Predicting local bioavailable $^{87}Sr/^{86}Sr$ isotopes and similarity search in multi-dimensional isotope data sets

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Abstract

The definition of local bioavailable $^{87}Sr/^{86}Sr$ isotopic signatures is crucial for the detection of non-local skeletal finds and provenance analysis. A multi-isotope approach by use of additional isotopic ratios permits the establishment of an isotopic fingerprint that can be used for similarity search. A mixing model was developed for the prediction of local $^{87}Sr/^{86}Sr$ signatures for selected sites along an archaeological important passage across the European Alps (Inn-Eisack-Adige passage across the Brenner Pass). This model was based on strontium concentrations and isotopic ratios of environmental samples (wood, water, soil) and correctly predicted the isotopic signatures of local vertebrates. A multi-isotope fingerprint consisting of stable strontium, lead, and oxygen isotopes in the bioapatite of archaeological animals and human cremations (omitting the thermally unstable $\delta^{18}O_{phosphate}$ in the latter) along the Alpine passage was forwarded to a Gaussian Mixture Model (GMM) clustering for the scope of similarity search. GMM clustering was capable of identifying groups of animals and humans that were spatially separated with a high probability (average $p > 0.9$). This way, local, non-local and also mixed isotopic signatures in the multi-isotope fingerprint were firmly detected. GMM clustering was also successfully applied to a palaeoecological study in an ecological complex region at the Baltic coast (Viking Age Haithabu and medieval successor Schleswig). A multi-isotope fingerprint in vertebrate skeletal finds including humans that consisted of $\delta^{13}C_{collagen}$, $\delta^{15}N_{collagen}$, $\delta^{13}C_{carbonate}$, $\delta^{18}O_{carbonate}$, $\delta^{18}O_{phosphate}$, $\delta^{34}S_{collagen}$, and $^{87}Sr/^{86}Sr_{apatite}$ permitted the definition of e.g. fishing grounds, quantification of the "sea spray" effect, and provenance analysis.

1. Introduction

– **bivariate analyses are insufficient for multi-isotopic data**
– extraction of spatial information from multi-isotope data is challenging
→ multi-isotope fingerprints can be defined using cluster analysis (here: **Gaussian Mixture Model (GMM) clustering**; see e.g. Göhring et al., 2016)
→ **similarity search**
→ individuals within a single cluster are more similar to each other with respect to their isotopic fingerprint than to individuals of other clusters

– **$\delta^{18}O_{phosphate}$, $^{87}Sr/^{86}Sr$, $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$, $^{208}Pb/^{207}Pb$, $^{206}Pb/^{207}Pb$ (data set I)**
→ spatial information, identification of probably non-local individuals (see e.g. Grupe et al., 2020; 2018)
→ problem: cremated skeletal remains do not allow to measure the thermally unstable $\delta^{18}O_{phosphate}$
→ the detection of non-local individuals and provenance analysis requires the definition of a **local bioavailable $^{87}Sr/^{86}Sr$ range (data set II**; see Lengfelder, in prep.; Lengfelder et al., 2019)

– **$\delta^{13}C_{collagen}$, $\delta^{15}N_{collagen}$, $\delta^{13}C_{carbonate}$, $\delta^{18}O_{carbonate}$, $\delta^{18}O_{phosphate}$, $\delta^{34}S_{collagen}$, $^{87}Sr/^{86}Sr$ (data set III)**
→ information on diet and drinking water, habitat (e.g. terrestrial vs. marine), provenance/migration
→ palaeoecological study in an ecological complex region at the Baltic coast
→ problem: "sea spray" effect causes a shift in isotope values towards more positive (seemingly more marine) values
→ "sea spray" effect validated for $\delta^{13}C_{carbonate}$, $\delta^{18}O_{carbonate}$, $\delta^{18}O_{phosphate}$, $\delta^{34}S_{collagen}$, and $^{87}Sr/^{86}Sr$ (Göhring, 2019; Göhring et al., 2019; 2018)

2. Material

<u>**isotopic data set I: GMM cluster analysis along the European alps**</u>
→ 30 sites along Inn-Eisack-Adige passage across the European Alps (Late Hallstatt and Fritzens-Sanzeno culture)
→ 184 (un-)cremated human remains, 79 cattle, 75 pig, 36 red deer
→ $\delta^{18}O_{phosphate}$, $^{87}Sr/^{86}Sr$, $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$, $^{208}Pb/^{207}Pb$, $^{206}Pb/^{207}Pb$, Sr and Pb concentration

