Denudation partitioning in carbonate regions reveal climatic and tectonic drivers of carbonate landscape evolution

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ABSTRACT

Carbonate rocks are highly reactive and presumably have higher ratios of chemical weathering to total denudation relative to most other rock types. Their high chemical reactivity affects the first-order morphology of carbonate-dominated landscapes and their sensitivity to climate. However, there have been few efforts to quantify the partitioning of denudation into physical erosion and chemical weathering in carbonate landscapes such that their sensitivity to changing climate and tectonic conditions and its effect on topography remains elusive. Here, we compile bedrock and catchment-average cosmogenic calcite-\(^{36}\)Cl denudation rates and compare them to weathering rates. Local bedrock lowering and weathering rates are comparable, ~20 – 40 mm/ka, whereas catchment-average rates, which exist only for the Mediterranean, are ~2.7 times higher. This discrepancy is lower than the 15-fold difference in silicate-rich rocks illustrating that elevated weathering rates make denudation more spatially uniform in carbonate-dominated landscapes. Catchment-average denudation rates correlate well with topographic relief and hillslope gradient. Comparing these results with weathering rates shows that mechanical erosion processes contribute ~50% of denudation in southern France and ~70% in Greece and Israel. Our results indicate that the partitioning between largely slope-independent chemical weathering and slope-dependent mechanical erosion varies based on climate and tectonics. In humid, slowly uplifting regions, carbonates are associated with low-lying, flat topography because slope-independent chemical weathering dominates denudation. In contrast, in more arid climates with rapid rock uplift rates, carbonate rocks form steep mountains that facilitate rapid, slope-dependent mechanical erosion required to compensate for inefficient chemical weathering coupled with runoff loss to groundwater systems.
INTRODUCTION

Landscapes denude through a combination of mechanical erosion processes and chemical weathering. In most silicate-rich landscapes, erosion is the dominant denudation process. In contrast, carbonates are more susceptible to chemical weathering, likely resulting in a different partitioning of denudation into erosion and weathering for the same climatic and tectonic conditions. These differences in denudation partitioning should have pronounced effects on landscape morphology, presumably making carbonate landscapes more sensitive to differences in climate. However, few studies have quantified the relative contributions of erosion and weathering to denudation in carbonate-dominated landscapes, the effect of climate and tectonics on such partitioning, and its impact on topography.

The limited number of studies investigating denudation partitioning in carbonates stems largely from challenges in quantifying long-term mechanical erosion. Solution fluxes from dissolved loads and direct outcrop measurements with limestone tablets have been used extensively to quantify limestone weathering rates (Plan, 2005; Calmels et al., 2014). Recent advances in cosmogenic $^{36}$Cl production rate calibration and calculation (Schimmelpfennig et al., 2009; Marrero et al., 2016b) now allow for accurate calculation of total denudation rates in carbonates at the outcrop and catchment scale. Through the comparison of cosmogenic radionuclide (CRN) measurements with weathering rates, it is possible to isolate the contribution of mechanical erosion by subtracting weathering from total denudation rates (Ott et al., 2019), provided that the disparate integration timescales of each measurement have a negligible impact on measurement comparisons, and if CRN weathering biases can be accounted for (Ott et al., 2022).
Studies applying some combination of these techniques have arrived at different conclusions regarding denudation in carbonates. Several recent studies assumed that the effects of mechanical rock removal are negligible (Ryb et al., 2014a, 2014b; Avni et al., 2018), implying chemical weathering dominates carbonate denudation budgets. However, other studies found that mechanical processes likely play a critical role in carbonate denudation (Newson, 1971; Covington et al., 2015; Thomas et al., 2017; Ben-Asher et al., 2021). Here, we contribute to this growing effort by compiling and comparing available cosmogenic calcite-\(^{36}\)Cl denudation rate measurements with catchment average weathering rates collected from the same areas to quantify the partitioning between mechanical and chemical surface lowering in carbonate terrains across climatic and tectonic gradients. We use this analysis to illuminate how denudation partitioning varies as a function of climate and tectonics and highlight that carbonate regions are more susceptible to climate-topography interactions than silicate-rich rocks.

