Determining Geographic Origins of Foods Using Stable Isotope Ratios

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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

Inputs
- Isotope analyses of evidence
- Environment where evidence was collected
- Local water sample
- Local climate conditions
- Climate and geography database layer
- Water database layer

Data and modelling efforts
- Database of food isotope ratios
- Process model
- GIS model
- Inverse spatial maps

Outputs
- Consistent or not with a set of known observations?
- What are predictions for evidence in that location?
- What are the predicted regions from which that evidence could have originated?
Stable isotopes are multiple forms of the same element

For example,

Hydrogen stable isotopes = $^1$H and $^2$H

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

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

carbon as an example

the isotope ratio is typically presented in delta notation

\[ \delta = \left( \frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) \times 10000 \ \% \]
An isotope ratio mass spectrometer measures isotope ratios

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

North America  South America  Africa

Europe  Asia  Australia

Gabe Bowen
Water isotopes reveal consistent predictable patterns

Bowen et al., 2007
Application: verifying the origins of bottled waters

![Graph showing hydrogen isotope ratio vs. oxygen isotope ratio for various bottled waters: Hawaii, Fiji, Evian, Pure American (Dunsmuir), and Dannon, SLC. The graph includes a linear regression line with the equation $y = 11.377 + 8.2076x$ and $R^2 = 0.98928$.](image-url)
An organism’s water isotopes reflect the water source geo-environment

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).
Application: predicting the origins of wine

West et al. (2007)
Stable isotopes predict origins of wine varietals

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

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.
Application: O isotopes in hair reveal geographic movement

Ehleringer et al. (2009)

“Saltair Sally”

Modeled mean hair $\delta^{18}$O (%o, V-SMOW)

strands of hair

travel histories

Ehleringer et al. (2009)
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

West et al. (2008)
Application: C & N isotopes can identify some geographic regions

Cocaine

- from soil
- from photosynthesis or humidity
- from water source and humidity
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

\[ \delta^{15}N + 0.1\text{TRUX}, \% \]

\[ \delta^{13}C - 10 \text{ TMC}, \% \]

Ehleringer et al. (2001)
Adulteration: C4 sugars in sparkling wines

Martinelli et al. (2003)

Carbon isotope ratios of brut, ‰

- Argentina, Chile, E.U.
- Australia, Brazil, U.S.
Adulteration: C4 sugars in sparkling wines

- Argentina, Chile, E.U.
- Australia, Brazil, U.S.

Calculated C4 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