<u>**isotopic data set II: strontium mixing model**</u>
→ $^{87}Sr/^{86}Sr$ and Sr concentration of modern environmental samples (wood, soil, and groundwater) from 49 sites along Inn-Eisack-Adige passage
→ archaeological reference material (46 sites): 223 humans, 66 cattle, 81 pig, 38 deer

<u>**isotopic data set III: GMM cluster analysis at the Baltic coast**</u>
→ sites: Viking Haithabu and medieval Schleswig at the Baltic coast
→ 306 human remains, 111 terrestrial mammals, 27 marine mammals, 46 fish, 177 birds
→ $\delta^{13}C_{collagen}$, $\delta^{15}N_{collagen}$, $\delta^{13}C_{carbonate}$, $\delta^{18}O_{carbonate}$, $\delta^{18}O_{phosphate}$, $\delta^{34}S_{collagen}$, $^{87}Sr/^{86}Sr$

Fig. 1: Map of the sample sites in Germany, Austria, and Italy (modified after a Google Earth map (12/14/2015)

3. Strontium isotope mixing model

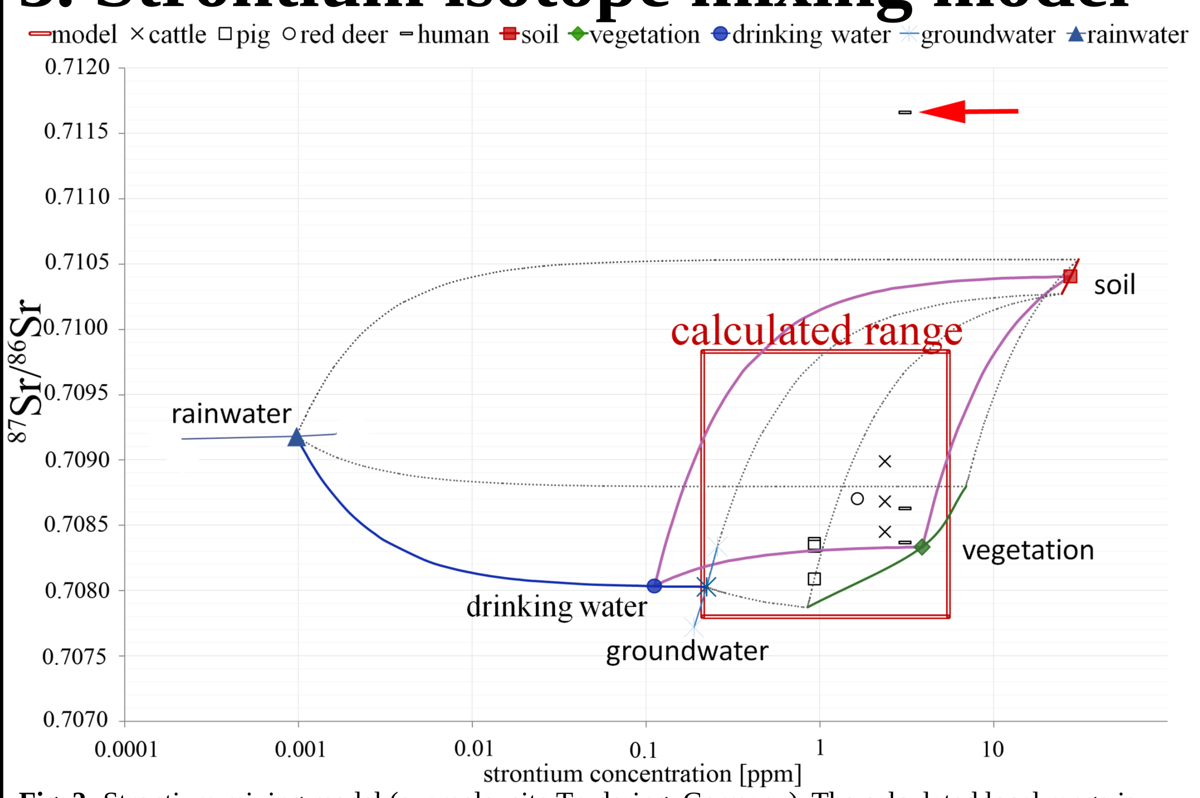

Fig. 2: Strontium mixing model (example: site Trudering, Germany). The calculated local range is circumscribed by the red rectangle (Lengfelder, in prep.)