**METHODS**

**\(^{36}\)Cl denudation rate compilation**

We compiled 232 bedrock and 43 fluvial sediment \(^{36}\)Cl measurements from the literature to determine denudation rates at the outcrop and catchment scale, respectively (Fig.1, Tab. S1). The bedrock denudation rates were sampled from bedrock outcrops or amalgamated clasts on hillslopes or ridgetops and thus record local bedrock lower rates. In contrast, the alluvial sediment samples are assumed to be well-mixed and provide an average denudation rate across the upstream drainage area. \(^{36}\)Cl denudation rate calculation requires knowledge of the chemical composition of the host rock and the target mineral analyzed (Schimmelpfennig et al., 2009). To compare denudation rates from different studies, we compiled all chemical sample data from the literature or, if unavailable, contacted the authors and recalculated all denudation rates with
CRONUS calc v2.1 (Marrero et al., 2016a) (Tab. S1). Ott et al. (2022) showed that weathering can bias CRN measurements of soluble target minerals such as calcite, but tests with weathering corrections show that this effect is negligible for the samples considered here, therefore, we present the uncorrected denudation rates (see supplement).

![Fig. 1: (A) Location of $^{36}$Cl denudation rate measurements compiled for this study in the Mediterranean and globally (inset). *samples not included due to lack of location and compositional data. (B) Typical carbonate catchment in the Lefka Ori range on Crete, Greece, with partial forest cover and small landslide scars. (C) Typical low relief catchment on Crete.](image)

**Carbonate weathering rate calculations**

We calculated carbonate weathering rates for areas of published catchment-average denudation rates to infer the rates of landscape-scale mechanical erosion. Erosion can be assumed to equal denudation minus weathering because, despite deep solution features such as
caves, the majority of carbonate dissolution takes place close to the Earth’s surface (Gunn, 1981; Worthington and Smart, 2004). In southern France, we used time-averaged water data of [Ca$^{2+}$] and [Mg$^{2+}$] for springs and wells, provided from the national portal of water data (ADES) (Tab. S3). For the catchments in Israel, regional well and spring data were published by Ryb et al. (2014b), and in Crete, weathering rates were reported by Ott et al. (2019). Water chemistry data were used in conjunction with satellite-derived averages of precipitation and actual evapotranspiration (AET) to calculate carbonate weathering rates following the approach of Ott et al. (2019) (see supplement, Tab. S4).

**RATES OF CARBONATE DENUDATION**

Bedrock denudation rates and weathering rates calculated from water data are similar and generally fall between 20 - 40 mm/ka (mean: 29 and 32 mm/ka, respectively) (Fig. 2). The similarity of lowering rates for outcrops and weathering suggests that weathering is the dominant denudation process for exposed bedrock on hillslopes. Correlations between bedrock denudation rate and topographic and climatic variables are weak ($r^2 < 0.26$) (Fig. S2). While region-specific bedrock denudation rates show positive relationships with mean annual precipitation (MAP) (Fig. 2D), the strong covariation of topography and precipitation (Fig. S3) does not allow isolating climatic effects from these data. Site-specific variations in bedrock lithology, soil cover versus bare bedrock, and geomorphic sampling position likely contribute to the data scatter. Thus, our further analysis focuses primarily on the alluvial samples.
Catchment-average denudation rates from alluvial samples are consistently higher (mean 81 mm/ka; 2.7 times higher) than bedrock denudation and weathering rates (Fig. 2). Discrepancies between bedrock and alluvial denudation rates have been interpreted as reflecting growing topographic relief (Small and Anderson, 1998; Hancock and Kirwan, 2007; Thomas et al., 2017), or higher weathering rates below soil-covered bedrock and therefore a sampling bias toward fresh outcrop surfaces (Portenga and Bierman, 2011). However, it is unlikely that all studies comparing bedrock-interfluve samples and catchment averages would find increasing relief. If the rate discrepancy is due to soil cover, measured soil production rates should equal the catchment-average rates. Yet, a global compilation of soil production rates shows that most measurements are significantly lower than the catchment-average denudation rates (Heimsath et
al., 2012). An alternative explanation is that the bedrock denudation and soil production rates, whether in carbonates or siliciclastic rocks, represent a biased sampling of stable portions of the landscape (e.g., areas not affected by recent mass-wasting). Catchment-average sampling incorporates all portions of the upstream landscape and reflects a mix between the locally high denudation rate associated with mass wasting and a “background” rate set by bedrock denudation and soil production.

