Comparison of Airborne Peroxy Radical Measurements with MECO(n) model simulation during EMeRGe in Europe

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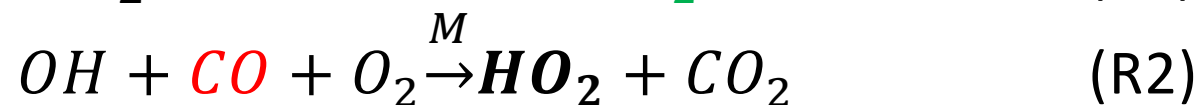[4]IEK-8, Forschungszentrum Jülich, Jülich, Germany

Online presentation for EGU2020 Session AS3.23: Impacts of emissions from major population centres on tropospheric chemistry and composition

Introduction

- The total sum of peroxy radicals, RO_2^*, measured by the PeRCEAS (<u>Pe</u>roxy <u>R</u>adical <u>C</u>hemical <u>E</u>nhancement and <u>A</u>bsorption <u>S</u>pectrometer) instrument have been compared with the MECO(n) (MESSy-fied ECHAM and COSMO models nested n times) model simulated RO_2 (defined as the sum of HO_2 + CH_3O_2 + ISOOH + CH_3CO_3 + $CH_3COCH_2O_2$ + $C_2H_5O_2$ in the model) for the EMeRGe campaign in Europe in 2017;

- In total 7 missions flights have been analyzed for this study and 3 will be showed in this presentation in details.

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Instrument measurement

The chemical amplification method is applied to amplify the conversion of radicals into NO_2 by a chain reaction with NO and CO as shown as (R1-R4):

$$\boldsymbol{HO_2} + \textcolor{cyan}{NO} \rightarrow OH + \textcolor{green}{NO_2} \qquad\qquad (R1)$$

$$OH + \textcolor{red}{CO} + O_2 \xrightarrow{M} \boldsymbol{HO_2} + CO_2 \qquad\qquad (R2)$$

$$RO_2 + \textcolor{cyan}{NO} \rightarrow RO + \textcolor{green}{NO_2} \qquad\qquad (R3)$$

$$RO + O_2 \rightarrow R'CHO + \boldsymbol{HO_2} \qquad\qquad (R4)$$

NO_2 is measured by a sensitive cavity ring-down detector, the peroxy radical ambient concentrations can be determined after the lab determination of the amplification factor.

Figure 1: Photo on the top is the view from outside HALO aircraft where the highlighted place is the inlet for the instrument; the photo at the show the PeRCEAS instrument and the part of inlet inside the aircraft cabin. The arrows indicate the sample flow direction.

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MECO(n) model

MECO(n) (Kerkweg and Jöckel, 2012a,b, Hofmann et al., 2012, Mertens et al., 2016, Kerkweg et al., 2018) is a global/regional chemistry-climate model developed by the MESSy consortium, which couples on-line the global chemistry-climate model EMAC with the regional chemistry-climate model COSMO-CLM/MESSy. The model results in this presentation provide COSMO-CLM/MESSy refinement with 7 km resolution, 330 x 310 horizontal gridboxes, timestep length = 60 s.