Idea

– assessment of the Sr isotopic signature of local mammals based on modern environmental samples (wood, soil, groundwater) and literature values (global rainwater) using a concentration weighted mixing model
– comparison of predicted local $^{87}Sr/^{86}Sr$ spans with measured values of archaeological bone samples → detection of (non-)local individuals

Method (see Lengfelder, in prep.; Lengfelder et al., 2019)

– model based on $^{87}Sr/^{86}Sr$ ratios and Sr concentrations of vegetation (wood), soil, groundwater samples, and atmosphere/rainwater (Lengfelder et al., 2019)
– calculation of the concentration of a two-component mixture using measured $^{87}Sr/^{86}Sr$ ratios and Sr concentrations as well as approximated fractionation proportions
→ calculation of $^{87}Sr/^{86}Sr$ ratios of mixtures
– local variability taken into account (see Table 1)
– user-friendly Excel-based calculation tool (Lengfelder, 2020) available from:
https://www.for1670-transalpine.uni-muenchen.de/tools/

Results & Discussion

– example: site Trudering (Munich, Germany)
– all animal samples are included in the calculated mixing triangle (purple lines in Fig. 2) and within the calculated local range (red rectangle in Fig. 2)
→ probably local animal remains
→ archaeozoological vertebrate data (cattle, pig, red deer) are **precisely predicted by the model**
– 2 out of 3 humans fall into the environmental span and the calculated local range (Fig. 2)
→ 2 probably local humans
→ 1 **non-local** human (see red arrow in Fig. 2)
→ **model performed well in 72 % of 46 tested sites** (Lengfelder, in prep.)

Table 1: $^{87}Sr/^{86}Sr$ and concentration range used for the mixing model (Lengfelder, in prep.)

sample material	$^{87}Sr/^{86}Sr$ range	concentration range
vegetation	x ± 0.00045	x ± 3
soil	x ± 0.0001	x ± (0.1*x)
groundwater	x ± 0,000285	x ± (0.15*x)
rainwater	0.70916 - 0.70920	0.20 ppb - 1.75 ppb

4. GMM cluster analysis of cremated human remains

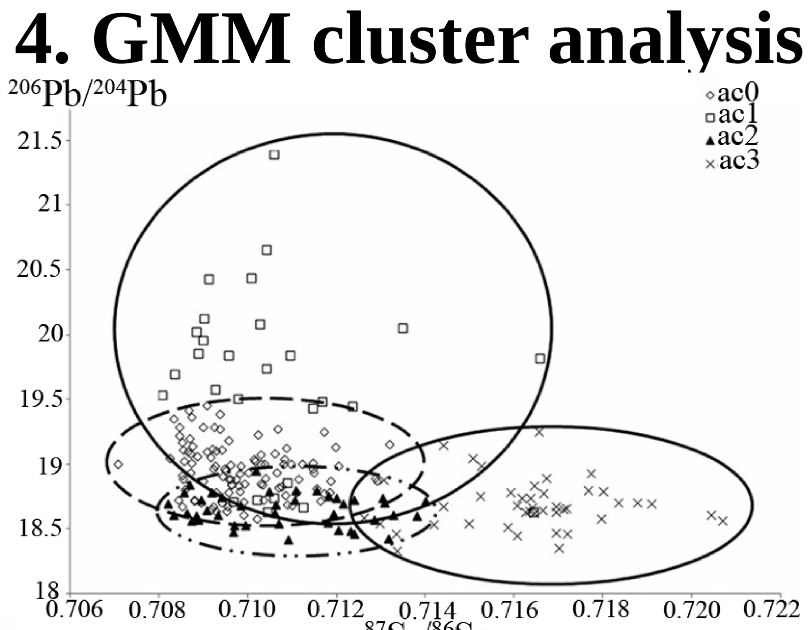

Fig. 3: Bivariate plot illustrating GMM clustering results (7 isotopic systems) for the animals from the Alpine transect; cluster 0: diamonds, cluster 1: squares, cluster 2: triangles, cluster 3: crosses (Grupe et al., 2018)

Idea

– multi-isotope fingerprints can help to identify (dis-)similarities between individuals
– Do animals group into isotopically defined micro-regions?
– Are the isotopic fingerprints of animal bones reflected by the human samples?
– Is a spatial assignment of cremated bones still possible ($\delta^{18}O_{phosphate}$ not available)?