*Fig. 3: Correlations between catchment average $^{36}$Cl denudation rates, carbonate weathering rates, and topographic and climatic metrics. Inset-C: Correlation between MAP and elevation illustrating orographic precipitation in the analyzed areas (Israel, grey; France and Crete, red).*
MECHANICAL DENUDATION IN CARBONATE LANDSCAPES

There are three explanations for the two-to three-fold difference between catchment-scale weathering rates and basin-average denudation rates: (1) Catchment-average rates overestimate denudation due to low CRN concentration material from deep (> 5 m, Yanites et al., 2009) landslides; (2) higher effective precipitation earlier in the averaging time window of CRNs (Ryb et al., 2014b, 2015); (3) mechanical denudation of carbonates. The deep-seated landslide bias seems unlikely due to the consistent discrepancy between catchment average denudation and weathering in a diverse array of topography, from low relief and slope areas in Israel to the rugged mountainous terrain of Crete (Fig. 1). Furthermore, most sampled catchments drain moderately sized areas (10’s of km²) and are therefore large enough to buffer potential biases arising from mass-wasting (Niemi et al., 2005; Yanites et al., 2009).

The disparate integration timescales of CRNs (10²– 10⁵ yrs) and weathering rate measurements (0 – 10² yrs) (Plummer et al., 2001) lead Ryb et al. (2014b; 2015) to suggest that higher CRN denudation rates integrated a history of more vigorous weathering associated with past wetter climates. Most catchment-average rates integrate from modern to mid-Holocene, with the lowest rates integrating until the last glacial maximum (LGM). To test this hypothesis, we compared modern precipitation to 1 km resolution WorldClim paleo-precipitation maps from three different climate models (Fick and Hijmans, 2017). We found that mid-Holocene precipitation in all three Mediterranean sampling areas is predicted to have been between 2 and 27% higher in the mid-Holocene and between 25% drier or 55% wetter for the LGM depending on the area and preferred model (Tab. S4). Since weathering scales linearly with the water availability (White, 1984), these changes in precipitation, even with a total drawdown of AET,
cannot explain the data discrepancy through an increased paleo-water flux when integrated over time.

Catchment-average denudation rates show strong-to-moderate correlations with topographic metrics such as local relief and slope (Fig. 3A, B). Catchment-average rates also scale strongly with precipitation, however, all study sites exhibit orographic precipitation such that precipitation is correlated with topography (Figs. 3C, S3). When comparing denudation rates to runoff, the correlation is weak, and slopes of the regression lines for catchment average denudation and weathering rate are similar (Fig. 3D). Weathering rates correlate weakly-to-moderately with topographic and climatic variables but remain consistently lower than catchment-average denudation rates across all variable gradients. Collectively, these observations suggest that in Mediterranean carbonate landscapes, mechanical erosion contributes significantly to denudation because (1) catchment average denudation rates are substantially higher than weathering rates, (2) denudation scales with topographic steepness, (3) the difference between denudation and weathering, which we interpret as erosion, also increases with increasing topographic steepness, whereas (4) the increase of denudation with runoff can be fully accounted for by an increase in weathering (Fig. 3D).

