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Measurement and modeling of methanol deposition/emission in a mixed forest.

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Methanol is one of Biogenic Volatile Organic Compounds (BVOC) showing the highest atmospheric concentration in the atmosphere. It influences OH concentration in the troposphere, tropospheric ozone production and formaldehyde production. In order to better understand the methanol emission/deposition mechanisms, micrometeorological flux measurements of methanol were carried out above a mixed forest (Fagus sylvatica, Pseudotsuga menziesii, Abies alba, Picea abies) at the Vielsalm experimental site (Belgium) from July to November 2009 and from April to November 2010. The flux measurements were obtained by the eddy-covariance technique using proton transfer reaction mass spectrometry (PTR-MS).

At Vielsalm, the methanol fluxes were found bi-directional, generally negative (deposition) during the night and positive during the day. During the summer-autumn period, the forest behaved mainly as a sink, night deposition being more important than the day emission, the inverse situation was observed during the spring period. The methanol deposition increased linearly with friction velocity and with atmospheric methanol concentration. The humidity seemed to be also an important driving variable, the depositions being more important when the atmospheric water vapor pressure was closed to saturation. This could suggest the presence of a deposition mechanism associated with the presence of water films on the vegetation surface in which methanol would dissolve. The formation of these films would indeed be favored by humid conditions.

In order to describe this mechanism, we developed an absorption/desorption model (MAD), based partially on the model developed by Sutton et al. (1998) to explore the ammonia adsorption/desorption effect. MAD took atmospheric methanol concentration, friction velocity, water vapor pressure deficit, precipitations and Henry's law constant temperature dependence into account. The model was calibrated on night deposition fluxes measured during the summer-autumn of 2009 and validated on 2010 data. Simulation reproduced well measurements during most of the measurement period, as well in summer-autumn 2009, as during summer 2010. This suggests that, most of the time, the measured fluxes (night and day) can be mainly explained by a physical absorption/desorption process. In contrast, during one 2010 spring week, significant differences appeared between simulations and measurements, suggesting the pre-eminence of biogenic fluxes at that time. They appeared during a period presenting a co-occurrence of sunny conditions and important leaf development. These biogenic emissions corresponded probably to the demethylation of pectin in the primary cell walls and were mainly controlled by the air temperature.

Using MAD, we will be able to disentangle emissions and depositions and propose a standard emission factor, to describe its seasonal evolution, to propose light and temperature dependencies for the emissions and to investigate more deeply the deposition process. These information are crucial for the improvement of the methanol exchange algorithms, depositions having been shown recently to be underestimated for oxygenated BVOCs.

Ref.: *M.A. Sutton, J.K. Burkhardt, D. Guerin, E. Nemitz and D. Fowler: Development of resistance models to describe measurements of bi-directional ammonia surface–atmosphere exchange, Atmospheric Environment, 32, 473-480, 1998.*