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The universal imprint of oxygen isotopes can track the origins of seafood

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ABSTRACT

Identifying the provenance of seafood is key to helping authorities combat seafood fraud, but current tools are predominantly applied and developed on a species-specific basis. This study investigates how multiple marine taxa could be geolocated at global scales by exploiting stable oxygen isotope compositions in carbonate biominerals ($\delta^{18}O_{biomin}$), where we expect to see universally-expressed and predictable spatial variation in $\delta^{18}O_{biomin}$ values across taxa. We constructed global ocean isoscapes of predicted $\delta^{18}O_{biomin}$ values specific to fish (otoliths), cephalopod (statoliths) and shellfish (shells), and a fourth combined "universal" isoscape, and tested their capacity to predict variations in $\delta^{18}O_{biomin}$ values among known-origin samples. High correspondence between isoscape-predicted $\delta^{18}O_{biomin}$ values and a compiled database of measured, geo-referenced values (3954 datapoints representing 68 species) indicated that this $\delta^{18}O_{biomin}$ approach works well, particularly in regions with highly-resolved projections of seawater $\delta^{18}O$ composition. The universal isoscape showed similar accuracy compared to taxon-specific isoscapes when predicting $\delta^{18}O_{biomin}$ values, demonstrating exciting potential for universal provenance applications. We tested the universal framework on a case study region, with machine learning models used to infer sample origins between regions of divergent climates (Tropical Asia vs Temperate Australasia) and similar climates (Temperate Asia vs Temperate Australasia). Classification accuracy averaged 75.3% between divergent regions and 66% between similar regions. However, when endothermic tuna species were removed from the analyses, accuracy increased up to 90% between divergent regions. This study is a first step towards developing universal chemical markers to support a more inclusive and global approach to verifying provenance of seafood.

Highlights:

- Universal chemical markers investigated for seafood provenance applications
- Global ocean oxygen isoscapes were constructed and tested
- Isoscape models accurately identified multiple taxa to geographical origins
- First steps towards a more inclusive and global provenance approach

Keywords:

Authentication; Biominerals; Fingerprinting; Global model; Isoscape; Provenance

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1. INTRODUCTION

A healthy and sustainable food system is underpinned by knowing where food comes from, and how it is produced. However, wild-caught seafood is a hard-to-monitor shared resource, and seafood supply chains are often opaque and complex (Fox et al., 2018; Kittinger et al., 2017), which in turn makes seafood particularly vulnerable to fraud. Provenance fraud occurs when consumers or businesses are intentionally deceived about where seafood is caught or its production method, with products often substituted with lower-quality, lower-value versions, or from locations with fewer regulations regarding sustainability or ethics concerns. Such substitution threatens our food system by risking sustainability, safety, and consumer confidence (Lindley, 2020; van Ruth et al., 2017). Provenance fraud can lead to over-exploitation of stocks, which undermines the long-term sustainability of fisheries, the balance of marine ecosystems, and ultimately, harms seafood businesses that rely on these resources (Jacquet & Pauly, 2008; Kompas et al., 2010). Substitute seafood products can also have safety and health implications for consumers, by containing hidden pathogens, banned antibiotics, unlisted allergens or reduced nutrient profiles (Gopi et al., 2019). While many factors influence fisheries sustainability, validating the provenance of seafood empowers authorities to combat seafood fraud and ensures consumers can make an informed choice about the seafood they eat.

The provenance of food can be authenticated using a range of techniques, including DNA profiling (Calosso et al., 2020; Rasmussen & Morrissey, 2008), paper-based and digital tracing, such as blockchain (Howson, 2020), and chemical profiling (Duarte et al., 2022; Martino et al., 2022). While chemical approaches for provenance testing are well-established in terrestrial food forensics, uptake in the aquatic food industry has been slower but is increasingly indicated to have wide potential (Gopi et al., 2019). Natural chemical tracers within seafood tissues can be effective at identifying geographical origins as they are carried

unaltered within the product itself and are difficult to falsify. However, existing provenance tools can be prohibitive for many seafoods due to the high cost of developing reference datasets, which are typically optimised for specific species, regions, or supply chains (Kelly et al., 2005). Reference databases are more commonly adopted as standard practise in terrestrial food systems as land-owning producers have more economic incentive to characterise and protect produce originating from land they own (e.g. Monahan et al., 2018; Soares et al., 2017). In marine systems with common access to waters and migratory species, there is less financial incentive for producers to fund chemical or genetic characterisation of specific geographic areas. Furthermore, small-scale fisheries comprise an estimated 70% of the total world catch (Kolding et al., 2014), with many targeting low value stocks, operated by independent fishers with no access to forensic tools, and in exploited waters of developing countries. Traceability methods that rely on development and maintenance of cost-intensive fishery-specific datasets are thus largely inaccessible to these producers. Therefore, a shared provenance system capitalising on consistent inherent properties across taxa could ease the burden of species-specific, cost-prohibitive provenance systems, and improve accessibility of seafood provenance for lower-income regions, smaller industries, or lower-value seafood species.

A shared provenance system could be developed using *universal* chemical markers. Here, we define a universal chemical marker as having predictable spatial variation coupled with common expression among taxa (Doubleday et al. in review). Essentially, a universal marker is primarily influenced by the environment whilst species-specific modifications of the environmental signal (e.g. physiological 'vital effects' that vary across taxa) are minimal, so that different taxa living in the same region would share a common chemical value in their tissues. The stable isotope composition of oxygen in calcium carbonate ($CaCO_3$) biominerals (expressed as $\delta^{18}O_{biomin}$ values) has such properties and is a promising candidate for universal provenance applications across broad geographic scales (100s to 1000s km). Oxygen is incorporated into marine animals from seawater with an isotopic composition ($\delta^{18}O_{water}$) that is

relatively constrained and predominantly varies as a function of salinity, water mass identity, and surface evaporation rate (Craig & Gordon, 1965). In general, typically lower $\delta^{18}O_{water}$ values are found in higher latitudes due to discharge of river water and glacial meltwater with low $\delta^{18}O$ values, and higher $\delta^{18}O_{water}$ values are observed in highly evaporative regions, such as some sub-tropical zones (Conroy et al., 2014; Singh et al., 2010; Voelker et al., 2015). Global compilations of seawater $\delta^{18}O_{water}$ values have been constructed (LeGrande & Schmidt, 2006), further constraining region-specific relationships between $\delta^{18}O_{water}$ values and salinity. Consequently, with high resolution salinity data, $\delta^{18}O_{water}$ values can be predicted with confidence on global scales. During biomineralisation, oxygen isotopes are fractionated with preferential incorporation of the lighter ^{16}O isotope. The extent of this fractionation depends on the temperature of mineral growth (Kalish, 1991; Kim et al., 2007), which in marine ectothermic organisms is the ambient seawater temperature. The form of the temperature-dependent equation of $\delta^{18}O_{biomin}$ has been investigated extensively as it forms the basis for palaeotemperature reconstructions (Zachos et al., 1994), with $\delta^{18}O_{biomin}$ thermometry equations experimentally derived for a range of fish (e.g. Geffen, 2012; Godiksen et al., 2010; Høie et al., 2004; Sakamoto et al., 2019; Thorrold et al., 1997), gastropod and bivalve species (e.g. Grossman & Ku, 1986; Nishida et al., 2015; Owen et al., 2008; Wanamaker Jr et al., 2007), as well as a single cephalopod species (Chung et al., 2020). However, there remains some debate on whether $\delta^{18}O_{biomin}$ values reflect ambient temperatures in fish with regional endothermy, such as some shark and tuna species (Graham & Dickson, 2004; Hane et al., 2020).

