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The origin of sulphur in gypsum and dissolved sulphate in the Central Namib Desert, Namibia

F.D. Eckardt [a,*], B. Spiro [b]

[a] *School of Geography, Mansfield Road, Oxford OX1 3TB, UK*
[b] *Natural Environment Research Council, Isotope Geosciences Laboratory, Keyworth, Nottingham NG12 5GG, UK*

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Abstract

This study investigates the sulphur source of gypsum sulphate and dissolved groundwater sulphate in the Central Namib Desert, home to one of Africa's most extensive gypsum ($CaSO_4 \cdot 2H_2O$) accumulations. It investigates previously suggested sulphate precursors such as bedrock sulphides and decompositional marine biogenic H_2S and studies the importance of other potential sources in order to determine the origin of gypsum and dissolved sulphate in the region. An attempt has been made to sample all possible sulphur sources, pathways and types of gypsum accumulations in the Central Namib Desert. We have subjected those samples to sulphur isotopic analyses and have compiled existing results. In addition, ionic ratios of Cl/SO_4 are used to determine the presence of non-sea-salt (NSS) sulphur in groundwater and to investigate processes affecting groundwater sulphate. In contrast to previous work, this study proposes that the sulphur cycle, and the formation of gypsum, in the Namib Desert appears to be dominated by the deposition of atmospheric sulphates of phytoplanktonic origin, part of the primary marine production of the Benguela upwelling cells. The aerosol sulphates are subjected to terrestrial storage within the gypsum deposits on the hyper-arid gravel plain and are traceable in groundwater including coastal sabkhas. The hypothesis of decompositional marine biogenic H_2S or bedrock sulphide sources, as considered previously for the Namib Desert, cannot account for the widespread accumulation of gypsum in the region. The study area in the Central Namib Desert, between the Kuiseb and Omaruru rivers, features extensive gypsum accumulations in a ca. 50–70 km wide band, parallel to the shore. They consist of surficial or shallow pedogenic gypsum crusts in the desert pavement, hydromorphic playa or sabkha gypsum, as thin isolated pockets on bedrock ridges and as discrete masses of gypsum selenite along some faults. The sulphur isotopic values ($\delta^{34}S$ ‰CDT) of these occurrences are between $\delta^{34}S$ +13.0 and +18.8‰, with lower values in proximity to sulphuric ore bodies ($\delta^{34}S$ +3.1 and +3.4‰). Damaran bedrock sulphides have a wide range from $\delta^{34}S$ −4.1 to +13.8‰ but seem to be significant sources on a local scale at the most. Dissolved sulphate at playas, sabkhas, springs, boreholes and ephemeral rivers have an overall range between $\delta^{34}S$ +9.8 and +20.8‰. However, they do not show a systematic geographical trend. The Kalahari waters have lower values, between $\delta^{34}S$ +5.9 and +12.3‰. Authigenic gypsum from submarine sediments in the upwelling zone of the Benguela Current between Oranjemund and Walvis Bay ranges between $\delta^{34}S$ −34.6 to −4.6‰. A single dry atmospheric deposition sample produced a value of $\delta^{34}S$ +15.9‰. These sulphur isotopic results, complemented by meteorological, hydrological and geological information, suggest that sulphate in the Namib Desert is mainly derived from NSS sulphur, in particular oxidation products of marine dimethyl sulphide CH_3SCH_3 (DMS). The hyper-arid conditions prevailing along the Namibian coast since Miocene times favour the overall preservation of the sulphate minerals. However, sporadic and relatively wetter

* Corresponding author. Tel.: +44 (1865) 271919; Fax: +44 (1865) 271929; E-mail: frank.eckardt@geography.oxford.ac.uk

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periods have promoted gypsum formation: the segregation of sulphates from the more soluble halite, and the gradual seaward redistribution of sulphate. This study suggests that the extreme productivity of the Benguela Current contributes towards the sulphur budget in the adjacent Namib Desert.

Keywords: Benguela Current; groundwater; gypsum; Namib Desert; sulphur; sulphur isotopes

1. Introduction

1.1. Origin of desert gypsum

Gypsum ($CaSO_4 \cdot 2H_2O$) is one of the most common evaporitic minerals occurring in terrestrial, lacustrine and marine sedimentary environments (e.g. Watson, 1979). Its occurrence in surficial and shallow pedogenic settings is restricted to arid environments, where evaporation exceeds precipitation. Desert gypsum can be found in either hydromorphic settings associated with salt-pans (playas) or coastal mudflats (sabkhas) and may accumulate in soil and sediments either through atmospheric deposition or groundwater evaporation (Watson, 1979). Examples of desert gypsum deposits are numerous in the literature with the primary sources being bedrock minerals (Visser, 1963; Yonge and Krouse, 1987; Bormann and Diedrich, 1995) and a mixture of marine sources such as sea spray and atmospheric non-sea-salt (NSS) sulphate such as dimethyl sulphide (CH_3SCH_3) (DMS) (Reimer, 1986; Chivas et al., 1991). In general, the formation of desert gypsum is suggested to be closely linked to the regional nature of the biogeochemical sulphur cycle (Charlson et al., 1985). The origin of gypsum sulphur has been summarised in the study of Australian playas by Chivas et al. (1991) and is equally valid for dissolved sulphate in surface and ground water: (1) atmospherically transported marine aerosols; (2) dissolution or deflation of evaporites in sediments within the drainage basin; (3) marine salt trapped in marine sedimentary rocks; (4) 'relict' sea-salts left by the regression of the ocean; (5) evaporated river water; (6) weathering of igneous or metamorphic basement rock in the basin.

