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Supplement of

Chemical and meteorological influences on the lifetime of NO_3 at a semi-rural mountain site during PARADE

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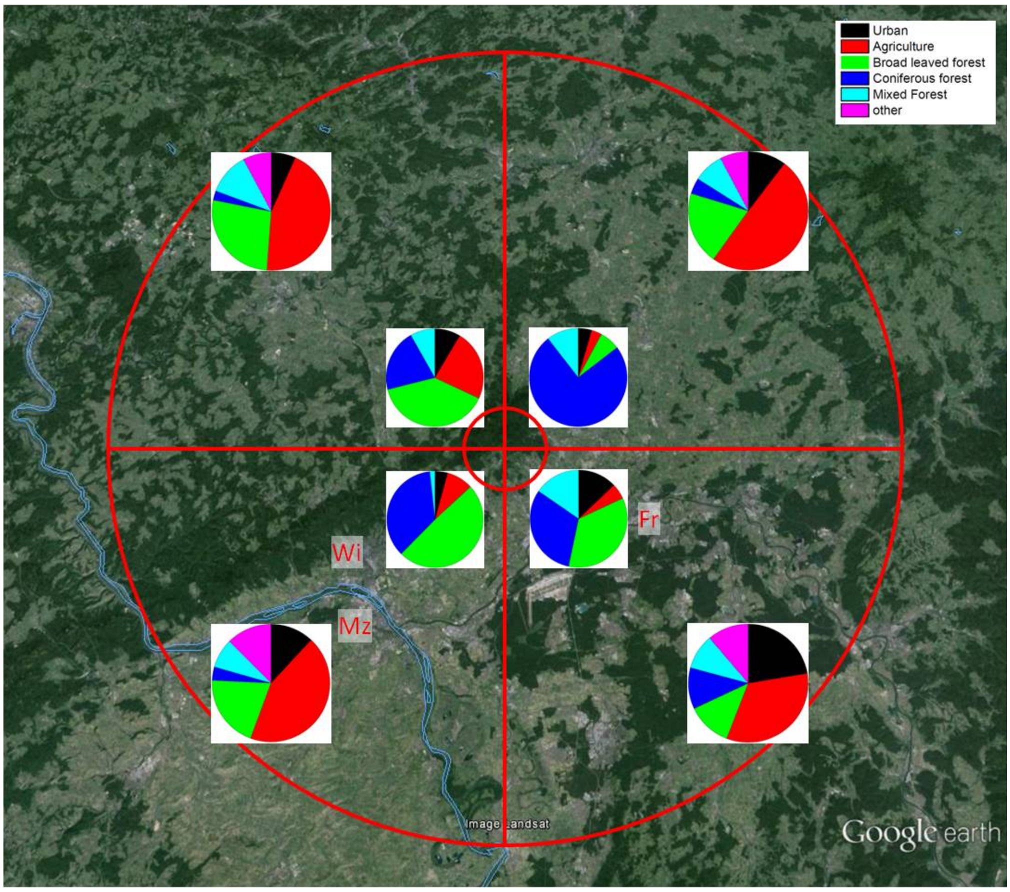

Fig. S1. Land-use in the area surrounding the PARADE campaign measurement site. The inner circle has a radius of 5 km, the outer circle a radius of 50 km. Both areas are divided into four sectors (North-East, South-East, South-West and North-West). For each of the 8 sectors the land-use is given by a pie chart. The Fr, Wi and Mz symbols represent the approximate location of the cities Frankfurt, Wiesbaden and Mainz, respectively. The map was generated from the CORINE Land Cover database by Dr. Pablo J. Hidalgo.

