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Chemistry, Transport and dry Deposition of Trace Gases in the Boundary Layer over the tropical Atlantic Ocean and the Guyanas during the GABRIEL Field Campaign

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We present a comparison of different Lagrangian and steady state box model runs with measurement data obtained during the GABRIEL campaign over the tropical Atlantic and the rainforest of the Guyanas in October 2005. Lagrangian modelling of boundary layer (BL) CO constrained by measurements of reactive trace gases and radiation is used to derive a horizontal gradient (≈ 5.6 pmol/mol km^{-1}) of this compound from the ocean to the rainforest (east to west). This is significantly smaller than that derived from the measurements (16-48 pmol/mol km^{-1}), indicating that photochemical production from organic precursors alone cannot explain the observed strong gradient. It appears that HCHO is significantly overestimated by the Lagrangian and "steady state" models, which include dry deposition but not exchange with the free troposphere (FT). Sensitivity runs show that only an unlikely combination of the variation of several parameters gives results close to the observed values. The relatively short calculated lifetime of HCHO (50-100 min) implies significant BL-FT exchange on small timescales. The mixing-in from above of FT air affected by African and South American biomass burning with an estimated rate of 0.12 h^{-1} (i.e. 12% of the BL air are exchanged with FT air per hour) increases the CO and lowers the HCHO mixing

ratios, leading to a better agreement of models and measurements. A 24 h mean deposition velocity of 1.35 cm/s for H_2O_2 over the ocean as well as over the rainforest is deduced assuming BL-FT exchange adequate to the results for CO. The measured increase of the organic peroxides from the ocean to the rainforest (≈ 0.66 nmol/mol d^{-1}) is significantly overestimated by the Lagrangian model, even when using high values for the deposition velocity and the entrainment rate. Our results point at heterogeneous loss of organic peroxides and/or their radical precursors. We calculate a mean integrated daytime net ozone production (NOP) in the BL of (0.2 ± 5.9) nmol/mol (ocean) and (2.4 ± 2.1) nmol/mol (rainforest). The NOP strongly correlates with NO and shows a positive tendency in the boundary layer over the rainforest.