LANDSCAPE EVOLUTION IN CARBONATES COMPARED TO OTHER ROCK TYPES

The trend of higher carbonate catchment-average rates compared to bedrock lowering rates is consistent with $^{10}$Be measurements from siliciclastic units where basin denudation rates are ~15 times higher than average outcrop lowering rates (Portenga and Bierman, 2011). The smaller ~2.7-fold discrepancy between local bedrock and catchment-average rates in carbonate terrains is best explained by the elevated role of chemical weathering in carbonates (~ 1/3 of
denudation, Fig. 3) compared to silica-rich rocks (< 5% of denudation) (Larsen et al., 2014). These findings have important implications for landscape evolution in carbonates and their topographic response to tectonic uplift and climate because mechanical erosion is slope-dependent, whereas weathering mostly depends on climate and vegetation.

**Fig. 4:** Mean weathering versus mean denudation rates for the sites with alluvial denudation data. We added Ireland as an example of a region dominated by chemical weathering processes (Simms, 2004) with rates by Drew (2001). Chemical weathering dominated landscapes will be subdued (white dot, southern Ireland); high erosion rates will lead to high relief areas dominated by slope-dependent mechanical processes (yellow dot, Crete).

Averaging catchment denudation and weathering rates for southern France, Israel (Soreq), and Crete, we find erosion-to-weathering ratios of 1.0, 2.2, and 2.5, respectively (Fig. 4). The substantial amount of mechanical erosion in these carbonate landscapes requires steep, high relief topography allowing slope-dependent processes to thrive. Carbonates in slowly
uplifting areas will denude sluggishly, and under favorable climatic conditions, weathering alone is sufficient to balance rock uplift with negligible mechanical erosion. In such a case, a carbonate landscape will remain subdued, and less-soluble lithologies will stick out because surface-lowering can only be achieved through erosion requiring local slopes to form. This can be observed in Ireland, where carbonates denude mostly through dissolution, and eroding sandstone ridges form the topographic highs (Simms, 2004).

The weathering rate will mostly be set by local climate but may be subject to a dissolution speed limit because (1) of a chemical threshold of <200 mg/l water hardness, which has been observed globally (Covington et al., 2015; Gaillardet et al., 2018), (2) an increase in precipitation will typically be partially compensated by an increase in AET, and (3) tropical regions with high runoff have lower water hardness due to a decrease in carbonate solubility with temperature. A global compilation of carbonate weathering rates finds a maximum rate of \( \sim 140 \text{ mm/ka} \) (Gaillardet et al., 2018), similar to our highest rate of 115 mm/ka. These rates may represent a carbonate weathering speed limit beyond which mechanical erosion becomes dominant, and topography steepens (Fig. 4).

In carbonate regions where mechanical erosion prevails, surface water infiltration will lower the discharge of surface streams. Reduction in stream discharge will decrease the erosional efficiency and cause steepening of the landscape compared to regions with less-soluble bedrock (Ott et al., 2019). This explains why carbonate terrain in locations with arid to semi-arid climate and significant uplift rates form steeper topography than areas underlain by silicate-rich lithologies (Ott et al., 2019; Ott, 2020), and carbonates form the low parts of the landscape, e.g. in Ireland or the Appalachians, where uplift rates are low, and the climate is favorable for weathering. Hence, the topography of carbonate regions is more sensitive to the interplay of
tectonics and climate compared to silicate-rich rocks. Studies investigating the climatic and
tectonic effects on topography typically report that tectonics govern the first-order landscape
morphology (Seybold et al., 2021). In contrast, we argue that carbonate regions offer the
potential to observe strong controls of climate and highly non-linear responses to tectonics on
landscape evolution.