1.2. Occurrence of gypsum in Namibia

Gypsum can be found along most of the hyper-arid coastline from South Africa through to Angola and is thus present in the entire Namib Desert (J.D. Ward, pers. commun., 1994) (Fig. 1). Miocene terraces of the lower Orange River have been reported to be capped by up to 4 m of pure gypsum extending inland to about 100 km (Pellet, 1995), while no gypsum has been reported from within the Namib Sand Sea. The area controlled by Consolidated Diamond Mines between Oranjemund and Lüderitz, is reported to hold some gypsum (J.D. Ward, pers. commun., 1994). The northern Namib Desert does hold gypsum, but too few studies have been concerned with this area to appreciate fully its extent. Minor sulphate accumulations outside the Namib Desert have been reported in and around the Etosha pan and in salt-pans of the Kalahari Desert (Schneider and Genis, 1992).

However, the general consensus is that the bulk of the accumulations are found in the Central Namib Desert underneath the dune-free gravel pavement (Watson, 1985). This relatively stable gravel plain is extremely flat and has a 1% gradient. It is subject to rare floods and easterly Bergwinds, which produce windstreaks, ventifacts and an ephemeral drainage network. Gypsum is largely of the pedogenic variety, forming a shallow but variable subsurface feature of low purity with preferential near-surface concentration in the thick Tertiary gravel cover. Gypsum occurs in the coastal half of the 30,000 km^2 of the Central Namib Desert. Its eastern limit is at about 50–70 km from the shore at an elevation of 400–500 m. The gypsum accumulations in this region are impressive but provide limited vertical access and have limited surface exposure. Judging by the 12 m high Tertiary terrace exposure overlooking the Swakop River at Goanikontes, the bulk of the gypsum is stored in the upper horizons of the colluvium, cementing angular clasts and forming mesocrystalline and alabastrine gypsum. Lower horizons show pronounced accumulations of more soluble halite and a distinct absence of gypsum (Gevers and Van der Westhuyzen, 1931) (Fig. 2). A regional survey on

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5. Summary

We propose that there is no isotopic association between Benguela H_2S and the terrestrial gypsum accumulations as suggested by Martin (1963). Hyper-productive coastal upwelling conditions alongside a hyper-arid desert dominates the hydrogeochemical sulphur cycle of the Namib Desert/Atlantic interface and can account for the observed isotopic composition of gypsum and groundwater sulphate (Fig. 7). For a more detailed description of the atmospheric sulphur cycle in the marine boundary layer, refer to Capaldo and Pandis (1997). Sulphur for the extensive Middle Miocene to recent gypsum deposits of the Central Namib Desert can be potentially derived from a single NSS sulphur source such as DMS. The present study shows that weathering of igneous and metamorphic basement rocks and associated sulphide mineralisation has only a local influence. Surprisingly, marine water sulphate does not seem to be a major contributor to the Namibian sulphates. This suggests that marine incursions, marine evaporites in sediments or sea spray are unlikely to be the primary cause in the formation of desert gypsum. Hydrogen sulphide is a major constituent in the shelf sediments but could not be traced in the terrestrial or marine water environment. Catastrophic H_2S gas release events are probably exceptional. Although memorable, they seem quantitatively insignificant. Atmospherically transported marine aerosols derived from biogenic sulphur compounds (DMS) appear to be the main sulphur source. The exact isotopic composition, dynamics and deposition rates are as yet unknown. The present study suggests a maximal value for this source of +21%. The major terrestrial process is the recycling of the pedogenic and groundwater sulphate through dissolution and reprecipitation in the sparse aqueous environments and the aeolian transport of surficial gypsum. It is large-scale terrestrial and atmospheric mixing that is responsible for the relatively homogeneous $\delta^{34}S$ of the gypsum crusts throughout the Central Namib Desert.

6. Note added in proof

It has recently been possible to obtain sufficient amounts of sulphate from fog at Gobabeb (11/9/97) by using a clean 1 m^2 polypropylene mesh instead of the less efficient collectors used in Eckardt and Schemenauer (1998), which has however a slightly higher risk of contamination. The $\delta^{34}S$ value of +14.7‰ suggests a NSS sulphur source for fog depositions in the Central Namib Desert, as already postulated for the dry aerosol deposition (+15.9 $\delta^{34}S$ ‰).

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