Given global scale predictions of $\delta^{18}O_{water}$ values and temperatures in the ocean alongside the well-established biogeochemical relationships, it is possible to create spatial models ("isoscapes") of predicted $\delta^{18}O_{biomin}$ values in tissues of marine animals (Trueman et al., 2012). Isoscapes are typically displayed as spatial maps of isotopic distributions and can be used to determine the most likely geographic origins of an organism by matching the isotopic composition of its tissue to area(s) on the isoscape with similar values. Isoscapes

have been used in both terrestrial and aquatic ecology to investigate animal movements and environmental histories (Artetxe-Arrate et al., 2021; Pearson et al., 2020; Trueman & Glew, 2019), but have had little application in marine food provenance.

Here, we propose that globally-predicted, universal $\delta^{18}O_{biomin}$ markers are effective indicators of provenance for multiple marine taxa. Our taxa of interest included teleost fish, bivalves and gastropods (hereafter called shellfish), and coleoid cephalopods, which are critical components of the world's seafood supply. Our specific objectives were to 1) develop and compare three global ocean isoscapes of predicted $\delta^{18}O_{biomin}$ values in fish, cephalopods and shellfish, with a fourth "universal" multi-taxa isoscape, 2) test predicted isoscape-derived $\delta^{18}O_{biomin}$ values through comparing against a global database of measured, geo-referenced $\delta^{18}O_{biomin}$ values, and evaluate predictors of variation in residuals; and 3) in a case study, assess the isoscape performance for geolocating multiple taxa between regions with divergent (Tropical Asia vs Temperate Australasia) or similar (Temperate Asia vs Temperate Australasia) climatic profiles.

2. MATERIALS AND METHODS

2.1 Development of isoscapes

Mechanistically-predicted global ocean isoscapes of $\delta^{18}O_{biomin}$ values were constructed using environmental datasets and established biogeochemical relationships from the literature. Three isoscapes were built specific to aragonitic calcium carbonate ($CaCO_3$) biominerals of fish (otoliths), shellfish (exoskeleton shells of bivalve and gastropod molluscs), and cephalopods (statoliths). A fourth "universal" isoscape was then created via an average of the taxon-specific isoscapes. All modelling and analysis was done in R (R Development Core Team 2008), with isoscapes developed using R packages raster (Hijmans et al., 2015), gstat (Pebesma, 2004), and rasterVis (Lamigueiro et al., 2022; Liaw & Wiener, 2002).

$\delta^{18}O_{biomin}$ values were predicted from literature datasets of sea surface temperature and $\delta^{18}O_{water}$ values. Sea surface temperatures (°C, 0.05° resolution) representing a depth of 20 cm were taken as weekly intervals from the ESA Sea Surface Temperature Climate Change Initiative through the Copernicus E.U. Copernicus Marine Service Information (Merchant et al., 2019). Data across a 4-year period (February 2017 – January 2021) were extracted and averaged across this period and transformed to 1° x 1° resolution using bilinear resampling. A global gridded data set of $\delta^{18}O_{water}$ values (‰, 1° x 1° resolution) were also obtained (LeGrande and Schmidt 2006). This gridded data set was constructed from direct $\delta^{18}O_{water}$ measurements combined with estimates calculated from regional $\delta^{18}O_{water}$ to salinity relationships in areas of sparse data.

The terms used in the thermometry equations for predicting the $\delta^{18}O$ values in biominerals included: $\delta^{18}O_{biomin.VPDB}$ indicating the predicted values of $\delta^{18}O$ in carbonates reported relative to Vienna Pee Dee Belemnite (VPDB), $\delta^{18}O_{water.VSMOW}$ indicating the $\delta^{18}O$ of seawater relative to Vienna Standard Mean Ocean Water (VSMOW), and SST indicating the sea surface temperature in Celsius (°C).

A single experimentally-derived thermometry equation for statoliths of cephalopods has been published and was used to predict $\delta^{18}O$ values in statoliths (Chung et al., 2020):

$$\delta^{18}O_{biomin.VPDB} = -0.20(\pm 0.005) \times SST(°C) + 2.88\ (\pm 0.14) + \delta^{18}O_{water.VSMOW}$$

Equation 1

A range of thermometry equations for otoliths of teleost fish species have been experimentally validated (Table S1). As we aimed to construct an isoscape that could be widely applied across species, an averaged thermometry equation was derived from these published experimental studies (Geffen, 2012; Høie et al., 2004; Kitagawa et al., 2013; Nakamura et al., 2020; Radtke et al., 1996; Sakamoto et al., 2017; Thorrold et al., 1997). The selection aimed to include a range of functional groups of marine fish and included those with equations in suitable algebraic forms. The averaged thermometry equation used to predict $\delta^{18}O$ values in otoliths was:

$$\delta^{18}O_{biomin.VPDB} = -0.2143\ (\pm 0.012) \times SST(°C) + 4.056\ (\pm 0.28) + \delta^{18}O_{water.VSMOW}$$

Equation 2

For shellfish, we first compared the suitability of two equation forms, an averaged thermometry equation averaged from validated aragonitic shells and one of the most widely used thermometry equation for shellfish, the biogenic aragonite calibration from Grossman and Ku (1986) which is derived from a range of species. The Grossman and Ku model was subsequently chosen due to lower residual sum of squares when comparing predicted $\delta^{18}O_{biomin}$ values against measured values (see Supplementary Information, Figure S1). The form of the Grossman and Ku equation used was modified by Hudson and Anderson (1989), with a 0.38 ‰ offset applied to compensate for differences in acid fractionation factors of calcite and aragonite (Caldarescu et al., 2021). The equation used to predict $\delta^{18}O$ values in shells was:

$$\delta^{18}O_{biomin.VPDB} = \frac{SST(°C) - 19.7}{-4.34} + \delta^{18}O_{water.VSMOW} - 0.38$$

Equation 3

Predicted $\delta^{18}O_{biomin}$ values were calculated for each 1° by 1° cell, and plotted as a global map using ggplot2 in R (Wickham, 2011). The universal biomineral isoscape was constructed by averaging the predicted $\delta^{18}O_{biomin}$ values per cell across the three taxon-specific isoscapes.

2.2 Testing of isoscapes

To test the correspondence between the isoscape models and measured $\delta^{18}O_{biomin}$ values, we used an expanded version of a previously compiled database of $\delta^{18}O_{biomin}$ values in fish, coleoid cephalopods and shellfish, that were collected alongside geographical harvest coordinates (Doubleday et al., in review). The database comprised of published $\delta^{18}O_{biomin}$ values represented as means per species per harvest location derived from the literature (1995 to 2021), as well as a smaller number of unpublished values, provided by the authors. This database was expanded for the current project through extracting individual datapoints of $\delta^{18}O_{biomin}$ from publications, where available, and represented wild-caught, wholly marine, subtidal taxa collected from depths of less than 500 m. However, due to the spatial resolution of the isoscapes along coastlines, some datapoints from coastal zones were ultimately excluded. As such, 3954 datapoints derived from 68 species were ultimately used to represent the measured values. The database represents data from both the whole carbonate structure or the period corresponding to the area of capture (i.e. marginal increment of otoliths of migratory species) where appropriate. To assess regional-specific trends in the isoscapes, measured values were assigned to groups corresponding to 10 ocean ecoregions – Arctic, Central Indo-Pacific, Mediterranean, Temperate Australasia, Temperate Northern Atlantic, Temperate Northern Pacific, Temperate South America, Tropical Atlantic, Tropical Eastern Pacific, and Western Eastern Pacific (Spalding et al., 2007). As one of the main objectives of

231 this study is the universal and practical application of the isoscape, shellfish species that had
232 calcitic shells, aragonitic shells, or shells that were a mixture of both calcite and aragonite
233 were included. All other measured values of carbonates represented aragonitic structures.