Measurement and model comparison

Three measurement flights were chosen as representative examples of the comparison between the PeRCEAS measurements and the MECO(n) model. Figure 2 is the flight track plotted with flight altitude.
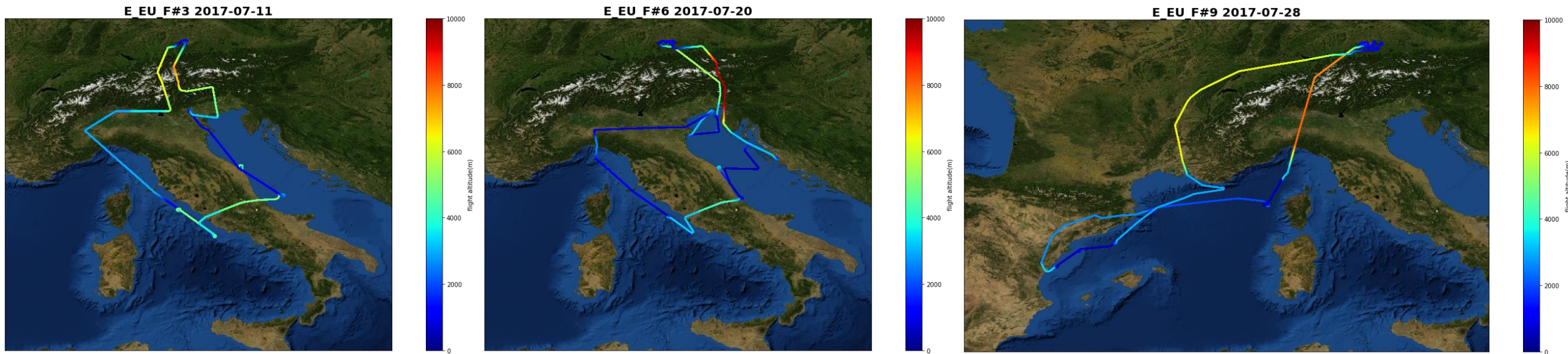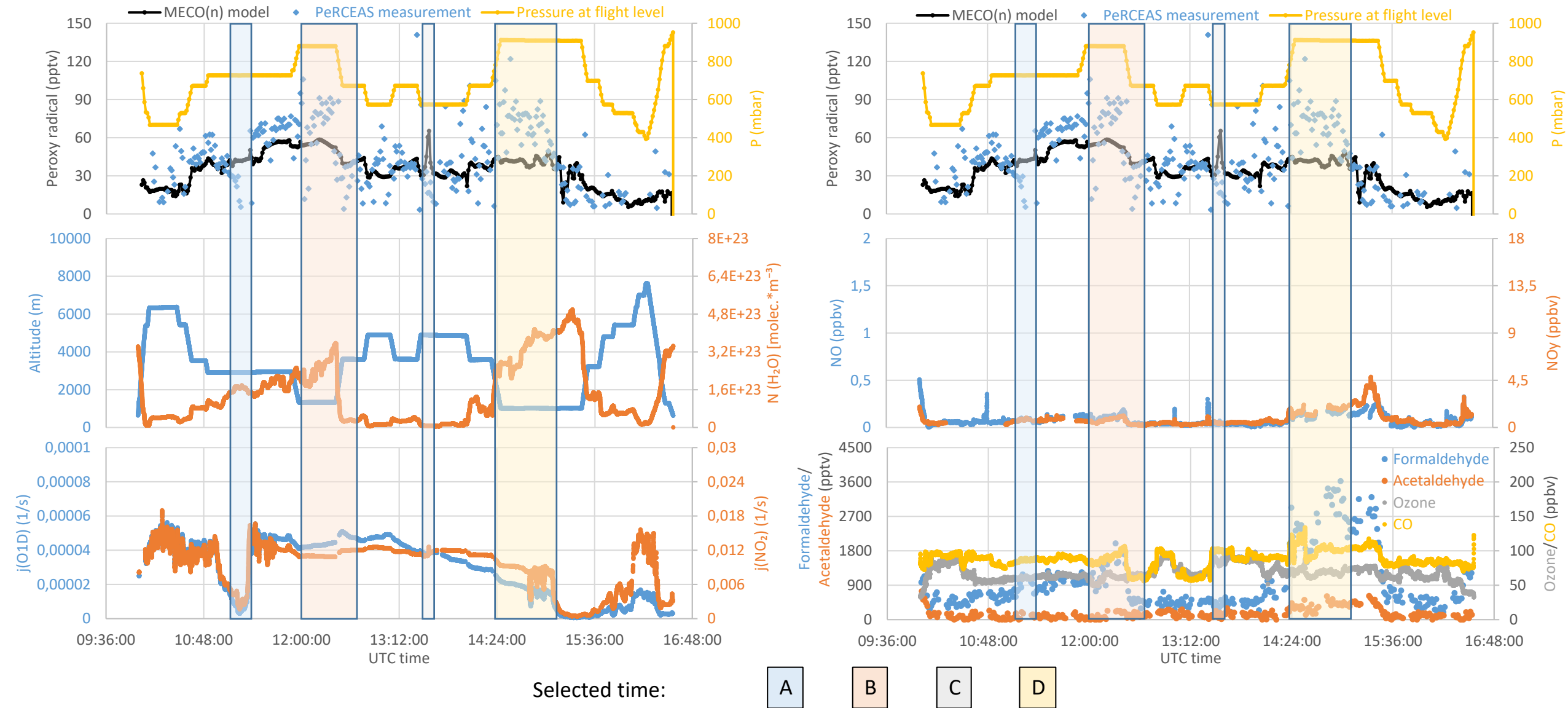

