

## ISOTOPIC SIGNATURES OF CARBON, OXYGEN AND STRONTIUM OF HISTORICAL MORTAR AND PLASTER IN STYRIA

Barbara KOSEDNAR<sup>1</sup>, Martin DIETZEL<sup>1</sup>, Albrecht LEIS<sup>2</sup>, Bettina WIEGAND<sup>3</sup>,  
Bernhard SCHRETTLE<sup>1</sup>, Karl STINGL<sup>1</sup>, Miriam BAUMGARTNER<sup>1</sup> & Ralf BENISCHKE<sup>2</sup>

<sup>1</sup> Institute of Applied Geosciences, Graz University of Technology, Rechbauerstrasse 12, A-8010 Graz. e-mail: kosednar@egam.tu-graz.ac.at

<sup>2</sup> Institute of Water Resources Management, Hydrogeology and Geophysics, Joanneum Research Graz, Elisabethstraße 16 / II, A-8010 Graz.

<sup>3</sup> Department of Geological and Environmental Sciences, Stanford University, CA 94305-2115, USA

Historical buildings are constructed of geo-materials, mortar, and plaster of various compositions. Mortar and plaster are man-made materials. Thus, the chemical and isotopic composition comprises information about the historical environment with respect to the provenance of the materials, processing, and specific applications. Moreover, isotopic data may provide additional information about the ancient composition of carbon dioxide and water. The present study is focused on the mineralogical, chemical and isotopic composition of dated and well-characterized carbonate mortar and plaster of roman, medieval, and early modern (pre-industrial) times in Styria (Austria).

Mortars and plasters were sampled from historical buildings in the area of Flavia Solva, Frauenberg, Deutschlandsberg, Seggauberg, Kleinstübing, Niederhofen, Södingberg, and Graz. Sampling was conducted from the exterior to interior mortar layer, wherever applicable. The sampled materials mostly consist of a CaCO<sub>3</sub> (calcite) cement with aggregates of quartz and additional silicates like clay minerals. The analyzed Sr/Ca and <sup>87</sup>Sr/<sup>86</sup>Sr ratios of the cement are between 0.00030 and 0.0023, and 0.7093 and 0.7104, respectively. These ratios reflect the composition of the natural deposits used for manufacturing of lime mortar. The respective values depend on the environment of formation and on the mineralogical composition (e.g. calcite or aragonite) of the primary limestone.

However, the distribution of <sup>13</sup>C/<sup>12</sup>C and <sup>18</sup>O/<sup>16</sup>O ratios in the carbonate mortars and plasters indicates a more complicated situation as isotopic compositions comprise a wide range of δ<sup>13</sup>C<sub>CaCO<sub>3</sub></sub>(PDB) from -24.2 to -0.8, and of δ<sup>18</sup>O<sub>CaCO<sub>3</sub></sub>(PDB) from -23.9 to -2.6 ‰. The stable carbon and oxygen isotope distributions in the carbonate cement displays an almost linear correlation. In general calcite is continuously isotopically "heavier" from the exterior to the interior mortar layer. The range and systematic correlation of the data reflect isotopic fractionation effects upon setting of the cement and during the history of the individual cement.

In principle isotope distributions depend on the composition of the gaseous CO<sub>2</sub> and aqueous OH<sup>-</sup> according to the overall reaction



during the formation of carbonate cement. Reaction 1 is accompanied by a kinetic isotope fractionation due to the hydroxylation of gaseous CO<sub>2</sub> (Dietzel, 2000), resulting in an enrichment of <sup>12</sup>C versus <sup>13</sup>C in the precipitated CaCO<sub>3</sub>. If gaseous CO<sub>2</sub> is delivered from the present Earth's atmosphere (δ<sup>13</sup>C<sub>CO<sub>2</sub>(atm)}</sub> = -7 ‰) a δ<sup>13</sup>C<sub>CaCO<sub>3</sub></sub> value of about -25 ‰ is obtained. Evolution of oxygen isotopes is more complex and yield δ<sup>18</sup>O<sub>CaCO<sub>3</sub></sub> values of about -20 ‰ for calcite precipitated according to reaction 1 (Dietzel et al., 1992). Upon setting of the cement, the diffusion of gaseous CO<sub>2</sub> and subsequent reaction to CaCO<sub>3</sub> leads to a continuous enrichment of <sup>13</sup>C and <sup>18</sup>O (versus <sup>12</sup>C and <sup>16</sup>O, respectively) of CO<sub>2</sub> within the gas

phase along the cement setting path. Accordingly, precipitated calcite is isotopically “lighter” at the exterior mortar layer.

The results show that analyses of carbon and oxygen isotopic compositions permit to follow the historic cementation process, and to detect potential variations of the composition of the atmospheric CO<sub>2</sub> and liquid (H<sub>2</sub>O). Variations may be caused by natural or anthropogenic impacts, e.g. evaporation of H<sub>2</sub>O and burning of coal, respectively. From another point of view, secondary processes like interaction with isotopically “light” soil-CO<sub>2</sub> or re-crystallization of carbonate cements in the presence of H<sub>2</sub>O from various origins may be deciphered.

## References

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Autor(en)/Author(s): Schrettle Bernhard, Dietzel Martin, Leis Albrecht, Wiegand Bettina, Schrettle Bernhard, Stingl Karl, Baumgartner Miriam, Benischke Ralf

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