234 For each coordinate of the measured database values, a matching predicted $\delta^{18}O_{biomin}$
235 value was extracted from the isoscapes as a 1° by 1° cell mean around the coordinate of
236 capture. Residuals (differences) between the measured $\delta^{18}O_{biomin}$ values from the database
237 and predicted values from the isoscapes were calculated to investigate discrepancies between
238 model-predicted and measured values. Global maps of residuals were constructed using
239 inverse distance weighting interpolation.

240 To investigate sources of variation in discrepancies between model-predicted and
241 measured $\delta^{18}O_{biomin}$ values, linear models were used to explore partitioning of the residuals
242 among potential predictor variables. Predictors included ocean ecoregion, taxa, latitude,
243 longitude and year of capture. For shellfish models, the $CaCO_3$ polymorph (aragonite, calcite,
244 or combined) was also included as a predictor. Model residuals were checked for normal
245 distribution and homogeneity of variance, while collinearity of predictors was assessed using
246 variance inflation factors below >2.5. Due to geographical predictors being highly collinear,
247 two models were compared either excluding longitude and latitude or excluding ocean region
248 using Akaike's information criterion corrected for small sample sizes (AICc; Burnham and
249 Anderson 2004). Relative importance weights were derived to assess the proportional
250 contribution of each predictor to the variability of the dependent value (Grömping, 2006).
251 Estimated Marginal Means derived from the optimal model was then used to investigate
252 whether the residuals differed significantly ($p < 0.05$) between ocean ecoregion, taxa, and
253 polymorph after accounting for the influence of all other predictors using package emmeans
254 (Lenth et al., 2018). Pairwise comparisons were conducted using a Tukey adjustment.
255 Subsequently, otolith isoscape-derived values were re-run in a separate linear model to
256 assess if residuals for endothermic tuna species (*Thunnus* sp.) were significantly different to
257 ectothermic fish. The species in our dataset we tested were albacore tuna (*Thunnus alalunga*),

Atlantic bluefin tuna (*Thunnus thynnus*), Pacific bluefin tuna (*Thunnus orientalis*), southern bluefin tuna (*Thunnus maccoyii*), and yellowfin tuna (*Thunnus albacares*).

2.3 Case study: Assessing isoscape performance for seafood geolocation across Asia and Australia

A case study was conducted to assess the efficacy of using $\delta^{18}O_{biomin}$ values to geolocate seafood to geographical origins in a real-world scenario, and to compare accuracy between regions of divergent or similar climatic profiles and latitudes. Regions in eastern Asia and Australasia were selected as they represent markets with high levels of seafood trade and suitable environmental profiles. “Temperate Asia” and “Temperate Australasia” were selected to represent similar climatic profiles and latitudes, while “Tropical Asia” was selected to represent a divergent climate profile from the temperate regions (Figure 5). A machine learning approach using random forest was used to classify region of origin of measured $\delta^{18}O_{biomin}$ values harvested within the boundaries of study regions. The random forest algorithm determines classifications using the aggregated results of an ensemble of individual classification and regression trees built using a random bootstrap of the data. Random forest models was built using R package randomForest (Liaw & Wiener, 2002) for each taxon-specific isoscape and the universal biomineral isoscape. Grid values of $\delta^{18}O_{biomin}$ representing 1° mean latitude and longitude steps within regions of interest (Temperate Australasia, Temperate Asia, and Tropical Asia) were extracted from each isoscape and then used to train each model with k-fold cross-validation incorporated. Out-of-bag (OOB) errors were derived to measure the predictive capacity of the trained model using bootstrap aggregating, whilst Cohen's kappa coefficient was calculated as an additional metric of classifier performance. Subsequently, random forest models predicted the region of origin of measured $\delta^{18}O_{biomin}$ values with accuracies derived by comparing the resulting matrix of class probabilities to the known regional classifications. To assess if model accuracies were improved with the

exclusion of endothermic tuna, we re-ran the universal and otolith-isoscape models without *Thunnus* sp.

3. RESULTS

3.1 Development of isoscapes

Isoscapes of mechanistically-predicted $\delta^{18}O_{biomin}$ values were successfully constructed for fish, cephalopod, and shellfish, as well as a universal average isoscape (Figure 1). Global trends were similar among taxa, and at global scales spatial variance was dominated by thermal effects, with more positive $\delta^{18}O_{biomin}$ values closer to the poles and more negative $\delta^{18}O_{biomin}$ values closer to the equator (Figures S2B, S3).

3.2 Testing of isoscapes

The trends in residuals between predicted values derived from the isoscape model and measured $\delta^{18}O_{biomin}$ values from the database provided an understanding of the variation between regions and taxa. The measured $\delta^{18}O_{biomin}$ values spanned between -5.3 to 3.9‰, corresponding well with the isoscape ranges (Figure 2A). Although residual sum of squares were similar between the universal and taxon-specific isoscapes when separating out the taxa (±1), the universal isoscape produced lower average residuals for statoliths (1.2 vs 1.6 ‰) and otoliths (-0.07 vs -0.4 ‰), but higher residuals for shells (0.5 vs 0.4 ‰) (Figure 2B, Figure 3).

Linear regressions uncovered the key influences on the residuals of $\delta^{18}O_{biomin}$ values. The linear model derived from the universal isoscape explained 41.0% of the variance ($p < 0.001$). Ocean region was the dominant predictor, responsible for 82.2% of the model variance. This regional variation was also demonstrated through significant differences ($p < 0.05$) between many regions. The estimated marginal means of residuals were the most negative in the Arctic (-0.96 ‰) and Mediterranean (-0.73 ‰), whilst the most positive estimated marginal means were found in Temperate South America (1.32 ‰) and the Central Indo-Pacific (1.49 ‰) (Figure 4). The estimated marginal means of residuals in the Temperate Northern Atlantic (0.09 ‰) had values closest to 0, indicating the predicted values closely aligned with the measured values. Taxon was responsible for 15.2% of the model variance. Cephalopods (0.75 ‰) and shellfish (0.8 ‰) had similar positive estimated marginal means of

312 residuals, whilst the estimated marginal mean for the group 'fish' was significantly lower (-
313 0.06‰). Polymorph was responsible for 1.9% of the variance but was not significantly different
314 between aragonite, calcite, or combined mixes of the two. Year of capture was responsible for
315 0.5% variation but was also not significant.

316 The linear model assessing the taxon-specific isoscapes explained 46.7% of the
317 variance (R^2) of the residuals ($p < 0.001$). Compared to the universal model, taxon was a more
318 dominant influence in these individual isoscapes. While ocean region remained the dominant
319 influence of variation at 66.3%, taxon was responsible for 31.0% of variance in residuals,
320 polymorph 1.9%, and 0.6% to year of capture. The estimated marginal means of the residuals
321 for ocean regions largely aligned with those described for the universal model, however the
322 estimated marginal means for taxon were significantly different between all groups. Compared
323 to the universal models, estimated marginal means of residuals were closer to zero (0.62 vs
324 0.8 ‰) for the shell isoscape, more negative but further from zero (-0.39 vs -0.06 ‰) for the
325 otolith isoscape, and more positive but similarly further from zero (1.25 vs 0.75 ‰) for the
326 statolith isoscape. Like the universe isoscape regression models, the polymorph and year of
327 capture did not significantly influence residuals.

