Airborne measurement of peroxy radicals during EMeRGe

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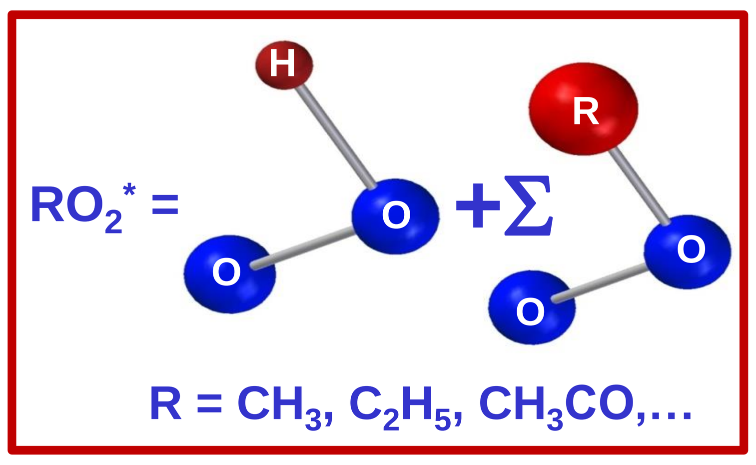
Introduction

The project **EMeRGe** (**E**ffect of **Me**gacities on the Transport and Transformation of Pollutants on the **R**egional to **G**lobal Scal**e**s, www.iup.uni-bremen.de/emerge) focuses on the understanding of the transport and transformation of polluted plumes of air flowing from selected major population centres (**MPC**) in Europe and Asia using the **HALO** (**H**igh **A**ltitude and **Lo**ng Range Research Aircraft, www.halo.dlr.de) platform.

One key issue with respect to transformation is the amount and role of peroxy radicals, HO_2 and RO_2 (where R is an organic group), as their reactions play a key role in oxidation processes and determine the O_3 budget and the oxidation capacity of the atmosphere (**OCA**).



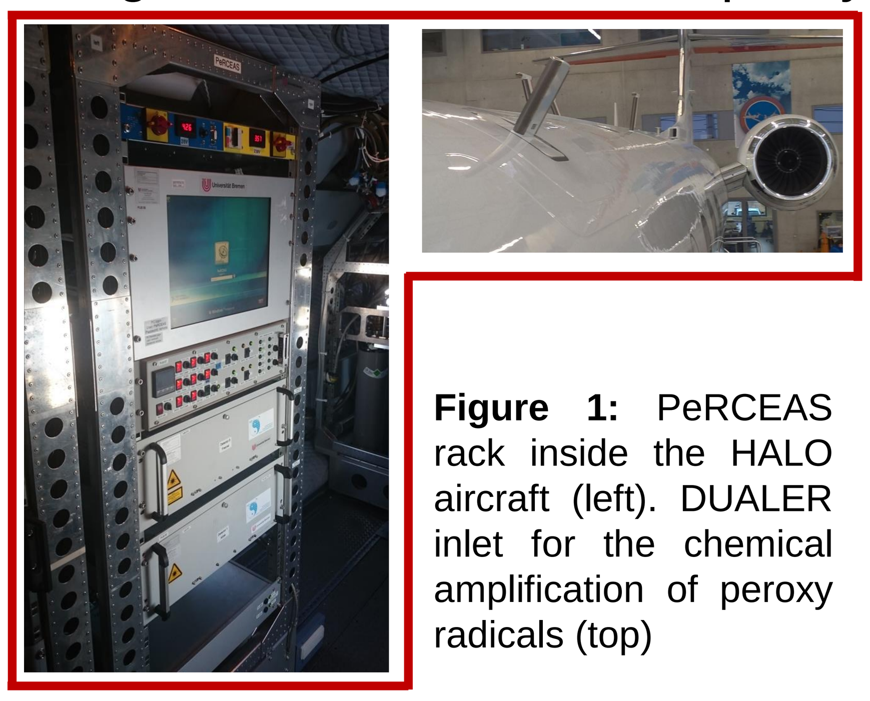


Figure 1: PeRCEAS rack inside the HALO aircraft (left). DUALER inlet for the chemical amplification of peroxy radicals (top)

Within EMeRGe**,** airborne measurements of the total sum of peroxy radicals, $\mathbf{RO_2^*} = HO_2 + \sum RO_2$, were conducted by using **PeRCEAS** (**Pe**roxy **R**adical **C**hemical **E**nhancement and **A**bsorption **S**pectrometer) which is part of the HALO EMeRGe payload.

