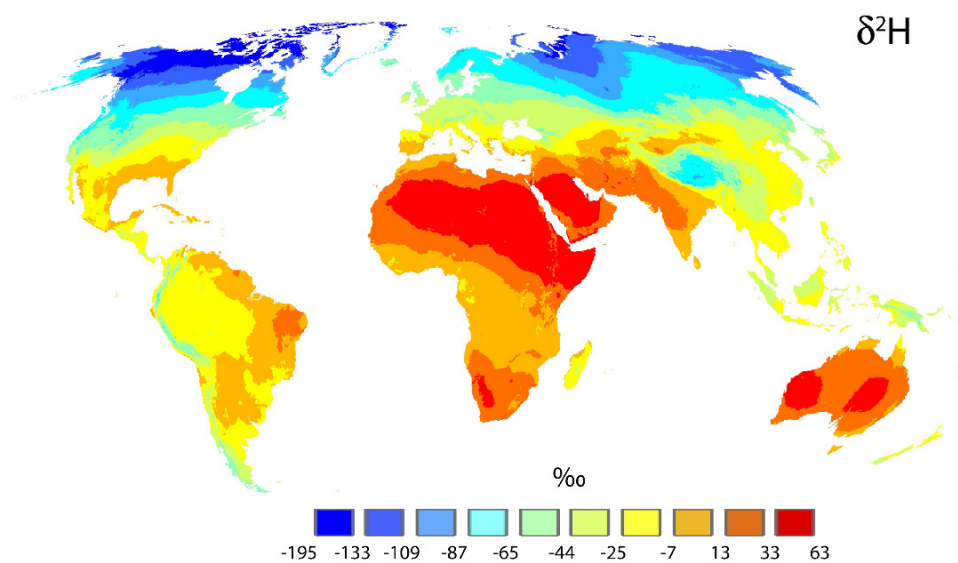
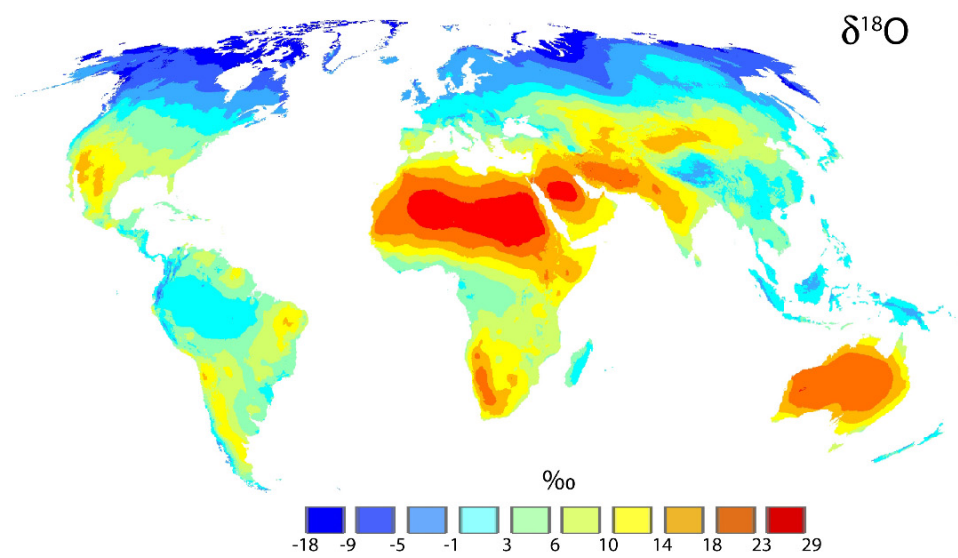
# Beef H and O isotope values exhibit significant correlation



