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Atmospheric sulphate increase in the past 200 years recorded in stalagmites from Italy and Oman

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Annually-resolved time-series of sulphur were generated by applying synchrotron radiation micro X-Ray Fluorescence (XRF) for two speleothems from Grotta di Ernesto (Dolomites) and Grotta Savi (Trieste) in northern Italy. X-ray absorption-edge spectrometry proved that S is in the form of sulphate and XRF mapping demonstrated that sulphate is incorporated into calcite growth layers. The stalagmites from Ernesto (ER78) and Savi (SV1) show a substantial rise in sulphate starting at c. 1880, which largely reflects an anthropogenically-induced increase of sulphate in the atmosphere, moderated by some ecosystem storage. Synchrotron micro-XRF measurements do not give absolute concentrations. Quantitative calibration of the S content in ER78 was carried out using high-mass resolution inductively-coupled mass spectrometer (ICP-MS), and shows an increase in S content from 15 to 66 ppm. Interestingly, the post-1980 decline in sulphur observed in Alpine ice cores is not apparent in the Alpine ER78 specimen (Frisia et al., 2005). In an annually laminated speleothem from southern Oman (Fleitmann et al., 2004), S was measured using the Stanford/USGS Sensitive High-Resolution Ion MicroProbe Reverse Geometry (SHRIMP RG) and a Cameca Ultrachron electron microprobe. The chronology of the S time series is based on both annual layer counts and supported U/Th ages. High annual growth rates ($\sim$0.3 mm/yr) and close sampling intervals (0.03 mm) make monthly resolution possible. The sulphur time-series from Southern Oman show high amplitude variations with

distinct sulphur peaks; some of which seem to coincide with large volcanic eruptions (e.g. Krakatau at 1883). A substantial increase in S since the late 1950s mimics the increase in sulphur emissions on the Arabian Peninsula and the Indian subcontinent. Similarities of the trend, and lack in both the Alpine and Middle eastern records of a decline in sulphur content in the past 20 years may be a significant indication that speleothems may record regional S emission that are still unaccounted for in the global anthropogenic sulphate emission estimates, and/or the effects of trajectories in the spatial distribution of anthropogenic sulphate aerosols. Our study nevertheless demonstrates that speleothems can provide detailed information on fluctuations in atmospheric sulphur.