PeRCEAS combines the **PeRCA** (**Pe**roxy Radical Chemical Amplification) measurement technique, for the amplified conversion of radicals entering the reactor into NO_2 in a chain reaction involving NO and CO, with the sensitive detection of NO_2 by **cw-CRDS** (**c**ontinues **w**ave - **C**avity **R**ing **D**own **S**pectroscopy) technique. The instrument shares a common inlet for two identical measurement lines (reactor-detector) to improve time resolution and sensitivity.

The amplification factor (chain length) and the detection limit were determined in the lab.

PeRCEAS airborne RO_2^* measurements within EMeRGe

PeRCEAS successfully participated in the EMeRGe HALO campaigns in summer 2017 and spring 2018. A total of 180 flight hours were distributed among seven mission flights over Europe (Figure 2, left), fourteen mission flights over Asia (Figure 2, right) and six transfer flights.

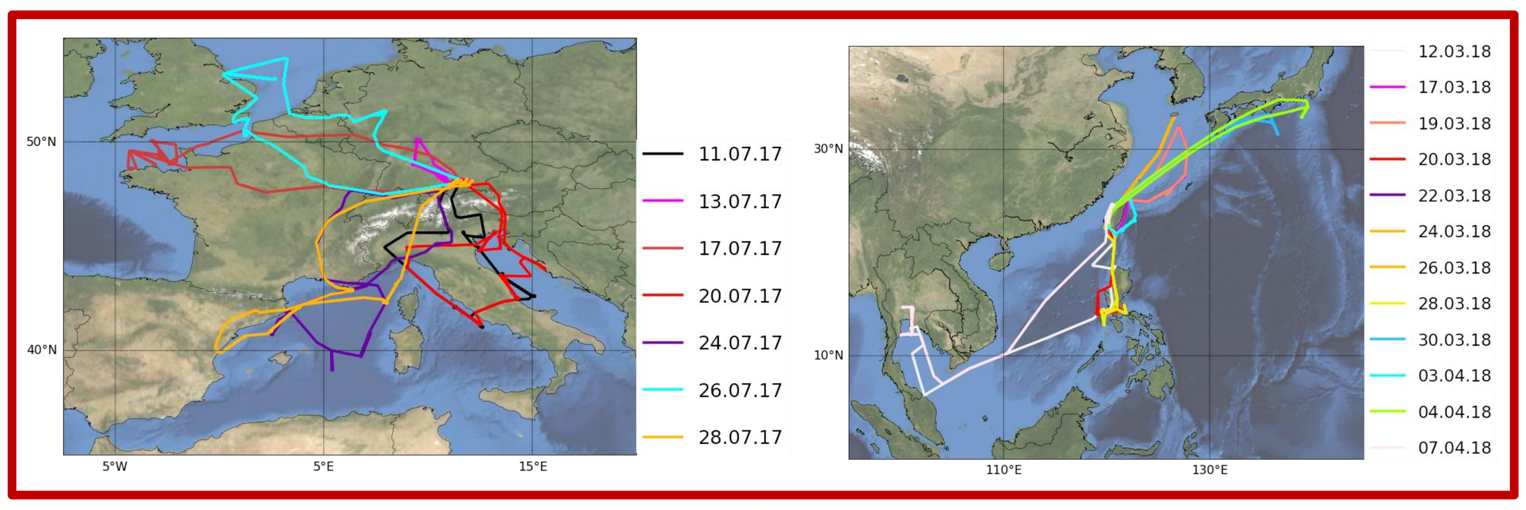


Figure 2: HALO mission flight tracks of the EMeRGe campaign in Europe (left) and in Asia (right)

In Europe the MPC outflows of London, Rome, Po Valley, Paris, Benelux/Ruhr, South France, Madrid and Barcelona were investigated.