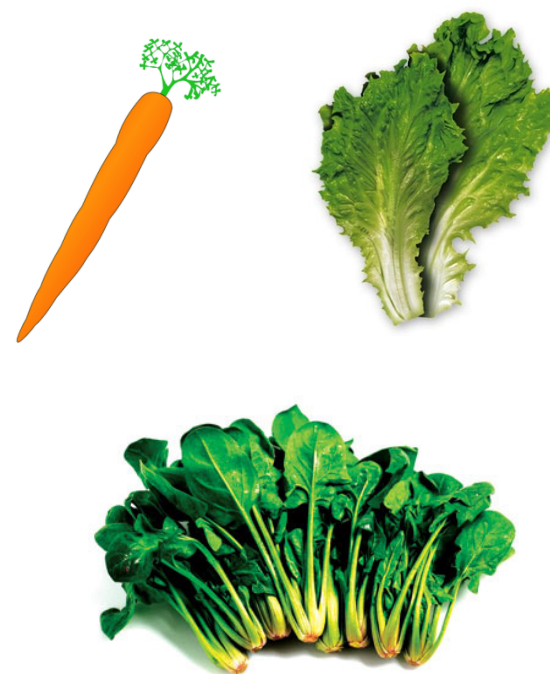
Chesson et al. 2008; in review



# Evaporation enriches leaf water above meteoric water



Applications:



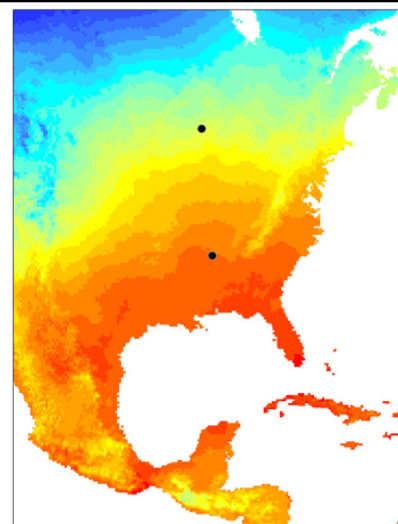
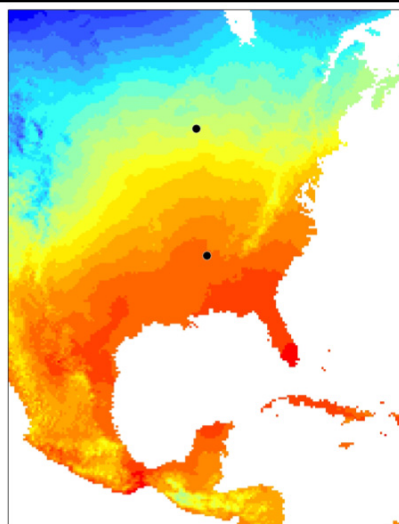
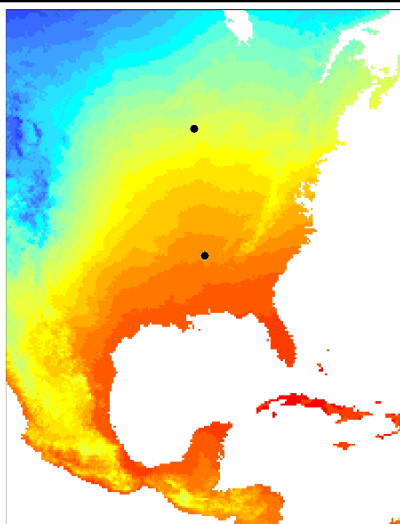
West et al. (2008)