328 **3.3 Case study: Assessing isoscape performance for seafood geolocation across Asia**
329 **and Australia**

330 In the case study, measured test samples (n = 1097, species = 16) were classified back to
331 geographical source between regions of both divergent (Tropical Asia) and similar (Temperate
332 Asia, Temperate Australasia) climatic profiles and latitudes (Figure 5). Lower values of
333 Cohen's kappa coefficients compared to classification accuracies were observed, primarily
334 due to class imbalances. For the universal isoscape model (OOB: 15.6%, Kappa: 0.2) overall
335 accuracy at classifying samples back to region of origin across the three regions was 50.5%
336 (554 out of 1097 samples) (Table 1). Samples could be correctly distinguished between
337 tropical and temperate zones with 72.6% (796 out of 1097 samples) accuracy, but this
338 accuracy rate dropped to 63.4% (434 out of 684 samples) when attempting to distinguish

between the two temperate zones (Kappa: 0.56). Across the taxon-specific models (mean OOB 13.5%, Kappa: 0.17), mean overall accuracy across the three regions was 54.4% (Table 1). For the otolith isoscape model, samples could be distinguished between tropical and temperate zones with a 58.5% (304 out of 520 samples) success rate. When discriminating between the two temperate zones, accuracy was 84.2% (107 out of 127 samples; Kappa: 0.55). For the statolith isoscape model, samples were distinguished between tropical and temperate zones with a 70% (28 out of 40 samples) success rate, and between the two temperate zones with 50% (10 out of 20 samples) correctly distinguished (Kappa: 0.53). For the shell isoscape model, no measured Tropical Asia samples from the database were available so only temperate samples were tested against the model. These included temperate samples were 100% correctly classified as temperate rather than tropical (537 out of 537 samples), whilst 62% (333 out if 537 samples) of these samples were correctly distinguished between the two temperate zones (Kappa: 0.5).

The tuna species were found to influence both the linear models and accuracy of the classification models. The otolith isoscape linear model that included a *Thunnus sp.* predictor variable showed that the estimated marginal means of residuals between predicted and measured $\delta^{18}O_{biomin}$ values were significantly more negative (-1.08 ‰) for the tuna group than the other teleost fish (0.2 ‰). When the classification models from universal and otolith isoscapes were rerun with *Thunnus* excluded, correct classifications increased when distinguishing between the temperate and tropical zones. The universal model increased from 72.6% to 90% (689 out of 766 samples) accuracy, and the otolith model increased from 58.5% to 69.3% (131 out of 189 samples) accuracy. However, excluding *Thunnus* did not alter the classification success between the two temperate zones.

4. DISCUSSION

Oxygen isoscapes were successfully applied to track the geographical origins of a broad range of marine species. The universal isoscape was a more accurate predictor of geographic origin than taxon-specific isoscapes for cephalopods and fish, whilst the shell isoscape was a more accurate predictor for shellfish. Classification models developed from isoscapes generally performed best when assigning animals back to their region of origin between test regions with high temperature and latitudinal contrast (i.e., tropical versus temperate waters). Our results indicate that oxygen isoscapes are most powerful for geolocating animals over larger latitudinal gradients (100s to 1000s km), where there are known regional differences in water temperature and salinity. While traceability tools are typically species or supply-chain specific, these results demonstrate the potential of oxygen isotopes for a universal and combined provenance approach for marine animals.

The promising implication of the universal isoscape being comparable, and sometimes more accurate, at predicting $\delta^{18}O_{biomin}$ than the taxon-specific isoscapes is that it demonstrates suitability for a wide-range of species without the need for prior taxon-specific collation of reference data. This higher accuracy is likely assisted by the averaging of a range of thermometry equations to develop the isoscape. While consistent negative linear relationships are demonstrated between $\delta^{18}O_{biomin}$ values and temperature, there are minor species and taxonomy variation in equation coefficients, particularly intercepts (Chung et al., 2020; Kitagawa et al., 2013; Shirai et al., 2018), potentially owing to differences in haemoglobin oxygen-binding affinity, or oxygen sources within body fluids (Macdonald et al., 2020). Differences in experimental set-up, such as experimental temperature range, alongside measurement and statistical errors, may also have had a minor influence on equation coefficients between species. Whilst there are examples of isoscapes being effectively developed and applied using a thermometry equation validated for a single-species (Artetxe-

Arrate et al., 2021; Pearson et al., 2020; Trueman et al., 2012), the idea here is that a combined equation may be more likely to be physiologically-relevant to a randomly chosen species on average. Further testing will help to refine the averaged equation for optimal applications across species and taxa. In particular, most experimental studies validating the relationship between temperature and $\delta^{18}O_{biomin}$ values were derived from temperate or sub-arctic temperature ranges and study species (Geffen, 2012; Høie et al., 2004; Radtke et al., 1996), although there are a couple of exceptions (Chung et al., 2020; Kitagawa et al., 2013). It is evident that more experimental research is needed to define thermometry equations specific to tropical species, which would allow for more accurate use of $\delta^{18}O_{biomin}$ as a universal proxy.

For endothermic fish like tuna our results show that $\delta^{18}O_{biomin}$ values need to be applied cautiously. Residuals between measured and predicted $\delta^{18}O_{biomin}$ values were significantly more negative for *Thunnus* species in our database compared to the other fish species and subsequently, when these samples were excluded from the classification models, accuracy improved. Many tuna species can elevate their muscle, visceral, and cranial temperatures using counter-current heat exchangers known as retia mirabilia (Graham & Dickson, 2004; Malik et al., 2020). As such, $\delta^{18}O_{biomin}$ values could reflect elevated, internal temperatures rather than just environmental temperatures, although it is difficult to consistently predict to what extent as the thermoregulatory ability of tuna can vary with somatic size and ambient ocean temperatures (Kitagawa et al., 2006). While further research is needed to determine the extent that endothermy affects $\delta^{18}O_{biomin}$ values for provenance applications, endothermy is a rare phenomenon in fish and is only relevant to tuna, billfish and a small number of shark species (Madigan et al., 2015).

The shells of bivalves and gastropods can comprise of either calcite or aragonite polymorphs, or a combination of both, which can influence $\delta^{18}O_{biomin}$ – temperature relationships due to differing fractionation factors. However, it was found that this variability in $CaCO_3$ polymorphs has minimal impact on the accuracy that can be achieved using $\delta^{18}O_{biomin}$

values as a provenance tool. No significant differences were found in $\delta^{18}O_{biomin}$ values between polymorphs in the bivalves and gastropods. While the coefficients of the oxygen thermometry equation vary based on the polymorph, we found that for broader scale provenance purposes the variation in $\delta^{18}O_{biomin}$ values due to polymorph type was small compared to spatial variation. This result is backed by a previous field study that found minimal isotopic differences between calcite and aragonite layers in different gastropod and bivalves species growing in the same environment (Lécuyer et al., 2012). While identifying and separating the polymorphs prior to isotopic analysis can be achieved (i.e., using x-ray diffraction and mechanically separating calcite and aragonite layers), this would be a time-consuming and costly process. The consistency of results here between calcite and aragonite structures indicates that for universal applications, where the aim is to save time and effort for industry and research, a combined system is likely optimal.

We were also interested in identifying whether universal geolocation is equally applicable across regions. Predictions of $\delta^{18}O_{biomin}$ values were most accurate in the Temperate Northern Pacific and Temperate Northern Atlantic regions but least accurate in the Central Indo-Pacific and Temperate South America. This difference in accuracy between regions may stem from the variability in the number of measurements between regions used to derive the gridded data of $\delta^{18}O_{water}$ (LeGrande & Schmidt, 2006). While the dataset was developed with a relatively consistent coverage of directly measured $\delta^{18}O_{water}$ values in the northern Pacific and northern Atlantic Oceans, areas of sparse coverage are clearly evident such as along the southern coastline of South America and tropical Asia. This insight highlights that isoscape applications work best for regions with highly resolved current projections of $\delta^{18}O_{water}$ values. For regions with high residuals, predicted accuracies could be improved through incorporating localised seawater measurements of $\delta^{18}O_{water}$, or coupling detailed salinity projections with specific regional $\delta^{18}O_{water}$ – salinity relationships.