The outflows of Bangkok, Manila, Taipei, and the transport of MPC emissions from China, South Korea and Japan over the Yellow Sea were in turn a main focus of the campaign in East Asia.

Overall significant RO_2^* mixing ratios up to 80 pptv were measured in both campaign phases.

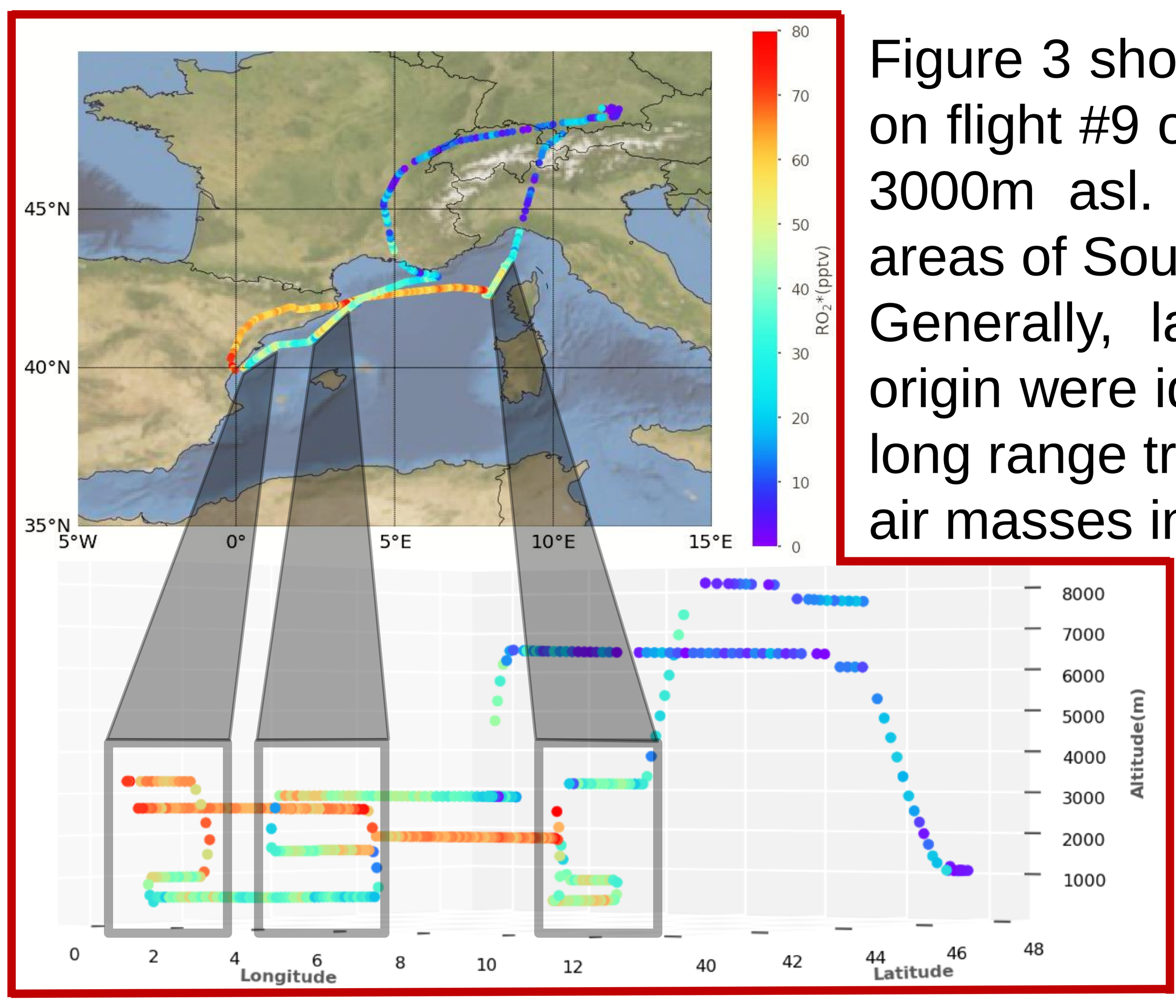


Figure 3: Preliminary RO_2^* volume mixing ratios measured on 28.07.2017 over the flight track (top). The RO_2^* vertical distribution at the shuttle points is additionally highlighted (bottom)