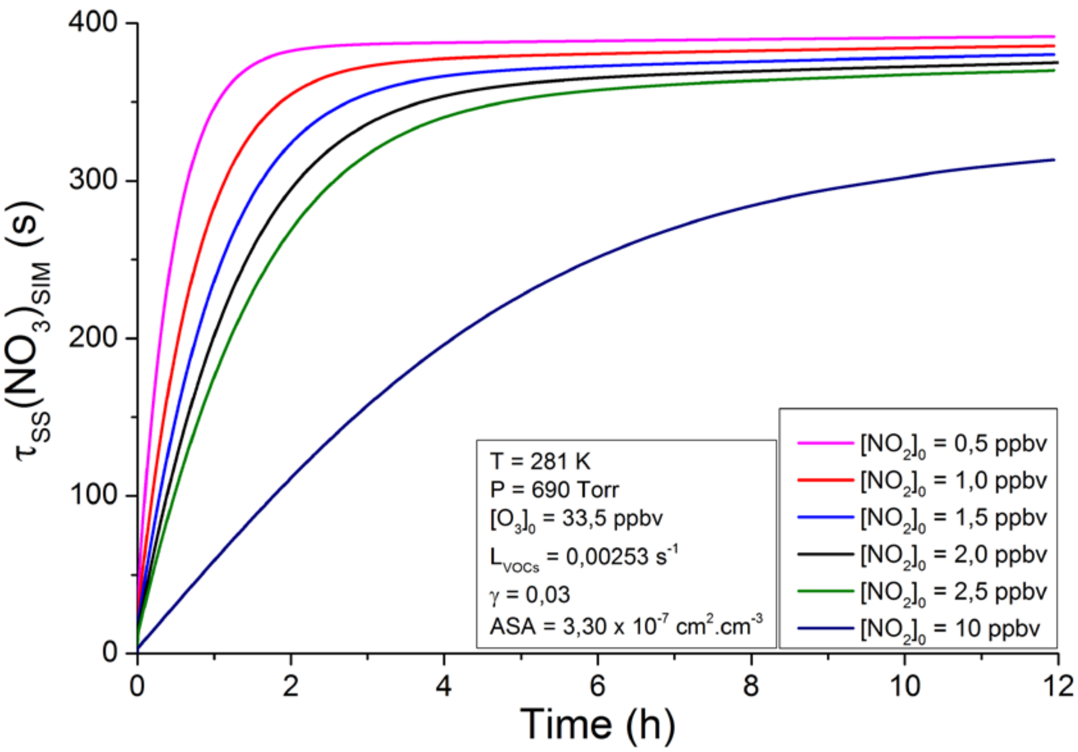

Fig. S2. Simulated evolution of the steady state lifetime of NO_3 assuming reaction of NO_2 with O_3 as the only NO_3 source, and both a homogeneous and a heterogeneous loss process. The simulation assumes variable $[O_3]$ and $[NO_2]$ as would be the case for a point source and a constant NO_3 reactivity in the gaseous phase (0.00253 s^{-1}, due e.g. to VOCs). The different curves each correspond to a different initial NO_2 mixing ratio (with a fixed initial $[O_3]$ for each simulation). The NO_3 losses through N_2O_5 uptake on aerosols is simulated by keeping the aerosol surface area (ASA) and the uptake coefficient γ constant at the values listed in the figure. The overall heterogeneous sink, depending on $[NO_2]$, is thus different for each simulation, which explains the slightly different effective lifetime ($\tau_{ss}(NO_3)$ for $t \rightarrow \infty$). At short times, the true steady state lifetime (350 to 380 s) is underestimated.

