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Formaldehyde over the eastern Mediterranean during MINOS: Comparison of airborne in-situ measurements with 3D-model results

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Abstract. Formaldehyde (HCHO) is an important intermediate product in the photochemical degradation of methane and non-methane volatile organic compounds. In August 2001, airborne formaldehyde measurements based on the Hantzsch reaction technique were performed during the Mediterranean INtensive Oxidant Study, MINOS. The detection limit of the instrument was 42 pptv (1σ) at a time resolution of 180 s (10-90%). The overall uncertainty of the HCHO measurements was 30% at a mixing ratio of 300 pptv. In the marine boundary layer over the eastern Mediterranean Sea average HCHO concentrations were of the order of 1500 pptv, in reasonable agreement with results from a three-dimensional global chemical transport model of the lower atmosphere including non-methane volatile organic compound (NMVOC) chemistry. Above the boundary layer HCHO mixing ratios decreased with increasing altitude to a minimum level of 250 pptv at about 7 km. At higher altitudes (above 7 km) HCHO levels showed a strong dependency on the airmass origin. In airmasses from the North Atlantic/North American area HCHO levels were of the order of 300 pptv, a factor of 6 higher than values predicted by the model. Even higher HCHO levels, increasing to values of the order of 600 pptv at 11 km altitude, were observed in easterlies transporting air affected by the Indian monsoon outflow towards the Mediterranean basin. Only a small part (~30 pptv) of the large discrepancy between the model results and the measurements of HCHO in the free troposphere could be explained by a strong underestimation of the upper tropospheric acetone concentration by up to a factor of ten by the 3D-model. Therefore, the measurement-model difference in the upper troposphere remains unresolved, while the observed dependency of HCHO on airmass origin might indicate that unknown, relatively long-lived NMVOCs - or their reaction intermediates - associated with biomass burning are at least partially responsible for the observed discrepancies.

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