Figure 2: Flight track for E_EU_F#3, #6, #9 data from the sensor system of the HALO aircraft (BAHAMAS)

Temporal evolution EMeRGe E_EU_F#3 flight

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6

Comparison of the measured and modelled temporal evolution

- The temporal evolution of modelled and measured peroxy radicals agree reasonably for air masses with low concentration of pollutants;

- Different air masses were probed during E_EU_F#3. High differences between model and measurements are marked in the temporal evolution plot (shaded areas). The model seems not to reproduce short-term variations in trace gases and water concentration in polluted air masses;

- Significant variations in formaldehyde and acetaldehyde (marked as B in the plot) correspond with variations in peroxy radical measurements that are not simulated by the model;

- The reasons for overestimations (marked as C in the plot) in the model data will be further investigated.
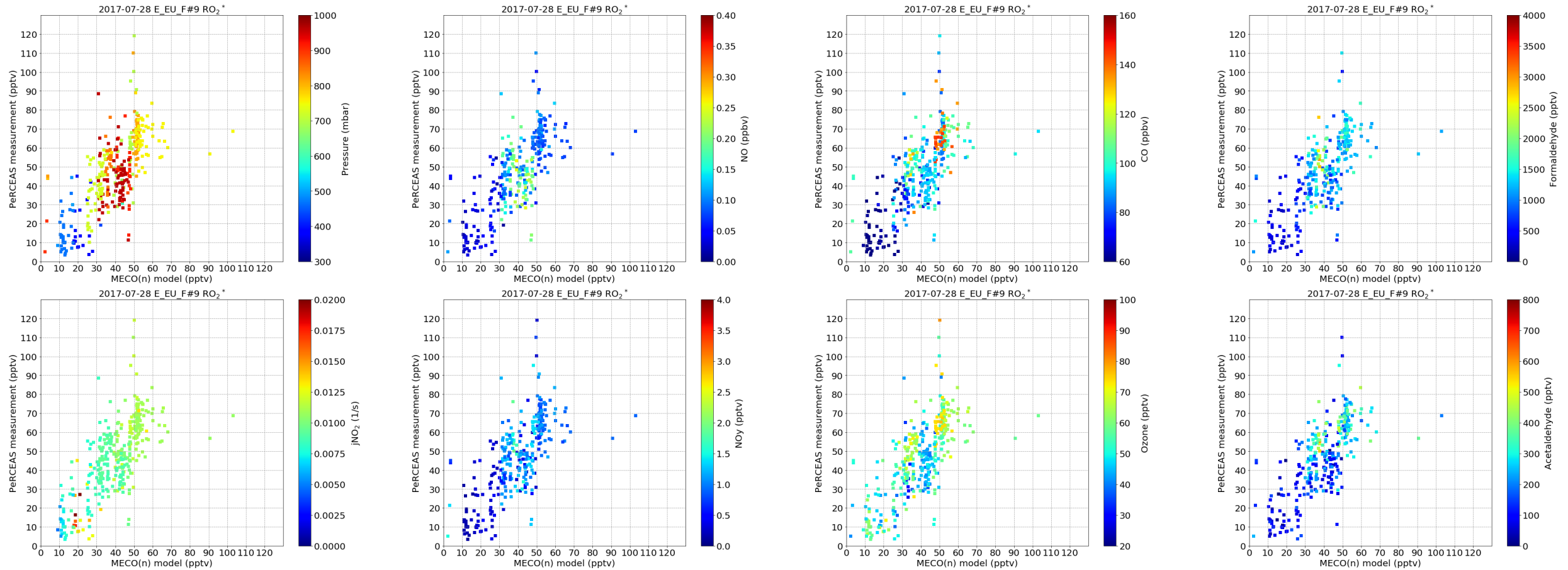
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Model and measurement correlations E_EU_F#3

