Tracing the geographical origin of cocaine

Cocaine carries a chemical fingerprint from the region where the coca was grown.

Here we show that cocaine originating from different geographic regions in South America can be identified by its isotopic-ratio signature. The distinct carbon (δ13C) and nitrogen (δ15N) isotope-ratio combinations allow the country of origin to be determined for the principal coca-growing regions along the Andean Ridge. By combining this information with detectable differences in the patterns of the trace alkaloids truxilline and trimethoxycocaine, we correctly identified the source of 96% of 200 cocaine samples.

Cocaine is the most widely used narcotic drug, making the determination of the geographic origin of illicit cocaine the focus of intense investigation by the forensic community1,2. Previous studies have concentrated on detecting the trace residues present in cocaine or the trace alkaloids that are extracted with it; these have met with limited success, although they have been valuable in identifying the processing methods peculiar to different regions. However, refined cocaine base is often transported from one country to another for its final conversion to cocaine hydrochloride, making the source harder to identify.

Stable-isotope ratios (δ) have been used as indicators of the geographical source of a wide variety of biological and non-biological materials, from migrating butterflies3 to isotopic-ratio analysis, determinations as indicators of the geographical source of a drug, making the determination of the geographical origin of illicit cocaine, right. Identification of cocaine-growing regions based on a combined model derived from carbon- and nitrogen-isotope ratios as well as abundance of minor alkaloid components. Squares, Bolivia; triangles, Colombia; and circles, Peru. Regions within a country are distinguishable by black and white symbols. Trux, truxilline; TMC, trimethoxycocaine. Isotope ratios are expressed as \( \frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \) × 1,000‰, where \( R \) is the molar ratio of heavy-to-light stable isotope; standards for carbon and nitrogen are PDB and air, respectively.

Table 1 Differences in trace alkaloid amounts among coca populations

<table>
<thead>
<tr>
<th>Region</th>
<th>Truxillines</th>
<th>Trimethoxycocaines</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chapare Valley, Bolivia</td>
<td>2.79 ± 0.41</td>
<td>0.16 ± 0.03</td>
<td>17.2</td>
</tr>
<tr>
<td>Huallaga/Ucayali Valleys, Peru</td>
<td>3.80 ± 0.76</td>
<td>0.19 ± 0.04</td>
<td>20.0</td>
</tr>
<tr>
<td>Apurimac Valley, Peru</td>
<td>4.11 ± 0.59</td>
<td>0.27 ± 0.06</td>
<td>15.2</td>
</tr>
<tr>
<td>Guaviare region, Colombia</td>
<td>4.94 ± 0.20</td>
<td>0.61 ± 0.06</td>
<td>8.1</td>
</tr>
<tr>
<td>Putumayo–Caqueta region, Colombia</td>
<td>14.66 ± 4.29</td>
<td>0.20 ± 0.29</td>
<td>73.30</td>
</tr>
</tbody>
</table>

Means and standard deviations of truxilline and trimethoxycocaine content of coca leaves, together with the ratio of truxilline to trimethoxycocaine growing in different regions of South America are shown. Data are given on a w/w% basis using structurally related internal standards.

Figure 1 Identification of geographic regions in South America where coca is commonly grown. Left, regions producing illicit cocaine; right, identification of cocaine-growing regions based on a combined model derived from carbon- and nitrogen-isotope ratios as well as abundance of minor alkaloid components. Squares, Bolivia; triangles, Colombia; and circles, Peru. Regions within a country are distinguishable by black and white symbols. Trux, truxilline; TMC, trimethoxycocaine. Isotope ratios are expressed as \( \frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \) × 1,000‰, where \( R \) is the molar ratio of heavy-to-light stable isotope; standards for carbon and nitrogen are PDB and air, respectively.

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The environmental basis for this regional variation in coca-leaf and cocaine stable isotopes may be due to differences in soils affecting δ13C, and in the humidity and length of the wet season affecting δ15N. It has been proposed7 that nutrient cycles in tropical forest ecosystems (typical of coca-growing regions) are more open than in drier, more temperate regions, and the range of coca-leaf δ13C values are consistent with this idea. δ13C differences are narrower, but are consistent with patterns predicted by δ15N theory for plants8,9.
Tracing the country of origin of cocaine is now feasible through automated, routine analysis of both stable isotopes and trace alkaloids, opening up strategic options for identifying source regions and trafficking routes. We have shown how ecological and isotope-fractionation principles used to predict isotopic-ratio patterns associated with plants from different ecosystems can also be applied to determine the distribution of an illegal drug, as well as to identify new coca-producing regions as they develop.

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Archaeology

Detecting milk proteins in ancient pots

Deciding whether to farm cattle for milk or beef was just as complex in the past as it is today. Compared with meat production, dairying is a high-input, high-risk operation indicative of an intensive, sophisticated economy, but this practice is notoriously difficult to demonstrate in the archaeological record. Here we provide evidence for the presence of milk proteins preserved in prehistoric vessels, which to our knowledge have not been detected before. This finding resolves the controversy that has surrounded dairying on the Scottish Atlantic coast during the Iron Age2–5 and indicates that farming by the early inhabitants of this harsh, marginal environment was surprisingly well developed.

The analysis of sorbed lipid residues in prehistoric ceramics has provided a powerful new indicator of how vessels were used4–6. Although proteins are more diagnostic of specific foodstuffs than lipids, they are difficult to extract from archaeological ceramics7. We have developed an immunological detection method, the digestion-and-capture immunoassay (DACIA)8–10, which overcomes this difficulty by dissolving the ceramic then capturing the liberated proteins for immunodetection.

We obtained sherds from nine coarseware cooking vessels, dated to the middle of the first millennium BC, from the fill of an Early Iron Age house at Cladh Hallan, South Uist, in the Outer Hebrides, and analysed them by DACIA. Extracts were tested using a monoclonal antibody raised against heat-degraded and dephosphorylated bovine α-casein (about 1.4% w/v milk), which was specific for bovine milk.

Immunological analysis of archaeological materials has been criticized for the lack of negative controls11, so we included an extensive array of reference samples (Fig. 1). Seven of nine of the interior sections of sherds recovered from Cladh Hallan tested positive for casein and the amounts were comparable to those found on experimentally buried milk sherds (Fig. 1). DACIA analysis failed to detect the presence of bovine α-casein in the associated sediment or exterior surfaces of the samples.

The large number of neonatal cattle remains found at this site (42% of individuals) has been attributed to the deliberate culling of young calves in order to preserve fodder in an adverse environment12–15 or to sustain a high-input dairying economy.16 The presence of bovine α-casein on a substantial number of sherds (Fig. 1) lends support to the latter interpretation. Our successful characterization of protein residues after 2,500 years demonstrates the potential of DACIA as a high-resolution technique for determining how archaeological ceramics were used.

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