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Unravelling the ozone-weather relationship: the role of vegetation and radical reactions

Tamara Emmerichs¹, Bruno Franco², Catherine Wespes², Simon Rosanka¹, and Domenico Taraborrelli¹

¹Forschungszentrum Jülich, Institute of Energy and Climate Research 8, Jülich, Germany (t.emmerichs@fz-juelich.de)

²Université libre de Bruxelles (ULB), Spectroscopy, Quantum Chemistry and Atmospheric Remote Sensing (SQUARES), Brussels 1050, Belgium.

Near-surface ozone is a harmful air pollutant, which is not only controlled by chemical production and loss processes. The major removal process of near-surface ozone is dry deposition accounting for 20 % of the total tropospheric ozone loss. Due to its significance, parameterizations used in atmospheric chemistry models represent a major source of uncertainty for tropospheric ozone simulations. This uncertainty might be one of the reasons why global models tend to overestimate ozone, when compared to observations. The model used in this study, the global atmospheric model ECHAM5/MESy (EMAC), is no exception. Like most global models, EMAC employs a “resistances in series” scheme, which is hardly sensitive to local meteorological conditions (e.g. humidity) and lacks non-stomatal deposition. In this study, these missing features have been implemented in EMAC affecting not only the deposition of ozone but also the removal of ozone precursors, resulting in lower chemical production of ozone.

Furthermore, near-surface ozone may be significantly impacted by water vapour forming complexes with peroxy radicals. The role of water in the reaction of HO₂ radical with itself and nitrogen oxides is known from the literature. However, in current models only the former is considered by assuming a linear dependence on water concentrations. Recent experimental evidence for the significant role of water on the kinetics of one of the most important reaction for ozone chemistry, namely NO₂ + OH, has been published. Here, the available kinetic data for the HO_x + NO_x reactions have been critically re-assessed and included in EMAC to test its global significance. Additionally, we considered the representation of isoprene and nitrous acid (HONO) as important oxidants for lower tropospheric chemistry. Namely, for isoprene emissions we added a drought stress factor which enables a higher sensitivity to meteorology leading to reduced emissions. Also, we firstly implemented soil emissions of HONO which is known as a missing source in models. The implications of these modifications on the global tropospheric composition are analysed, focusing on near-surface ozone and related precursors. The improved representation of ozone in EMAC is demonstrated using measurements from the Infrared Atmospheric Sounding Interferometers (IASI), the Tropospheric Ozone Assessment Report (TOAR) database and from the Trajectory-mapped Ozone-sonde dataset for the Stratosphere and Troposphere (TOST). The overall changes might help to reduce the uncertainty and overestimation

of models predicting near-surface ozone.