While we demonstrate the promising potential of universal oxygen isoscapes to geolocate seafood, there are current limitations. As the oxygen isoscapes developed here

cover fully marine, surface conditions, it is necessary to have a good understanding of the life-history, habitat use, and movement and migratory patterns of the study species, particularly identifying taxa with nearshore, subtidal or estuarine habitats or life stages. It is also possible that for highly migratory species, such as tuna, chemical markers may represent the migratory path rather than harvest location. Using age data alongside targeted sampling of new otolith growth is recommended to isolate $\delta^{18}O_{biomin}$ values that represent the harvest location. Moreover, the compiled database of measured geo-referenced samples used to test the isoscape was relatively scarce for offshore, pelagic samples compared to samples from inshore or shelf sea settings (Figure 2A). This is likely driven by coastal marine ecosystems accounting for 95% of the biomass of fisheries catch, likely in part due to easier accessibility and lower costs for fishers, despite covering only 22% of ocean area (Stock et al., 2017). As such, the assignment potential estimates presented should be cautiously applied to species in open ocean. Similarly, our isoscape models represent shallow ocean depths and further investigation is needed to test the accuracy of the approach in deeper water species. However, previous research indicates that predicted $\delta^{18}O_{biomin}$ values in otoliths are highly consistent in depths between 0 – 50 m, and while values became more positive in deeper depths, spatially-driven variation in $\delta^{18}O_{biomin}$ values still remained dominant (Artetxe-Arrate et al., 2021). Lastly, $\delta^{18}O_{biomin}$ approaches are clearly not applicable to seafood taxa that do not possess calcitic or aragonitic tissues. While most seafood taxa have $CaCO_3$ tissues, sharks and other elasmobranch fish are a key group that do not. However, phosphate and structural carbonate ions within bioapatite of skeletal structures of vertebrate animals are potential targets for applying universal oxygen isotope markers (e.g. Bryant et al., 1996). Furthermore, more research is required on understanding oxygen thermometry relationships in decapod crustaceans, which form a significant component of the seafood supply. Crustacean exoskeletons are mixture of organic matrices, calcite or Mg-calcite, and amorphous polymorphs of calcium carbonate (Luquet, 2012), so oxygen thermometry equations developed for fish and molluscs may not be as accurate.

In addition to these taxonomic considerations, the isoscapes presented here are constructed from annual means in temperature and salinity. However, seasonal fluctuations of temperature and salinity can influence $\delta^{18}O_{biomin}$ values and the magnitude of this influence would vary between regions. For example, when global $\delta^{18}O$ values in barnacle calcite were predicted separately for individual seasons, mid-latitudinal regions (30° to 60°) with large annual temperature gradients had the largest differences in isotope values (up to 1.9 ± 0.7 ‰) (Pearson et al., 2020). Conversely, minimal changes in isotopic values were found in the tropics and southern polar regions, where seasonal temperature fluctuations are limited. While in the current study higher residuals were not observed in mid-latitudinal zones, addressing season of capture is an area for refinement in future studies. Local variations in time-resolved $\delta^{18}O_{biomin}$ values may indeed provide finer-scale assignment potential, particularly for shellfish where high-resolution time-resolved sampling is relatively simple across the shell, and the sessile nature ensures that variation through time is directly linked to local variations in temperature (and possibly salinity).

To improve the predictive ability and spatial resolution of $\delta^{18}O_{biomin}$ values as a provenance tool, particularly among samples collected from similar latitudes, additional isotopic or elemental layers with universal properties could be introduced to the isoscape. Like $\delta^{18}O_{biomin}$ values, complementary universal chemical markers require predictable spatial variation coupled with common expression amongst species. While carbon isotopes ($\delta^{13}C$) in biominerals are generally analysed concurrently with $\delta^{18}O$, they are unsuitable in this context as $\delta^{13}C$ values in seawaters are generally homogenous (~0 ‰) whilst values in carbonates can vary across individuals due to diet and metabolic influences (Chung et al., 2021; Chung et al., 2019; Martino et al., 2020). In contrast, neodymium isotopes ($^{143}Nd/^{144}Nd$ ratios expressed as ε_{Nd} values) are indicated to be driven by continental geology and thus exhibit distinct geographic profiles in the ocean with high spatial resolution (Jeandel et al., 2007). Recent research suggests that ε_{Nd} values in the soft and hard tissues of bivalve molluscs are highly successful at identifying the origins of individuals across even small spatial scales (10s

497 km) (Saitoh et al., 2018; Tanaka et al., 2022; Zhao et al., 2019). Concentrations of barium
498 ratioed to calcium (Ba/Ca) could also be effective for provenance as biomineral signatures
499 reflect seawater values (Bath et al., 2000; Walther & Thorrold, 2006), and are little affected by
500 individual life-history, such as physiology or diet (Grammer et al., 2017; Hüssy et al., 2021;
501 Martino et al., 2021). However, significant work first needs to be undertaken to estimate spatial
502 variations in these potential universal markers. Alternatively, chemical markers could be used
503 in conjunction with taxa-specific provenance methods, such as DNA markers or fatty acid
504 profiling. While using multiple methods and biomarkers to infer the marine food provenance
505 can strengthen the accuracy and precision of provenance assignment (Cazelles et al., 2021),
506 the additional time and financial costs need to balance with likely benefit. For example, a
507 recent meta-analysis of provenance testing using DNA and isotopic markers, found that for 3
508 of the 63 teleost fish species investigated a combined DNA-isotope approach reduced the
509 chance of misassignment (Cusa et al., 2022). The study thus concluded that using both DNA
510 and isotopes would be likely most useful where the provenance regions of interest include
511 water bodies that are isotopically similar (i.e. between Temperate Asia and Australasia in this
512 study) or where populations that are genetically similar.

513 Chemical markers are used extensively in ecological and fisheries applications (e.g.,
514 defining stock structure or fishery management units), but their application to address seafood
515 provenance fraud, as well as their integration into associated management and policy
516 decisions, is scarce (Camin et al., 2016; Cusa et al., 2021). This is also the case for DNA-
517 based approaches, whereby uptake by managers and policymakers for seafood
518 authentication has stagnated (Bernatchez et al., 2017; Cusa et al., 2021; Martinsohn et al.,
519 2019). For DNA-based approaches, roadblocks to uptake include limited awareness on the
520 true costs and benefits of DNA analyses (e.g. perceived analytical costs versus value of
521 confiscated catches and associated fines), as well as poor communication between scientists,
522 managers, and policymakers about the relevant methods (Bernatchez et al., 2017; Martinsohn
523 et al., 2019). We suggest that awareness and communication about chemical markers, and

524 the efficiencies that could be achieved using a universal approach, is also key to uptake,
525 alongside demonstrating tangible outcomes. For instance, universal markers could be applied
526 to distinguish multiple marine taxa from protected designations of origin (PDO), which has
527 been successfully achieved using a species-specific marker approach, e.g. the authentication
528 of mussels from Galicia (Costas-Rodríguez et al., 2010), or between key trade locations, as
529 demonstrated here between south-east Asia and Australia. Furthermore, a universal marker
530 approach negates the need for developing time-consuming reference databases for different
531 species and target regions (Li et al., 2016), which is a drawback of current chemical marker
532 methods and a roadblock to uptake by industry (Camin et al., 2016). However, incentives to
533 use provenance technology to support sustainable seafood practices do vary, with seafood
534 labelling policies and regulations being vastly different among jurisdictions (Lindley, 2021).
535 Involvement from PDO protection consortia, producers' associations, or eco-certification
536 organisations can drive real world applications (Camin et al., 2016). For example, the Marine
537 Stewardship Council used DNA barcoding to verify the species of 100s of sampled seafood
538 products collected from multiple countries (Barendse et al., 2019). Chemical markers,
539 alongside DNA-based approaches, are well regarded methods for delineating population
540 structure for fisheries management applications, therefore we are optimistic the universal
541 approach outlined here could achieved for seafood provenance applications through ongoing
542 communication with stakeholders and a coordinated international approach to method
543 development.