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Supplemental material - Denudation partitioning in carbonate regions reveal climatic and tectonic drivers of carbonate landscape evolution

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This supplement includes a description of the carbonate dissolution rate calculation, the estimation of actual evapotranspiration, ³⁶Cl weathering bias correction, and the calculation of topographic metrics. Moreover, it includes the ³⁶Cl denudation rate compilation and water chemistry data used for southern France. Supplementary figures show correlations between bedrock lowering and topographic and climatic metrics. We also show the estimated recharge areas used for carbonate dissolution rate calculation and a p-value matrix for the significance of our correlations.

Carbonate dissolution rate calculation

All chemical data were corrected for precipitation input by assuming that all [Cl⁻] is derived from precipitation, and that other cations are scaled by seawater ratios (Stallard and
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Edmond, 1981). Water data from Israel were not corrected due to unknown [Cl\textsuperscript{-}]; however, the correction for [Ca\textsuperscript{2+}] was usually < 1% and is therefore assumed to be negligible.

To calculate the water flux, we used satellite-derived precipitation and actual evapotranspiration data (AET). Precipitation was derived from the WorldClim 1 km dataset (Fick and Hijmans, 2017) and averaged for each catchment. To determine the runoff available for dissolution, AET was estimated from a parameterization of vegetation indices (PaVI-E) model (Helman et al., 2015) using MODeerate resolution Imaging Spectroradiometer (MODIS) satellite data from 2000 to 2016 at 1 km resolution and water vapor flux data from the eddy covariance tower international net (FLUXNET) (see description below for details). The dissolution rate is then calculated by assuming that all [Ca\textsuperscript{2+}] and [Mg\textsuperscript{2+}] in the water are derived from carbonate dissolution and using the water flux from our local runoff calculation for each catchment. A recharge area for runoff averaging was estimated for groundwater water samples based on surface topography and local geology (see Fig. S1 for estimated groundwater recharge areas).

Dissolution rates were calculated with an uncertainty of 10% on the precipitation data and 20% on the actual evapotranspiration. Because recharge areas of springs and wells in karstic terrains can deviate significantly from topographically estimated areas (Fig. S1), we show a second estimate based on the P and AET average of the entire mountain range where the spring or well is located (Tab. S3). However, the difference in the average dissolution rate between both approaches is < 3%, and we, therefore, choose to use the topographic recharge area estimates outlined in Fig. S1.

**Estimation of actual evapotranspiration**

Actual evapotranspiration (AET) was calculated on a yearly basis at a 1 km spatial resolution using an empirical model (PaVI-E) based on relationships found between satellite-
derived vegetation indices and eddy covariance fluxes (Helman et al., 2015). The advantage of PaVI-E is its independence of meteorological data, which avoids parameter dependency issues in climate-related studies.

PaVI-E was successfully validated against basin-scale AET derived from water balance calculations (i.e., $\text{AET} = \text{P} - \text{Q}$) in the Eastern Mediterranean region ($R^2 = 0.85, p<0.05$) and was shown to be comparable with other well-established physically-based AET models (Helman et al., 2015, 2017a). It has been used to study climate change impacts on the terrestrial water cycle and is considered a reliable tool for water balance calculations (Helman et al., 2017c, 2017b).

**Topographic metric calculation**

Topographic metrics were measured on a 1 arc-second (~30 m) Shuttle Radar Topography Mission dataset. Slope, mean elevation, mean slope, and mean local relief (500 m radius) were calculated using TopoToolbox (Schwanghart and Scherler, 2014).

**Weathering corrections for denudation rates**

Chemical weathering can bias cosmogenic nuclide-derived denudation rates. Regolith weathering can overestimate $^{36}\text{Cl}$ denudation rates because the soluble target mineral calcite may have a shorter regolith residence time than the bulk rock due to its high solubility. We use the methods proposed by Ott et al. (2022) to correct all alluvial denudation rates for regolith weathering. The correction requires knowledge of the regolith or bedrock composition. In the absence of direct bedrock compositional data, we use the reported bulk chemical composition of the samples as an estimate of regolith composition. We multiply the weight-percent CaO by 1.4 to estimate the calcite fraction in the regolith, take the SiO$_2$ weight-percent as fraction quartz, and assign the remainder as other insoluble minerals.

