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Gaseous (DMS, MSA, SO_2, H_2SO_4 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 (SO_2), sulfuric (H_2SO_4) and methanesulfonic acids (MSA) and particulate sulfate (SO_4^{2-}) 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 NO_3 radicals during the campaign indicate that NO_3 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. SO_2 levels ranged from 220 to 2970 pmol·mol^{-1} (mean value of 1030 pmol·mol^{-1}), while nss-SO_4^{2-} 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 H_2SO_4. MSA ranged from below the detection limit ($3x10^4$) to $3.7x10^7$ molecules cm^{-3}, whereas H_2SO_4 ranged between $1x10^5$ and $9.0x10^7$ molecules cm^{-3}. The measured H_2SO_4 maxima are among the highest reported in literature and can be attributed to high insolation, absence of precipitation and increased SO_2 levels in the area. From the concurrent SO_2, OH, and H_2SO_4 measurements a sticking coefficient of 0.52±0.28 was calculated for H_2SO_4. 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

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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