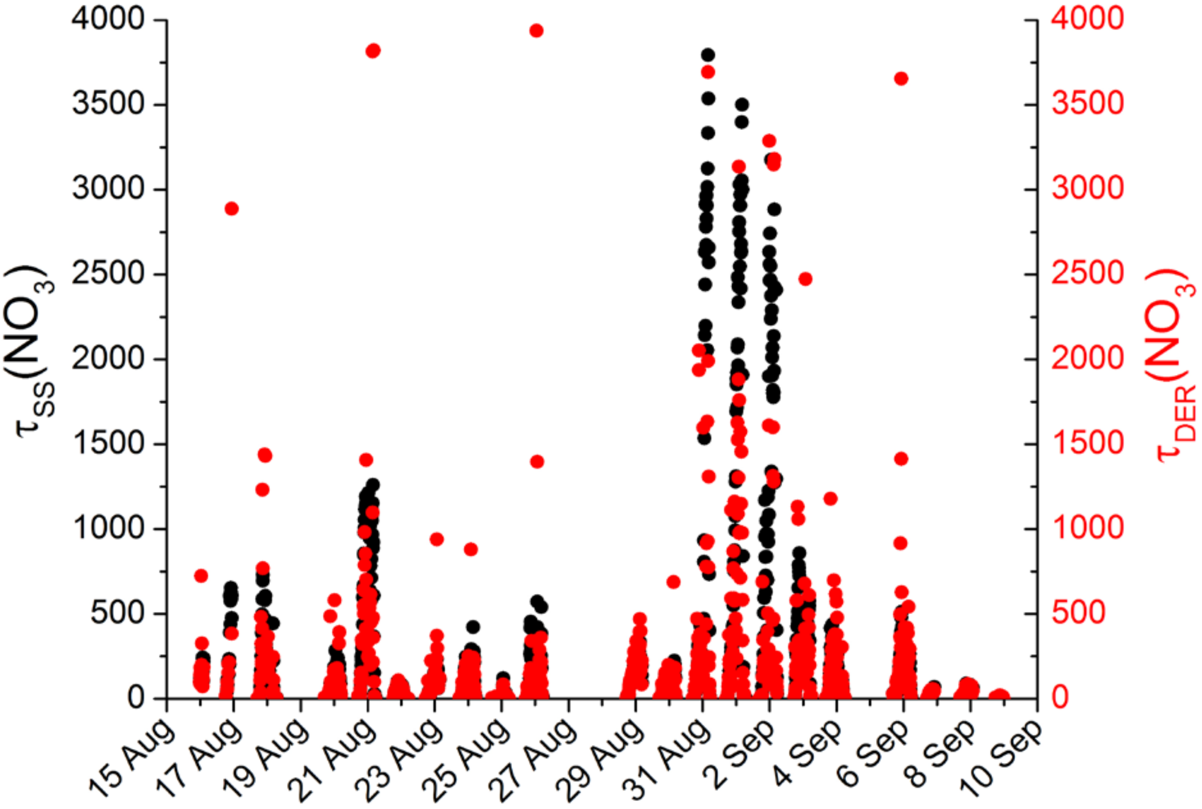

Fig. S3. Comparison between $\tau_{ss}(NO_3)$ and $\tau_{DER}(NO_3)$ using the same $[NO_3]$, $[NO_2]$ and $[O_3]$ datasets. For most of the nights both datasets agree even though outliers for $\tau_{DER}(NO_3)$ are sometimes calculated due to noise in the raw data.

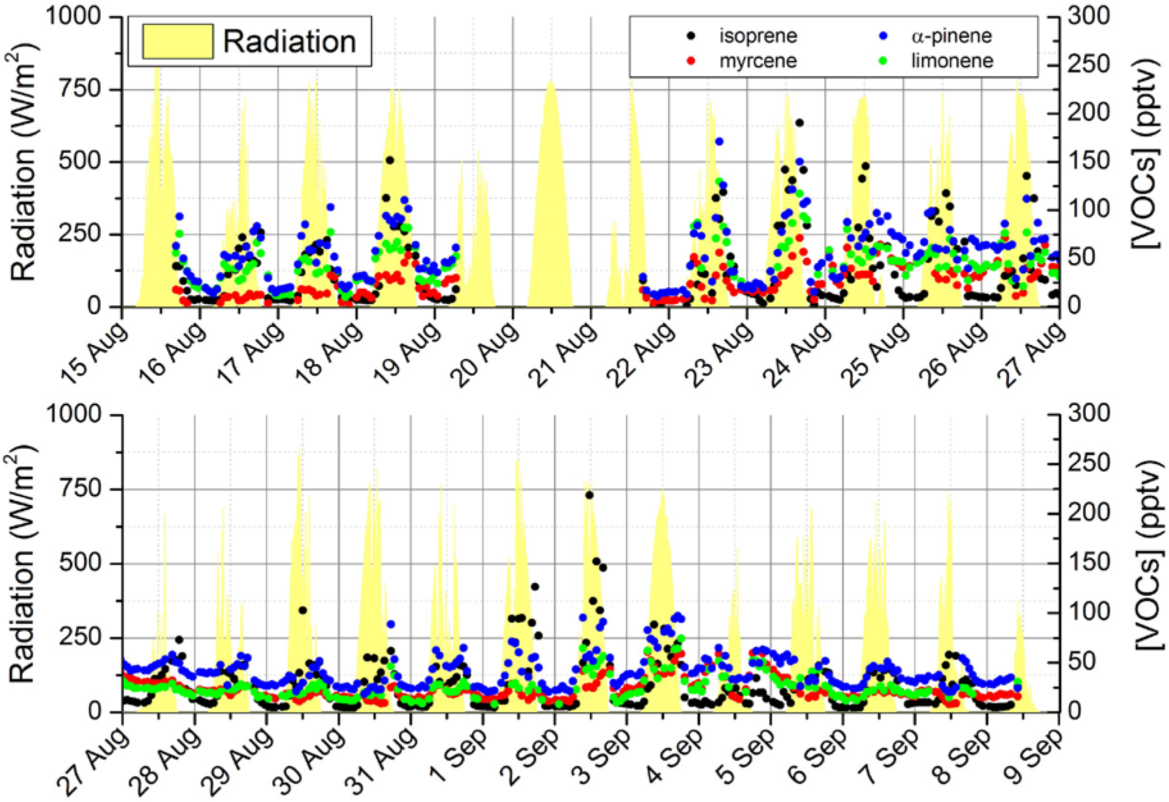

Fig. S4. Time series of the mixing ratios of the reactive (highlighted) VOCs in Table 2.