Figure 3 shows exemplary the RO_2^* mixing ratios measured on flight #9 over Europe. HALO shuttles between 500m and 3000m asl. were carried out at the expected downwind areas of South France, Madrid and Barcelona.

Generally, layers of different photochemical activity and origin were identified confirming the effect of convective and long range transport processes on the composition of air masses in the boundary layer and beyond.

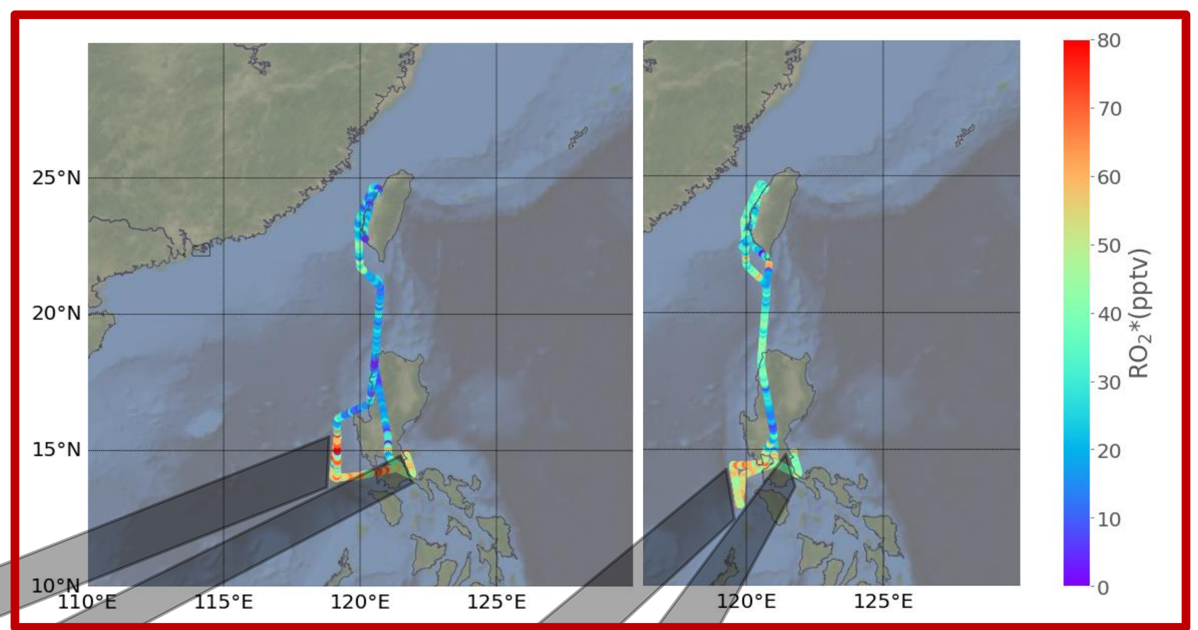


Figure 4: Preliminary RO_2^* volume mixing ratios measured on the EMeRGe flights on 20.03.2018 (left) and 28.03.2018 (right) over Manila