544

545 **4.1 Conclusions**

546 Here, we demonstrate how universal chemical markers and isoscape mapping approaches
547 can successfully geolocate a diverse range of seafood back to the region of origin. These
548 techniques have the potential to change the way we verify the provenance of seafood on a
549 more unified, global, and equitable scale. Oxygen isotope compositions in biominerals provide
550 an accurate tool for geolocating samples across large latitudinal and thermal gradients, but

could be augmented with additional isotopic and elemental layers or species-specific approaches (e.g. DNA markers) to improve the spatial resolution and precision of the framework. Furthermore, biomineral structures have several key advantages over soft tissues for seafood provenance applications: they permanently retain lifetime chemical fingerprints; do not decay; are often discarded or low-valued by the seafood industry; and are commonly routinely retained for other purposes, such as aging of stocks (e.g., otoliths). Additionally, we can draw upon the extensive ecological and fisheries literature developed using chemical profiling of biominerals for reconstructing environmental or movement histories. Validating the provenance of seafood products on a global scale is a large and complex undertaking due to the diversity of species, fisheries, and socio-economic drivers. Here, we present a more universal method that is applicable to multiple taxa and seafood products and could provide access to provenance for smaller or lower-income producers, and to species with lower commercial value. Provenance fraud continues to be a significant issue that threatens food security, equitable access to resources, food safety, and consumer confidence in seafood products. The development of universal chemical markers, such as oxygen isotopes, may be a valuable step towards a more inclusive, global approach for tracing the provenance of seafood.

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DATA AVAILABILITY – Data is available in supplementary information, except unpublished data which is available from the corresponding authors upon reasonable request.

DECLARATION OF INTERESTS - The authors declare no competing interests.

5. TABLES

Table 1 – Confusion matrix of class classifications with percentage accuracies from random forest models, derived from a universal biomineral oxygen isoscape and isoscapes specific to statoliths of cephalopods, otoliths of fish, and exoskeleton shells of shellfish (bivalves and gastropods). This case study investigated whether samples can be classified back to region of origin between divergent climates and latitudes (Tropical Asia) or similar climates and latitudes (Temperate Asia, Temperate Australasia). Accurately classified samples are indicated in bold.

		True class		
		Tropical Asia	**Temperate Australasia**	**Temperate Asia**
Isoscape	*Predicted class*			
Universal biominerals	**Tropical Asia**	**120**	0	8
	Temperate Australia	7	**89**	216
	Temperate Asia	286	26	**345**
	Class accuracy (%)	29.1	77.4	60.6
	Overall accuracy (%)			50.5
Cephalopod statoliths	**Tropical Asia**	**8**	0	0
	Temperate Australia	10	**10**	0
	Temperate Asia	2	10	**0**
	Class accuracy (%)	40.0	50.0	-
	Overall accuracy (%)			45
Fish otoliths	**Tropical Asia**	**185**	0	8
	Temperate Australia	1	**25**	0
	Temperate Asia	207	12	**82**
	Class accuracy (%)	47.1	67.6	91.1
	Overall accuracy (%)			63
Shellfish shells	**Tropical Asia**	**0**	0	0
	Temperate Australia	0	**30**	176
	Temperate Asia	0	28	**303**
	Class accuracy (%)	-	51.7	63.3
	Overall accuracy (%)			62.0

6. FIGURES

Figure 1 – Universal isoscape of predicted oxygen isotopes in biominerals ($\delta^{18}O_{biomin}$) of marine organisms. This isoscape represents an averaging of temperature-dependent fractionation equations of $\delta^{18}O_{biomin}$ from the statoliths of cephalopods, otoliths of fish, and exoskeleton shells of shellfish (bivalves and gastropods). Taxon-specific isoscapes are found in supplementary information (Figure S3).

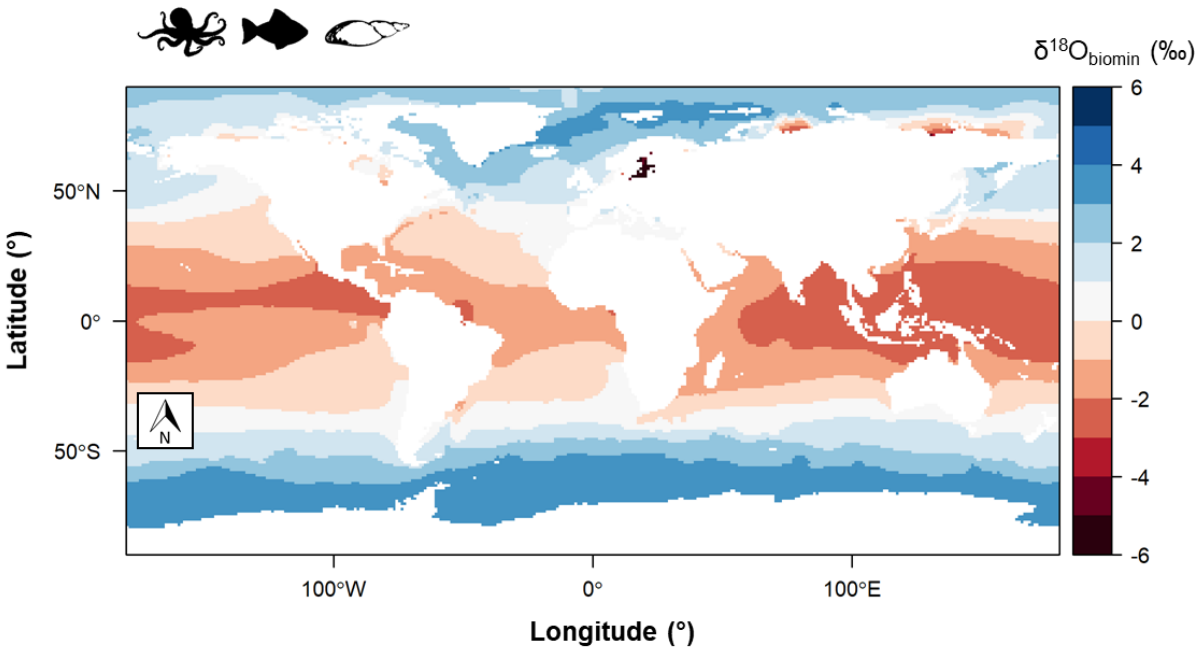


Figure 2 – Global maps representing A) measured oxygen isotopes ($\delta^{18}O_{biomin}$) values of geo-referenced samples (n = 3954, 68 species) from compiled database with ocean ecoregions labelled, and B) the residuals between measured and universal isoscape-derived predicted $\delta^{18}O_{biomin}$ values calculated per datapoint and then interpolated as a continuous surface onto a global map. The ocean ecoregions defined here include the Arctic (Arc), Temperate Northern Pacific (TeNP), Temperate Northern Atlantic (TeNA), Mediterranean (Med), Tropical Eastern Pacific (TrEP), Tropical Atlantic (TrAt), Central Indo-Pacific (CIP), Western Indo Pacific (WIP), Temperate Australasia (TeAus), and Temperate South America (TeSA) (Spalding et al., 2007).