Method (see Grupe et al., 2020; 2018)

– Gaussian Mixture Model (GMM) clustering of multi-isotope data of uncremated animal (7 isotopic ratios) and both uncremated and cremated human (6 isotopic ratios) bone samples
– GMM based on multivariate normal distribution and EM (Expectation Maximization) algorithm

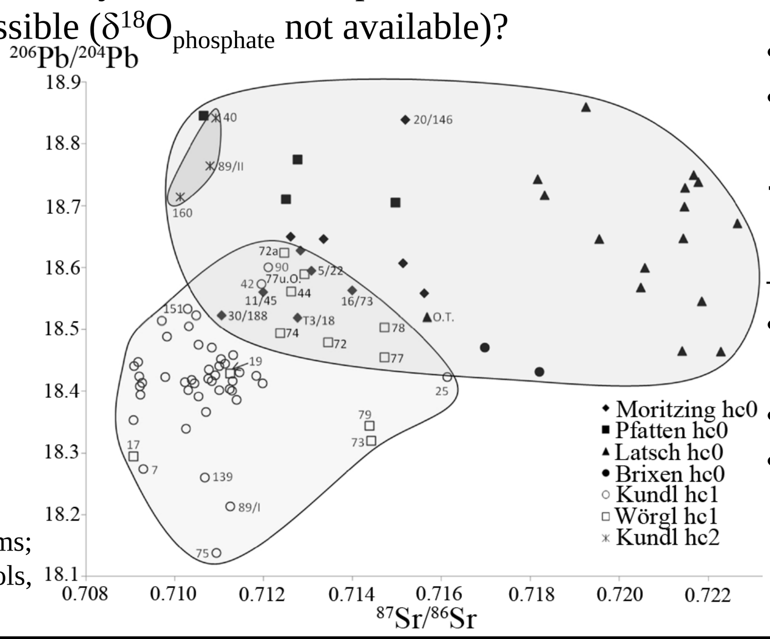

Fig. 4: Bivariate plot illustrating GMM clustering results (6 isotopic systems; without $\delta^{18}O$) for the humans from the Alpine transect. cluster 0: filled symbols, cluster 1: open symbols, cluster 2: crosses (Grupe et al., 2020)

Results & Discussion

– GMM analyses identified four animal clusters (ac0 - ac3; Fig. 3) and three human clusters (hc0 - hc2; Fig. 4; without $\delta^{18}O_{phosphate}$) with high probability ($p > 0.9$)
– **animal clusters** (Grupe et al., 2018):
- ac0: mostly excavated in the north → show declining frequency to inneralpine and southern regions
- ac1: no animal found in the south
- ac2: nearly evenly distributed between sites to the northern and inneralpine regions; only two individuals found in the south (probably imported)

→ ac3: no animal found in the north
→ **clusters reflect spatial distribution** along alpine transect

– **human clusters** (Grupe et al., 2020; 2018):
- hc0: all uncremated human remains from Southern Tyrol (n =33; Moritzing, Latsch, Pfatten, Brixen) → **high similarity to animal cluster ac3**
- hc1: cremated human remains from the Inn Valley (n=56; Kundl, Wörgl)
- hc2: three individuals from Kundl → **immigrants** (archaeologically conspicuous)

→ **GMM analysis using Sr and Pb permits allocation of samples** along the alpine transect (inner Alpine vs. Southern Tyrol) **even without $\delta^{18}O$ values**