# Application: sourcing of vegetable foods

environment water

organism water

organic matter



Precipitation  $\delta^2\text{H}$

Leaf water  $\delta^2\text{H}$

Leaf CHD  $\delta^2\text{H}$



environment water

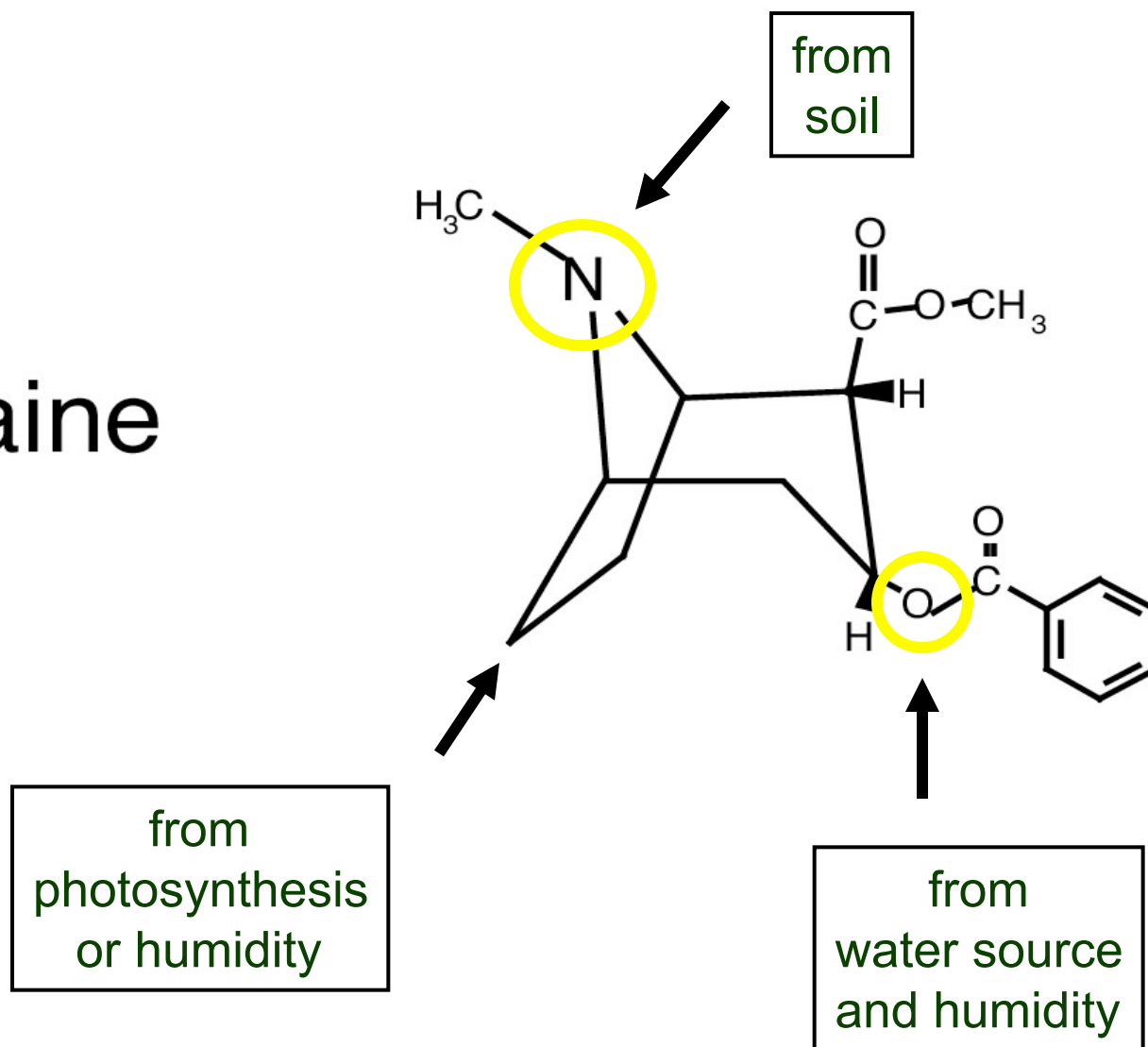
leaf water

carbohydrate

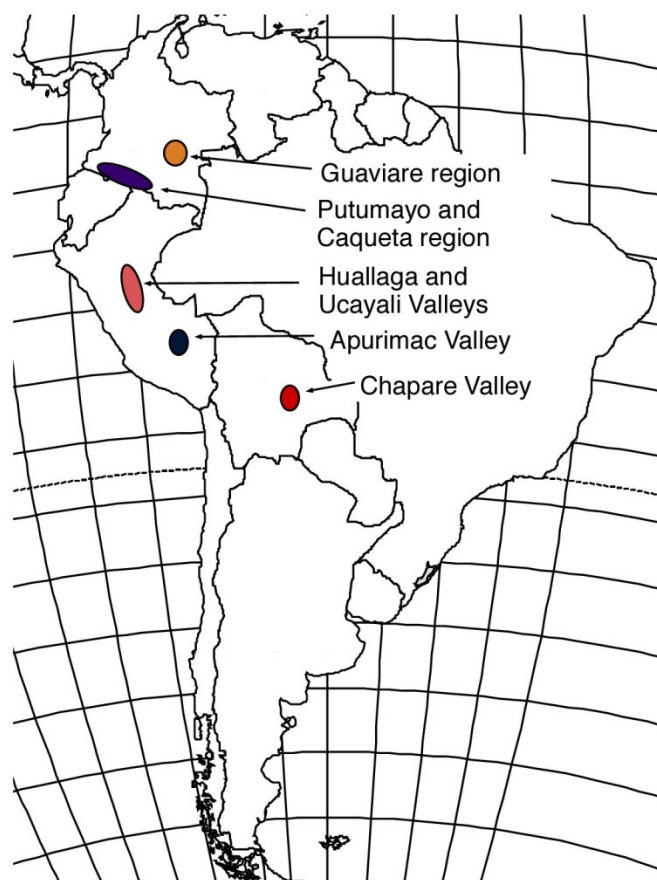