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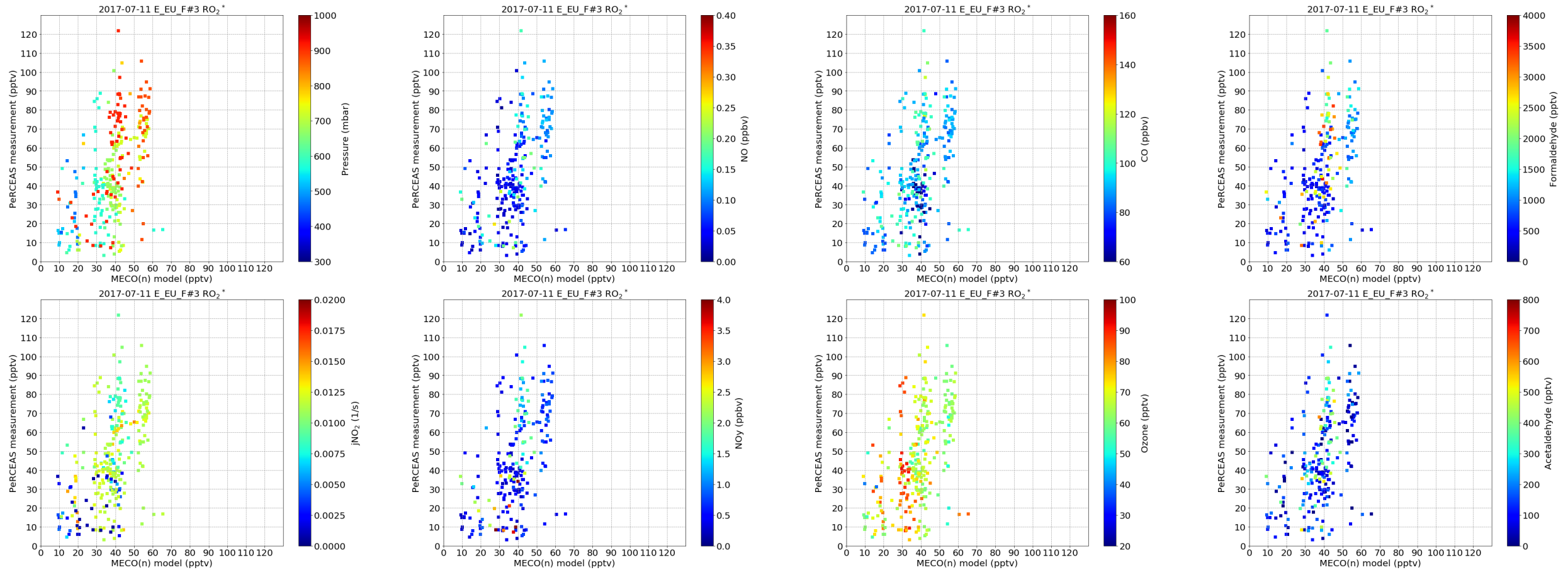
Model and measurement correlations E_EU_F#6