5. GMM cluster analysis in a palaeoecological study

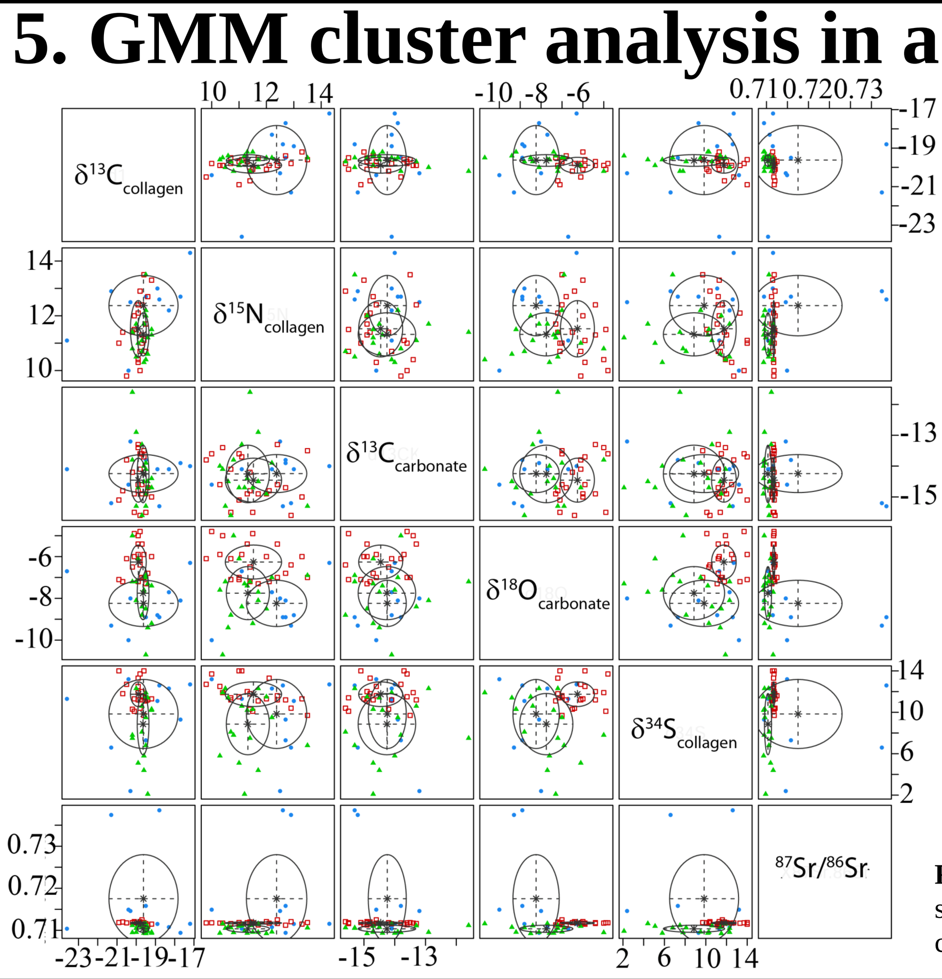

Fig. 5: Bivariate plots illustrating GMM clustering results (6 isotopic systems) for the humans from Haithabu and Schleswig (cluster 1: blue circles, cluster 2: red squares, cluster 3: green triangles)

Idea

– use GMM cluster analysis for provenance analysis of humans, to identify e.g. fishing grounds or migration of birds, and highlight the impact of the "sea spray" effect on terrestrial individuals (see Göhring, 2019; Göhring et al., 2016)

Method (see Göhring et al., 2016)

– GMM clustering of multi-isotope data (up to 7 dimensions) of archaeological humans and animals
– R package "mclust" (Scrucca et al., 2016)

Results & Discussion

– **human clusters** (Fig. 5):
- cluster 1: enriched in $\delta^{15}N_{collagen}$ and ^{87}Sr, depleted in $\delta^{18}O_{carbonate}$ → **probably not local**
- cluster 2: enriched in $^{18}O_{carbonate}$ and $^{34}S_{collagen}$
  → marine impact = **"sea spray" effect**

– **bird clusters** (Fig. 6):
- cluster 1: enriched in $^{18}O_{carbonate}$
  → **many domesticated birds** (chicken, goose), mainly terrestrial → **"sea spray" effect**
- cluster 2: **"outlier"** cluster (n = 5)
  → different diet → non-local individuals?
- cluster 3: enriched in all systems
  → **mainly piscivores** (e.g.cormorant, guillemot, white-tailed eagle)
  → **marine signal**
- cluster 4: depleted in all systems
  → **many archaeozoologically non-local species** (winter visitors, imported)