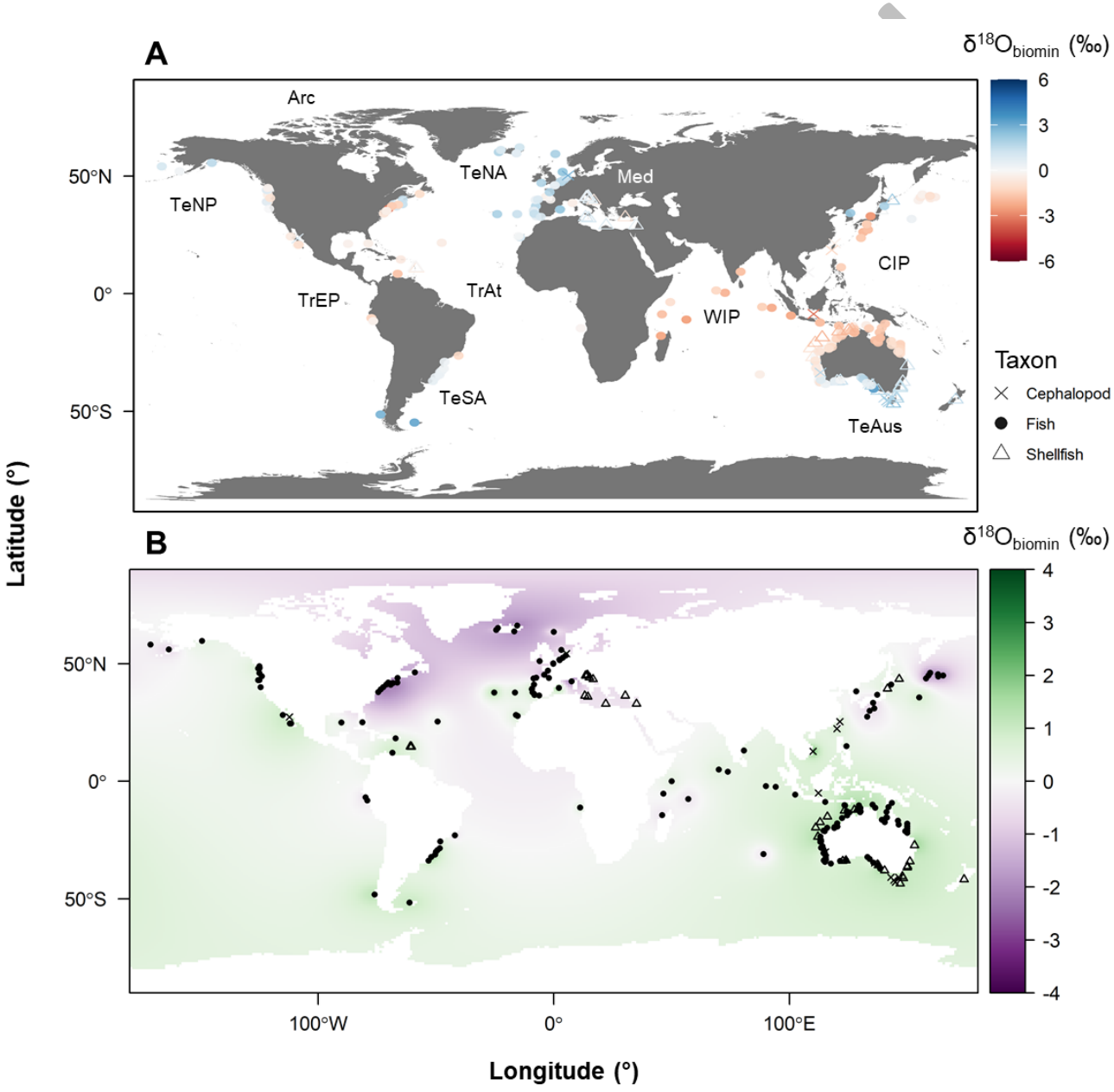


Figure 3 – The residuals between measured and taxon-specific isoscape-derived predicted oxygen isotope values in biominerals ($\delta^{18}O_{biomin}$) calculated per datapoint and then interpolated as a continuous surface onto a global map for the A) statoliths of cephalopods, B) otoliths of fish and C) exoskeleton shells of shellfish (bivalves and gastropods).

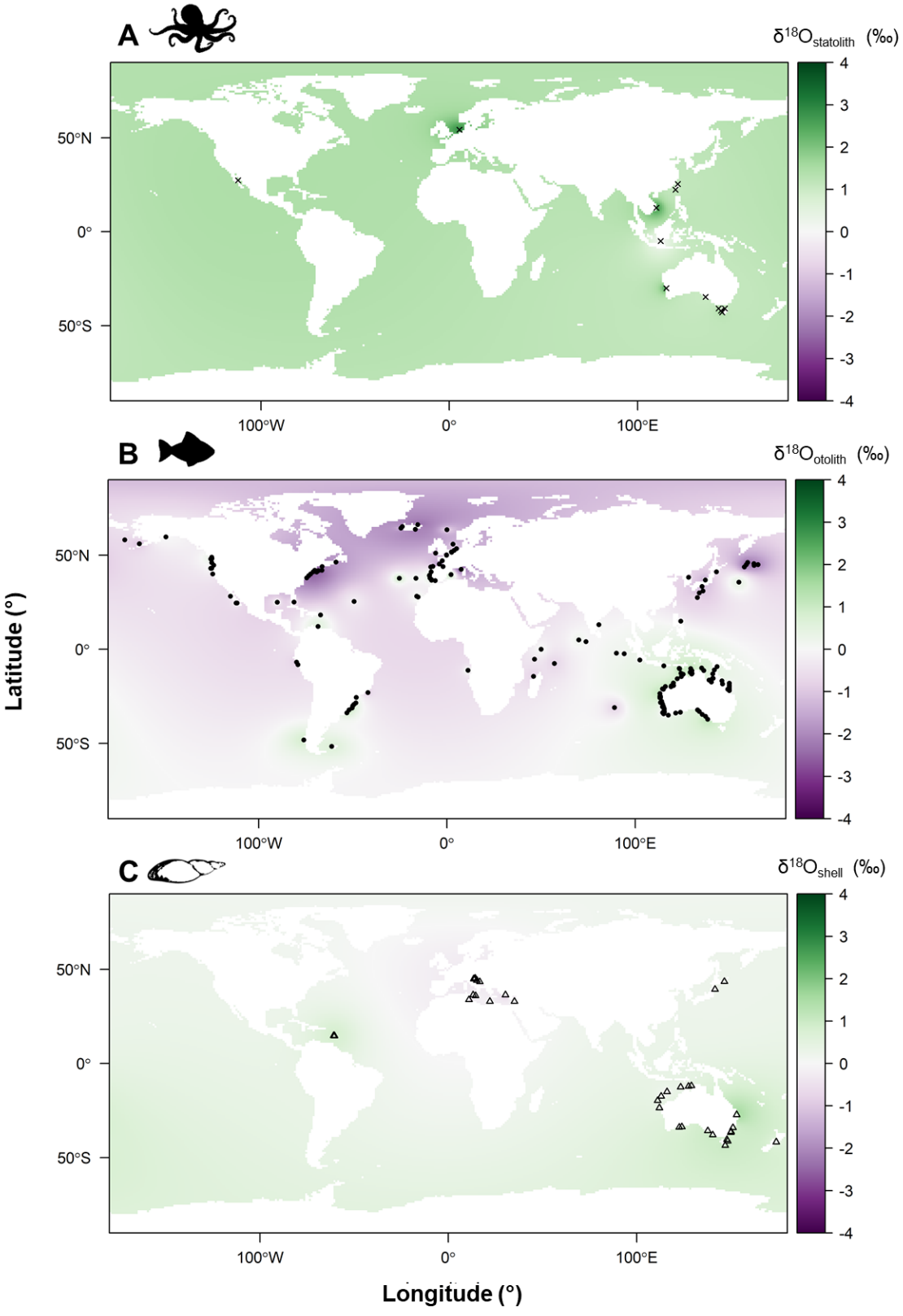


Figure 4 – Estimated marginal means of residuals between measured and universal isoscape-derived predicted oxygen isotope values in biominerals ($\delta^{18}O_{biomin}$) and separated into ocean bioregions and taxon of A) cephalopods B) fish, and C) shellfish. The bars indicate confidence intervals, whilst the red arrows are for comparisons between groups with overlaps across the horizontal axis indicating groups are not significantly different ($p > 0.05$). Dashed vertical lines indicate the estimated marginal mean of residuals averaged across regions for each taxon.

