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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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Abstract. We present a comparison of different Lagrangian and chemical box model calculations with measurement data obtained during the GABRIEL campaign over the tropical Atlantic Ocean and the Amazon rainforest in the Guyanas, October 2005. Lagrangian modelling of boundary layer (BL) air constrained by measurements is used to derive a horizontal gradient (≈5.6 pmol/mol km^{-1}) of CO 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 overestimated by the Lagrangian and chemical box models, which include dry deposition but not exchange with the free troposphere (FT). The relatively short lifetime of HCHO implies substantial BL-FT exchange. The mixing-in of FT air affected by African and South American biomass burning at an estimated rate of 0.12 h^{-1} increases the CO and decreases the HCHO mixing ratios, improving agreement with measurements. A 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 either heterogeneous loss of organic peroxides and/or their radical precursors, underestimated photodissociation or missing reaction paths of peroxy radicals not forming peroxides in isoprene chemistry. 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 has a positive tendency in the boundary layer over the rainforest.

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