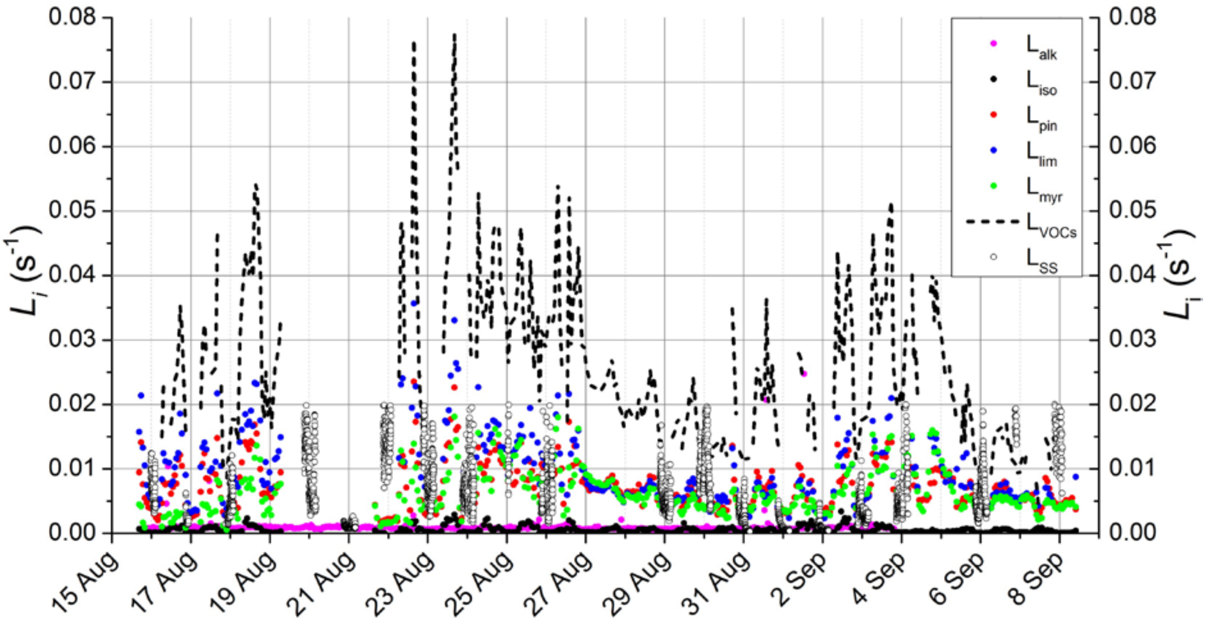

Fig. S5. Individual and summed reactivities (towards NO_3) of the VOCs listed in Table 2. L_{iso}, L_{pin}, L_{lim} and L_{myr} represent respectively the losses due to isoprene, α-pinene, limonene and myrcene. All the alkenes and alkanes reactivities are summed in the L_{alk} term. The L_{VOCs} term sums up the individual terms (L_{iso}, L_{pin}, L_{lim}, L_{myr} and L_{alk}). The steady-state loss rate constant for NO_3 (L_{ss}) is also plotted.

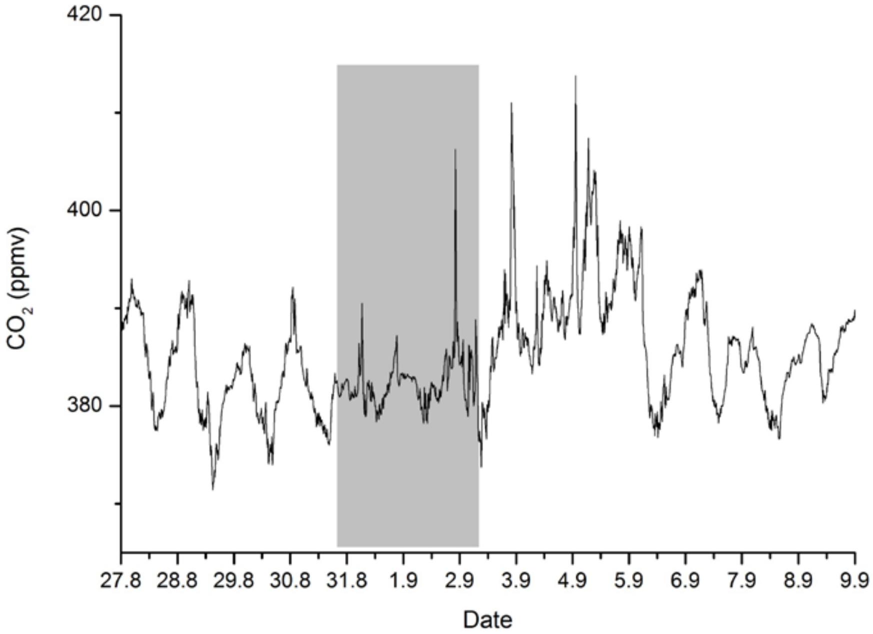

Fig. S6. CO_2 during PARADE. The grey shaded area indicates no significant nocturnal build-up of CO_2 (contrast e.g. the nights of 27.8 - 30.8 or 6.9 - 9.9) and is coincident with NO_3 lifetimes exceeding 2000 s.