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Gaseous (DMS, MSA, SO₂, H₂SO₄ and DMSO) and particulate (sulfate and methanesulfonate) sulfur species over the northeastern coast of Crete

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Abstract. A detailed study of the levels, the temporal and diurnal variability of the main compounds involved in the biogenic sulfur cycle was carried out in Crete (Eastern Mediterranean) during the Mediterranean Intensive Oxidant Study (MINOS) field experiment in July-August 2001. Intensive measurements of gaseous dimethylsulfide (DMS), dimethylsulfoxide (DMSO), sulfur dioxide (SO₂), sulfuric (H₂SO₄) and methanesulfonic acids (MSA) and particulate sulfate (SO₄²⁻) and methanesulfonate (MS⁻) have been performed during the campaign.

Dimethylsulfide (DMS) levels ranged from 2.9 to 136 pmol·mol⁻¹ (mean value of 21.7 pmol·mol⁻¹) and showed a clear diurnal variation with daytime maximum. During nighttime DMS levels fall close or below the detection limit of 2 pmol·mol⁻¹. Concurrent measurements of OH and NO₃ radicals during the campaign indicate that NO₃ levels can explain most of the observed diurnal variation of DMS. Dimethylsulfoxide (DMSO) ranged between 0.02 and 10.1 pmol·mol⁻¹ (mean value of 1.7 pmol·mol⁻¹) and presents a diurnal variation similar to that of DMS. SO₂ levels ranged from 220 to 2970 pmol·mol⁻¹ (mean value of 1030 pmol·mol⁻¹), while nss-SO₄²⁻ and MS⁻ ranged from 330 to 7100 pmol·mol⁻¹, (mean value of 1440 pmol·mol⁻¹) and 1.1 to 37.5 pmol·mol⁻¹ (mean value of 11.5 pmol·mol⁻¹) respectively.

Of particular interest are the measurements of gaseous MSA and H₂SO₄. MSA ranged from below the detection limit (3×10⁴) to 3.7×10⁷ molecules cm⁻³, whereas H₂SO₄ ranged between 1×10⁵ and 9.0×10⁷ molecules cm⁻³. The measured H₂SO₄ maxima are among the highest reported in literature and can be attributed to high insolation, absence of precipitation and increased SO₂ levels in the area. From the concurrent SO₂, OH, and H₂SO₄ measurements a sticking coefficient of 0.52±0.28 was calculated for H₂SO₄. From the concurrent MSA, OH, and DMS measurements the yield of gaseous MSA from the OH-initiated oxidation of DMS was calculated to range between 0.1-0.4%. This low MSA yield implies that gaseous MSA

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levels can not account for the observed MS^- levels. Heterogeneous reactions of DMSO on aerosols should be considered to explain the observed levels of MS^- .

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