West et al. (2008)

Application: C & N isotopes can identify some geographic regions

# Cocaine



## Application: C and N isotopes can identify geographic region



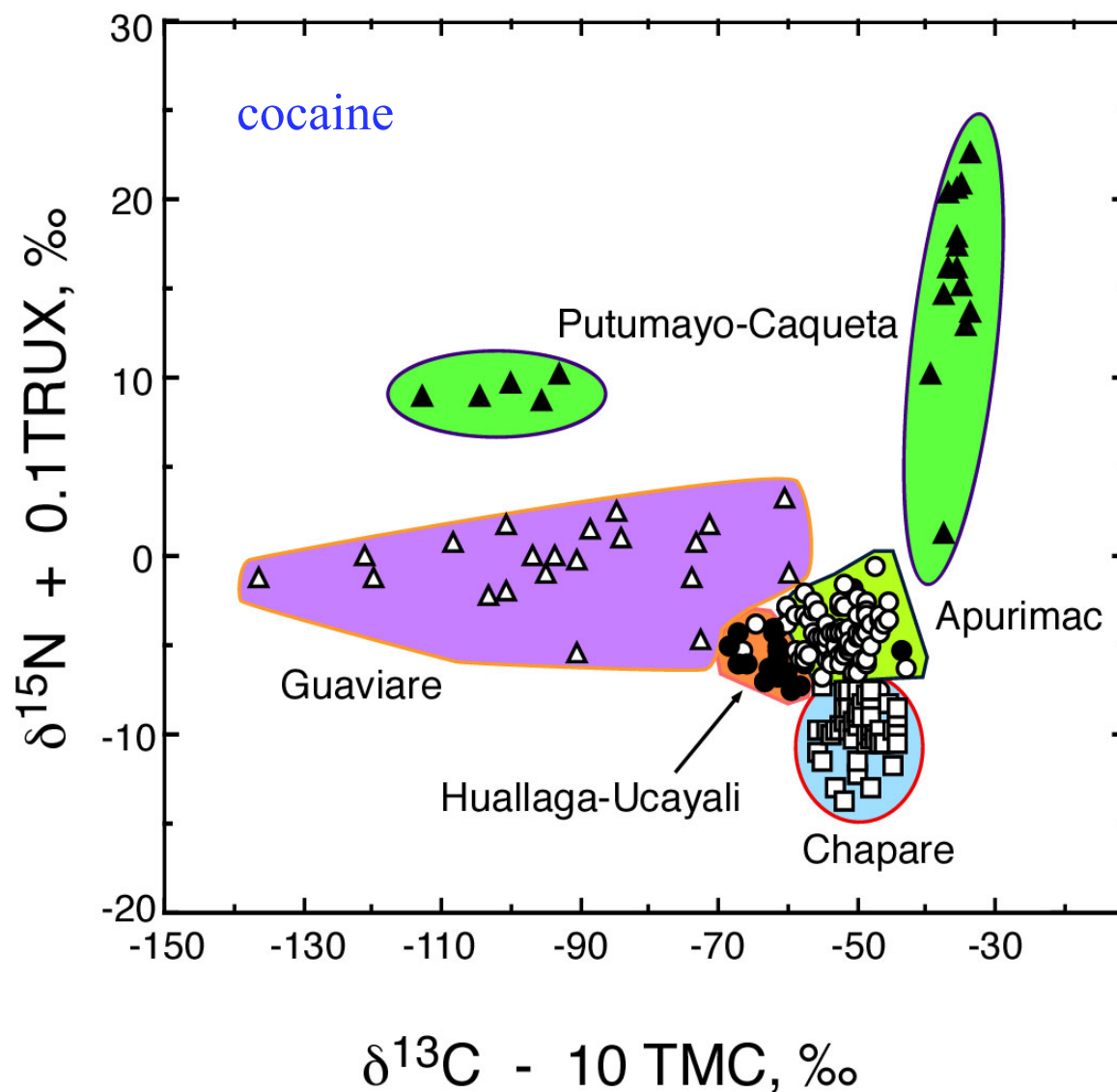
Almost all of the cocaine in South America originated from one of only 5 regions.