Additionally, we use the mean weathering rates for every area (Israel: $21 \pm 3$ mm/ka, Crete: $48 \pm 11$ mm/ka, France: $37 \pm 8$ mm/ka) for the corrections. On Crete, we estimate 30 cm
regolith thickness within the sampled catchments from the European soil database (ESDAC) (Panagos et al., 2012). We use this thickness in conjunction with a density of 1.5 g/cm³ to derive a soil mass of 45 g/cm². The same value is found in southern France, with soil depth estimated from the maps of Chen et al. (2019). For the Soreq catchment in Israel, Ryb et al. (2014) report regolith thicknesses between 0 and 75 cm. We take the middle (37.5 cm) and use it with the reported regolith density of 1.4 g/cm³ to derive an average soil mass of 52.5 g/cm².

Weathering-corrected denudation rates are similar to conventional denudation rates, with a maximum difference of about 7%. The sample compositions reported from southern France indicate almost pure limestone. Therefore, the bias for these rocks is very small. The WeCode package by Ott (2022) used for weathering corrections does not account for radioactive decay. This explains why the weathering-corrected denudation rates from southern France are marginally higher than the conventional rates, including decay. For five samples, the weathering correction could not be applied because the reported nuclide concentration was greater than the theoretical maximum nuclide concentration for the provided parameter combination (N_{max}, for definition, see Ott et al., 2022). This is likely linked to uncertainties in the input parameters, such as weathering rate, regolith mass, etc. Likely, for some samples considered here, the actual catchment weathering rate is lower than the mean weathering rate for the area. However, the distribution of water sampling stations does not allow deriving weathering rates close to every location sampled for cosmogenic nuclides. In general, the weathering bias in the investigated samples is low due to the low weathering rates and clean limestone composition of many samples. Because the corrections are low and uncertainty exists in some of the correction parameters, we chose to report uncorrected denudation rates in the main text.
Tab. S2: Output of weathering correction calculation for denudation rates.

<table>
<thead>
<tr>
<th>Location</th>
<th>Sample</th>
<th>Denudation rate mm/ka</th>
<th>err</th>
<th>Conventional denudation rate mm/ka</th>
<th>Rate difference %</th>
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### Tab S3: Raw and precipitation corrected water data from southern France. \([\text{Ca}^{2+}]\) and \([\text{Mg}^{2+}]\). \(n\) – number of samples, \(\text{min}\) – minimum concentration, \(\text{max}\) – maximum concentration measured.

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Tab. S4: Dissolution rates calculated using estimated recharge areas from topography (Fig. S1) together with average $P$ and AET values for the entire mountain range of the sampled spring, well or river.

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Tab. S5: Comparison of estimated annual paleo-precipitation rates from climate models for all areas with published catchment average denudation rates. For three different WorldClim paleo-climate models (Fick and Hijmans, 2017), paleo-precipitation values were extracted for all sampled $^{36}Cl$ sampling locations, averaged for each region and compared to modern precipitation rates. LGM – Last Glacial Maximum

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Fig. S1: Hypothetical recharge areas used to calculate the water flux required for the carbonate dissolution rate calculation for wells and springs. (a) and (b) show recharge areas for southern France, (c) for Israel, where the Western Mountain Aquifer provides the water for most wells in the coastal plain (Sheffer et al., 2010).
Fig. S2: Correlation between $^{36}$Cl bedrock denudation rates, topographic metrics and mean annual precipitation. Colors indicate different study areas.
Fig. S3: (a) Correlation coefficient matrix for the data set and seven topographic and climatic metrics. Red colors indicate that most metrics are positively correlated for carbonate catchments. (b) P-value matrix for the correlation coefficient matrix in (a). The cells are colored by different significance levels.
References


7 Ott, R.F., 2022, WeCode - Weathering Corrections for denudation rates V. 1.0: GFZ Data