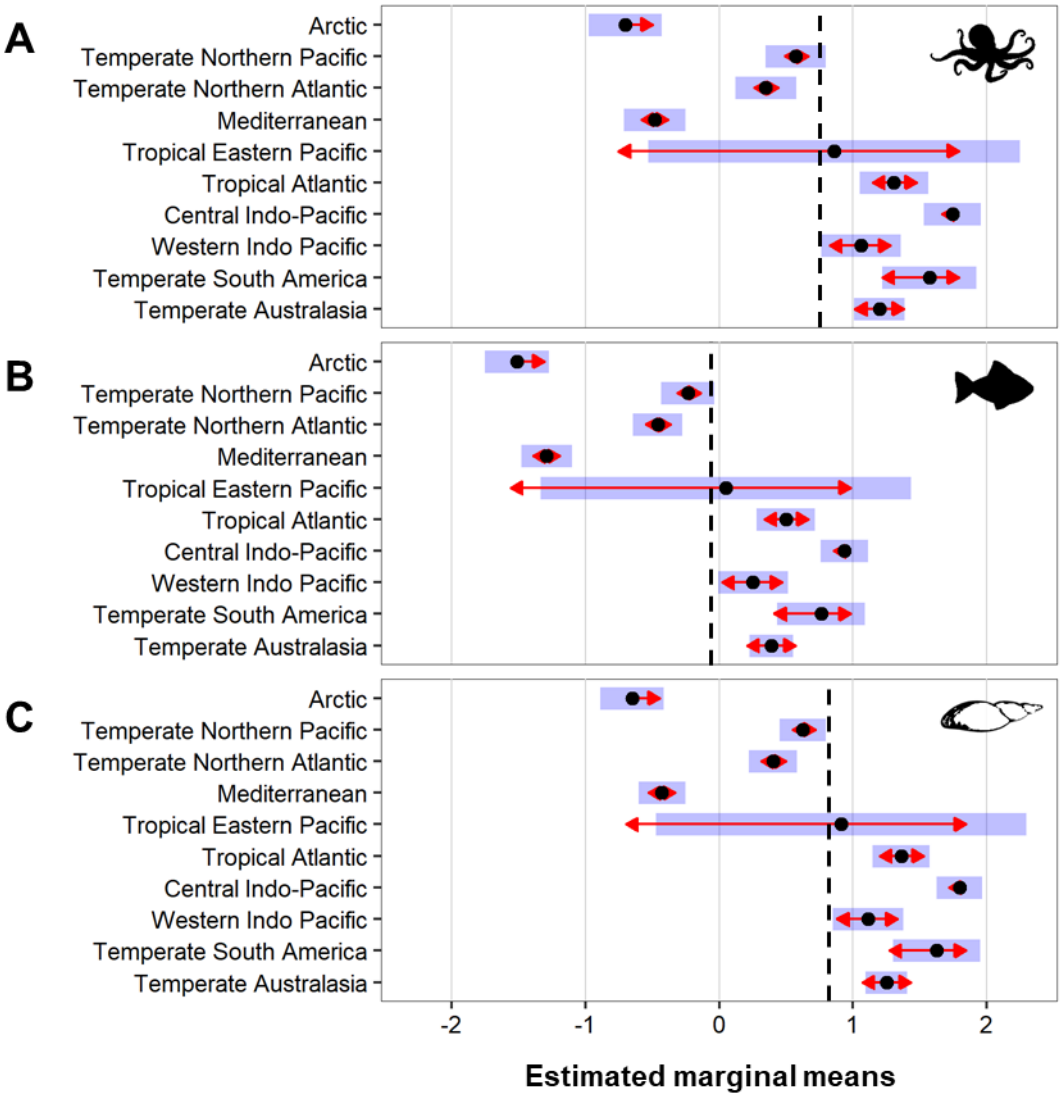
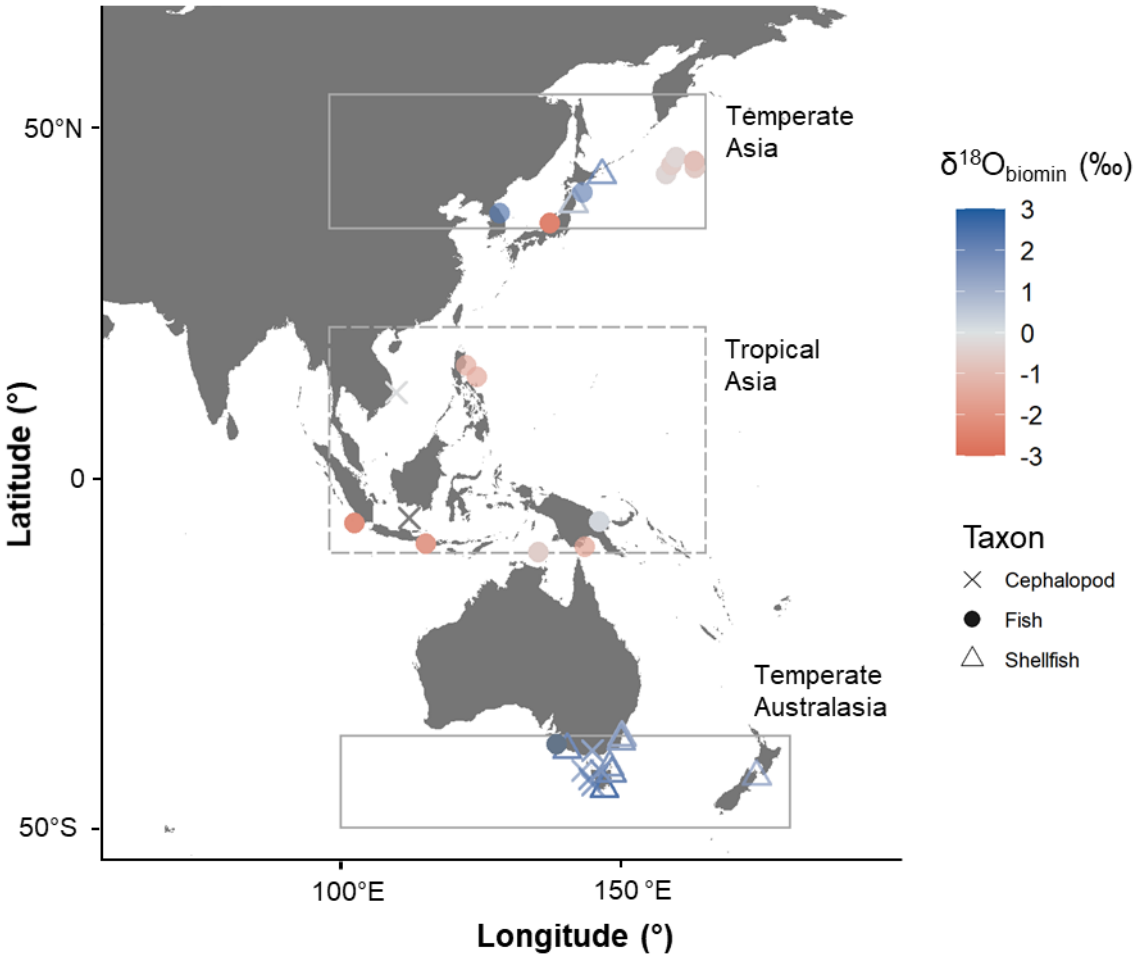


Figure 5 – Measured oxygen isotope values (n = 1097, species = 16) in biominerals ($\delta^{18}O_{biomin}$) of fish, shellfish, and cephalopods used in case-study to geolocate animals back to regions with climates and latitudes that are divergent (Tropical Asia) and similar (Temperate Asia, Temperate Australia). Also indicated are the boundary boxes of the case study regions where predicted $\delta^{18}O_{biomin}$ values were extracted from isoscapes and used to train random forest classification models.



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