These regions fall along a climatic gradient with the wettest regions in the north and driest in the south.

Ehleringer et al. (2001)

Collaboration with John Casale, DEA

# Isotopes, TMC, and TRUX explain ~96 % of cocaine variation

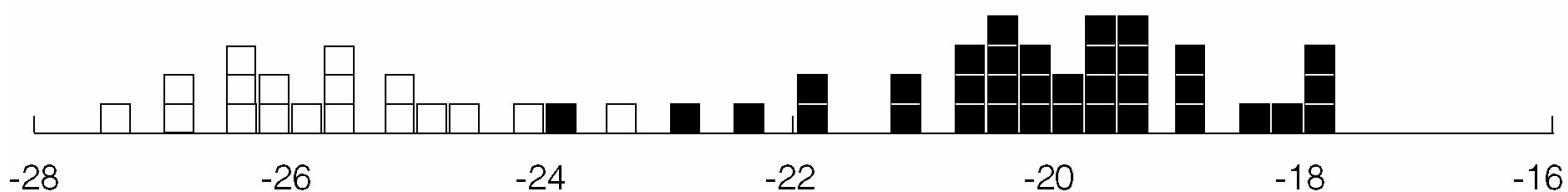


Ehleringer et al. (2001)

# Adulteration: C4 sugars in sparkling wines

□ Argentina, Chile, E.U.

■ Australia, Brazil, U.S.