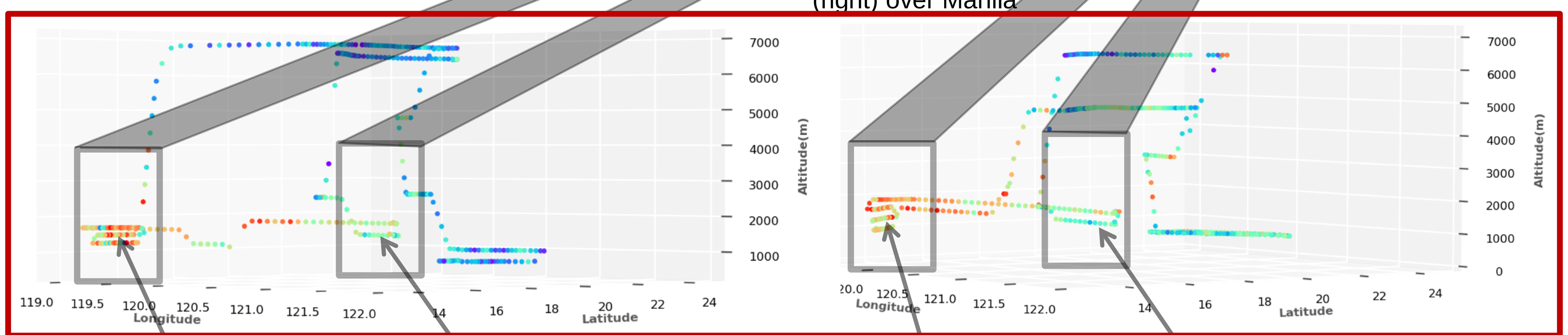


Figure 5: Preliminary RO_2^* vertical distribution measured during the EMeRGe flights over Manila on 20.03.2018 (left) and 28.03.2018 (right). The vertical distribution at the shuttle points upwind and downwind are highlighted.

Figure 4 and 5 shows RO_2^* mixing ratios measured during the EMeRGe flights in Asia investigating the outflow from Manila in March 2018. HALO shuttles at altitudes between 800m and 3000m asl. were carried out upwind and downwind of Manila as confirmed by the FLEXPART sensitivity maps shown in Figure 6.

The higher RO_2^* mixing ratios encountered downwind Manila are in agreement with the expected input of precursors and photochemical processing in within the MPC plume.

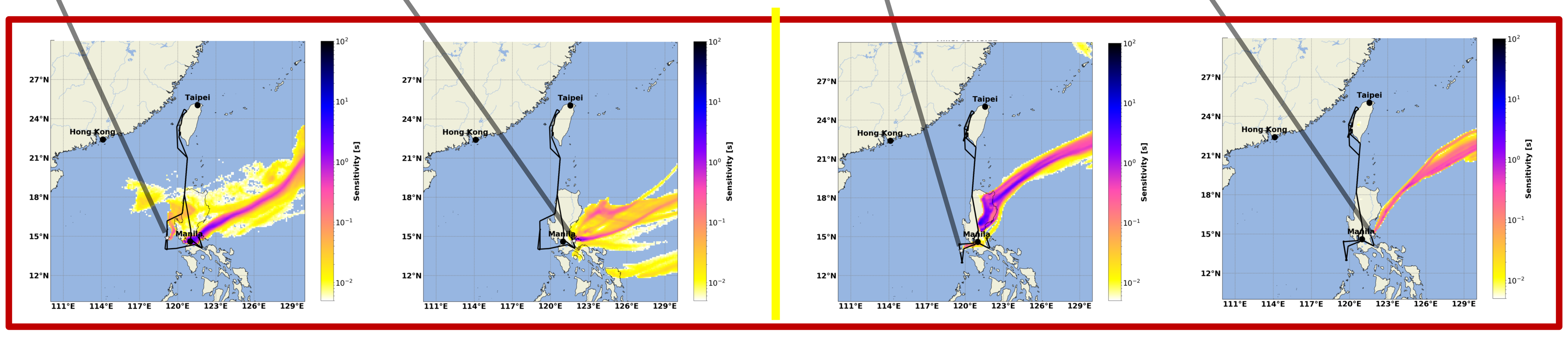


Figure 6: Sensitivity maps produced by FLEXPART back trajectory calculations using black carbon as a tracer for the EMeRGe flight on 20.03.2018 (left) and 28.03.2018 (right) showing the origin of the air mass probed.

Further investigation of the OCA and the regional O_3 production upwind and downwind of the selected MPCs is required. Moreover, laboratory experiments are presently being performed to improve the accuracy of the chain length used at different measurement conditions.

Acknowledgement

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