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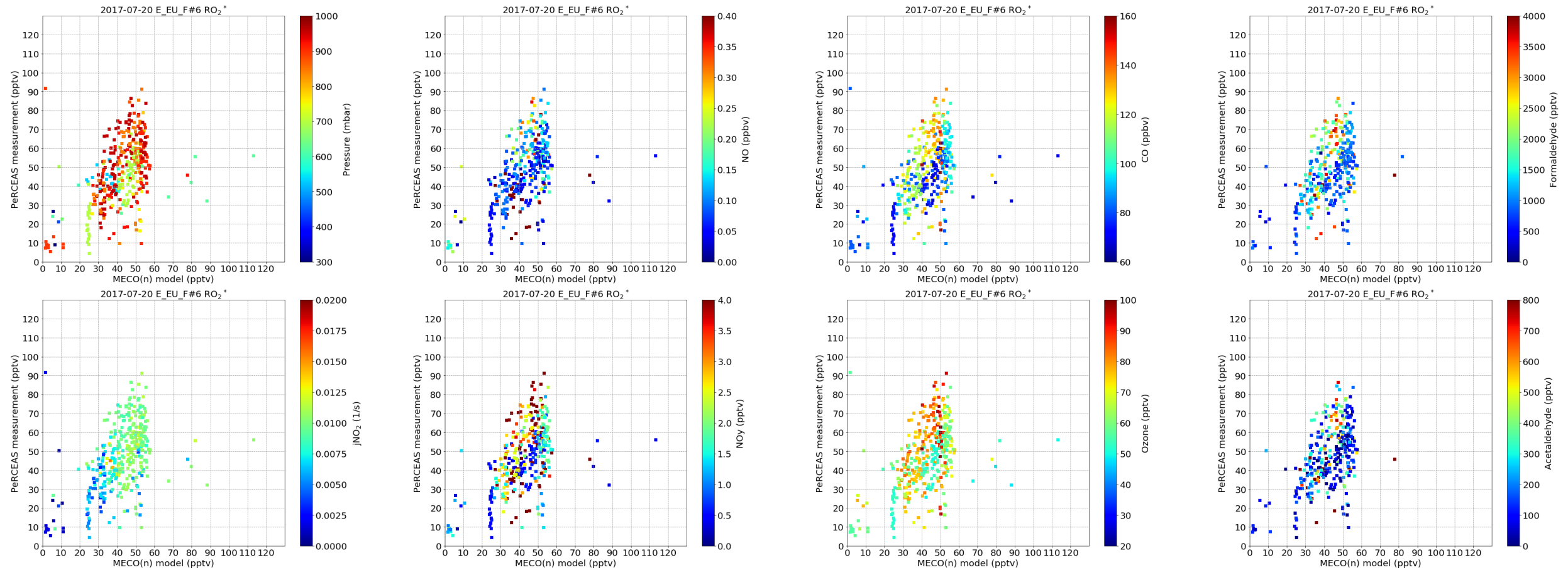
Model and measurement correlations E_EU_F#9

Discussion of the correlation plots

- Good correlations between model and measurements are systematically found at the lower values of the selected parameters, which agreed with the results from temporal evolution comparison;

- Sensible correlations between model and measurement correspond to periods of stable solar radiation as indicated by stable values in the photolysis rates;

- The number of points in the correlation plots changes due to missing measurement values;

- The model seems to underestimate significantly the measurements for events of high CO, O_3 formaldehyde (HCHO) and acetaldehyde (CH_3CHO).

Summary and outlook

- Model and measurements agree reasonably for periods with low to moderate variations in the peroxy radical mixing ratios. Short-term variations seem to be systematically underestimated by the model;

- The model seems to systematically underestimate the peroxy radicals measured in polluted plumes, typically associated with variations in peroxy radical precursors such as formaldehyde, acetaldehyde, and CO.

- Further comparison using different model sensitivities and trace gas inventories will be continued.

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