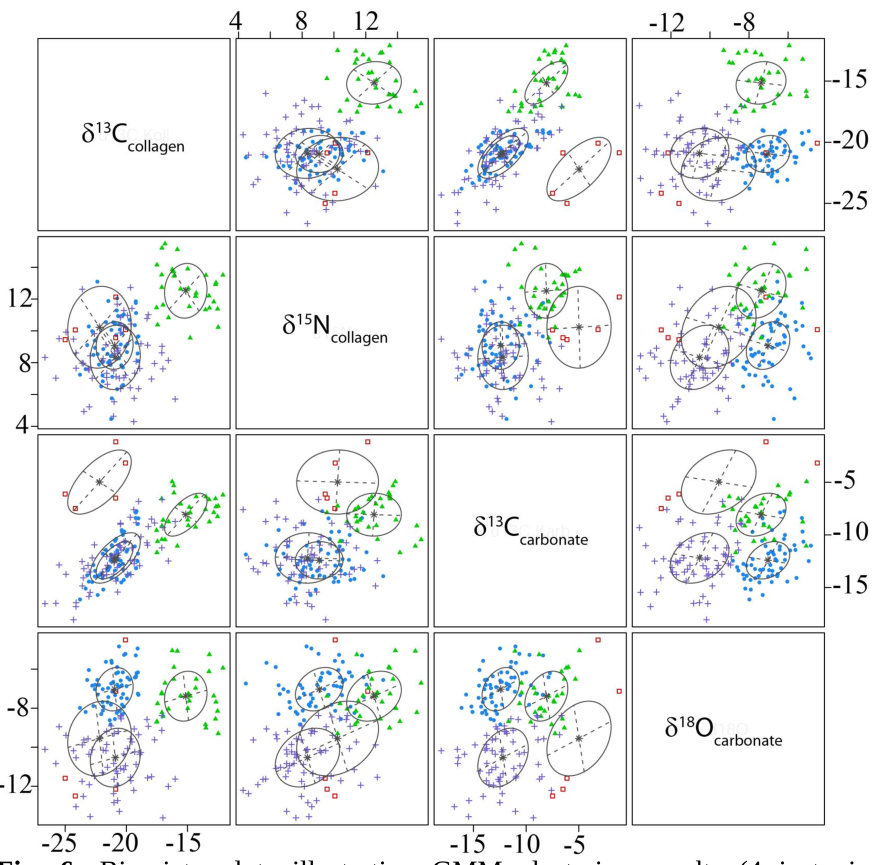

Fig. 6: Bivariate plots illustrating GMM clustering results (4 isotopic systems) for the birds from Haithabu and Schleswig (cluster 1: blue circles, cluster 2: red squares, cluster 3: green triangles, cluster 4: violet pluses)

6. Conclusion

– Sr mixing model based on environmental samples is a new tool for the prediction of local $^{87}Sr/^{86}Sr$ ratios of mammals
– Sr mixing model allows the identification of non-local individuals
– GMM clustering enables detection of multi-isotopic (dis-)similarities of both archaeological human and animals
– GMM identifies e.g. spatial distribution patterns, non-local individuals, migration/trade and individuals affected by the "sea spray" effect
– GMM reveals new information in multi-isotope data which are not visible in bivariate analyses
→ archaeometry will benefit from strontium mixing model and cluster analyses in future studies
→ advice to use strontium mixing model and GMM cluster analysis, which has been proven for multi-isotope data

Literature

Göhring (2019): Anwendung KDD-basierter Methoden zur Interpretation multi-dimensionaler Isotopen-Fingerabdrücke. Dissertation, LMU Munich, Germany. **Göhring et al. (2019)**: Evidence for sea spray effect on oxygen stable isotopes in bone phosphate – Approximation and correction using Gaussian Mixture Model clustering. *Sci Total Environ 673, 668-684*. **Göhring et al. (2018)**: Palaeobiodiversity research based on stable isotopes: Correction of the sea spray effect on bone carbonate $\delta^{13}C$ and $\delta^{18}O$ by Gaussian Mixture Model clustering. *Palaeo3 290, 673-686*. **Göhring et al. (2016)**: Using Gaussian Mixture Model clustering for multi-isotope analysis of archaeological fish bones for palaeobiodiversity studies. *Rapid Commun Mass Spectrom 30(11), 1349-1360*. **Grupe et al. (2020):** The genesis and spread of the early Fritzens-Sanzeno culture (5th/4th cent. BCE) – Stable isotope analysis of cremated and uncremated skeletal finds. *J Archaeol Sci Rep, 29, 102121*. **Grupe et al. (2018):** Multi-isotope provenancing of archaeological skeletons including cremations in a reference area of the European Alps. *Rapid Commun Mass Spectrom 32(19), 1711-1727*. **Lengfelder (in prep.):** Der Einfluss der Umwelt auf die Isotopensignaturen von Mensch und Tier. Dissertation, LMU Munich, Germany. **Lengfelder (2020):** Modelling local vertebrate Sr isotopes- a Sr calculation tool. Version 2. https://www.for1670-transalpine.uni-muenchen.de/tools/. **Lengfelder et al. (2019):** Modelling strontium isotopes in past biospheres – Assessment of bioavailable $^{87}Sr/^{86}Sr$ ratios in local archaeological vertebrates based on environmental signatures. *Sci Total Environ 648, 236-252*. **Scrucca et al. (2016):** mclust 5: Clustering, Classification and Density Estimation Using Gaussian Finite Mixture Models. *The R Journal, 8(1), 289-317*.