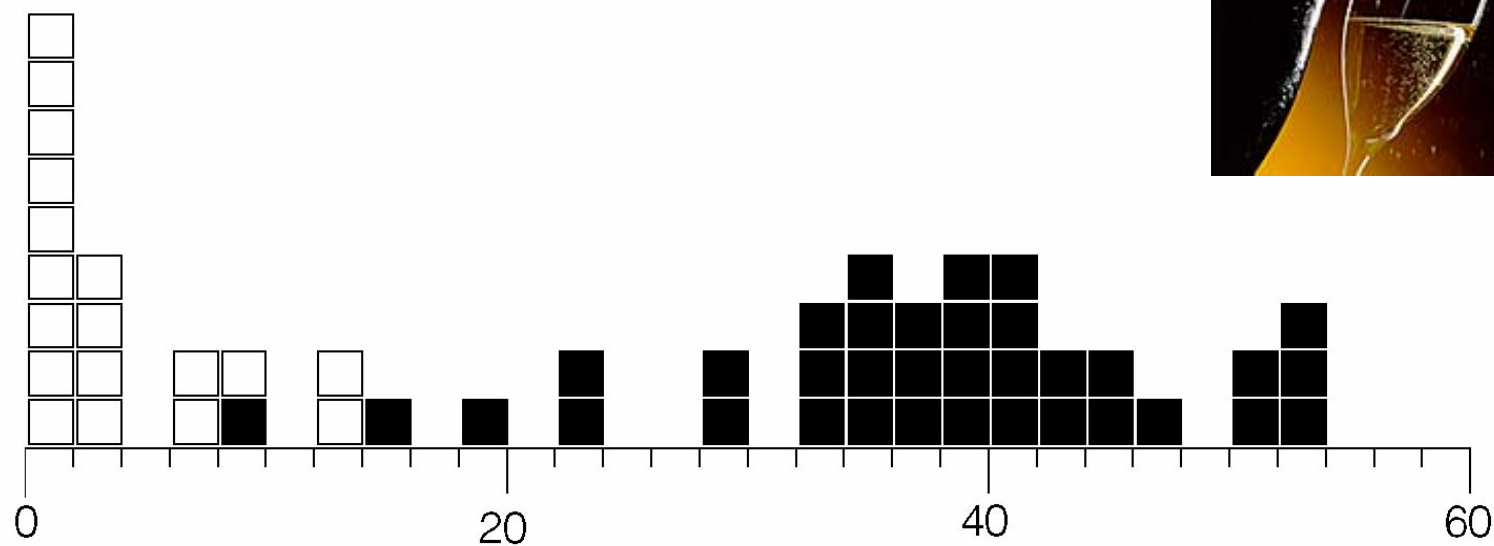
Carbon isotope ratios of brut, ‰

Martinelli et al. (2003)



# Adulteration: C<sub>4</sub> sugars in sparkling wines

□ Argentina, Chile, E.U.  
■ Australia, Brazil, U.S.



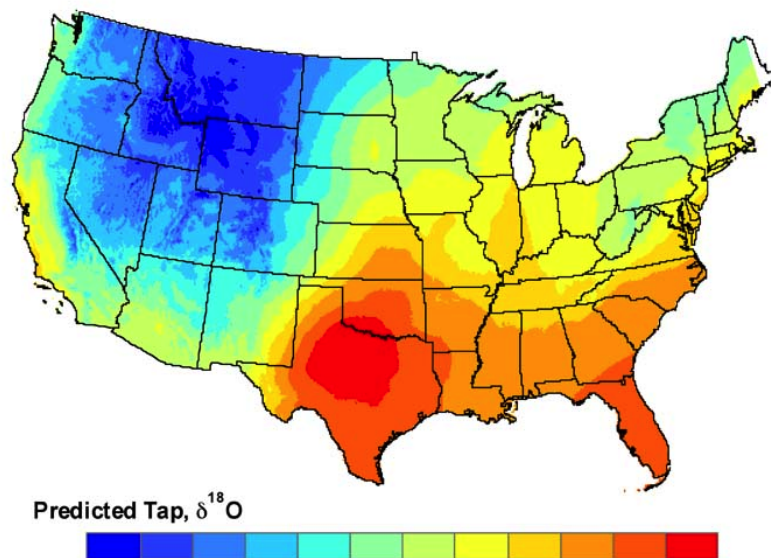
Calculated C<sub>4</sub> sugar contribution to brut, ‰



Martinelli et al. (2003)

# Nationwide and international stable isotope applications

- eliminating regions as a source of contaminant
- delineating regions consistent with source of a threat
- contribute to traceability of food items
- region-of-origin authentication



- beverages and juices
- protein sources
- vegetables and fruits
- fiber
- oils
- extracted compounds
- adulteration in foods
- adulteration in beverages

