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**Airborne peroxy
radicals within AMMA**

M. D. Andrés-Hernández
et al.

Peroxy radical observations over West Africa during the AMMA 2006 campaign: Photochemical activity in episodes of formation of convective systems on the basis of radical measurements

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Peroxy radical measurements made on board the DLR-Falcon research aircraft over West Africa within the African Monsoon Multidisciplinary Analysis (AMMA) campaign during the 2006 wet monsoon are presented in this study. The analysis of data focuses on the photochemical activity of air masses sampled during episodes of intense convection and biomass burning. Generally, the total sum of peroxy radical mixing ratios, measured in the outflow of convective clouds, are quite variable but occasionally are coupled with the NO variations indicating the coexistence, or simultaneously emission of NO_x, with a potential radical precursor (i.e., formaldehyde, acetone or peroxides) which has likely been transported to higher atmospheric layers. Based on the measurements, significant O₃ production rates up to 2 ppb/h in the MCS outflow are estimated by using a box model with simplified chemistry. Peroxy radicals having mixing ratios around 20–25 pptv and with peak values of up to 60–70 pptv are measured within biomass burning plumes, detected at the coast in Ghana. Calculations of back-trajectory densities confirm the origin of these air masses being a biomass burning region at southern latitudes and close to the Gulf of Guinea, according to satellite pictures.

Measured peroxy radical concentrations agree reasonably with modelled estimations taking into account simple local chemistry. Moreover the vertical profiles taken at the aircraft base in Ouagadougou, Burkina Faso, indicate the common feature of having maximum concentrations between 2 and 4 km, in agreement with other literature values obtained under similar conditions.

1 Introduction

Hydroperoxyl (HO₂) and organylperoxyl (RO₂; R=organic chain) radicals, are involved in most of the oxidation mechanisms taking place in the troposphere. Knowledge of their amounts and distributions provides essential information about the aging and his-

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tory of an air mass. In spite of their importance in the chemical processing of the troposphere, only a very limited number of measurements are available in the literature. This arises from their high reactivity, which complicates their measurement and results in small mixing ratios. Measurements of tropospheric free radicals are particularly scarce between 30 degrees South and 30 degrees northern latitudes as reported by Cantrell et al. (2003a).

The mesoscale convective systems (MCS) enclosed into synoptic-scale African Easterly Waves during the West African Monsoon are considered to be the origin of about 40% of the Atlantic tropical cyclones and responsible for troposphere-stratosphere exchange (Agusti-Panareda and Beljaars, 2008). Africa is therefore a suitable environment for investigating the photochemical activity in air masses impacted by MCS. Little is known about the chemical composition of these air masses during intense convective episodes. HO_x in the upper troposphere (6 to 12 km) cannot generally be sustained by the primary production of OH from the reaction of O¹D, produced by the photolysis of O₃ with H₂O and the subsequent reactions of OH with CO and O₃. Transported HO_x precursors other than O₃ and H₂O seem to be the primary sources of hydrogen containing radicals in much of the upper troposphere (Wennberg et al., 1998). In the presence of enough NO from lightning, biomass burning, and aircraft emissions the resultant catalytic mechanism can be responsible for rapid and effective production of O₃ in the higher layers of the atmosphere and have a global impact (Jaeglé et al., 2001). Prather and Jacob (1997) suggested that given a 10 days overturning rate of the tropical upper troposphere, deep convection could cause a persistent chemical imbalance in HO_y (defined as OH, HO₂ and their non-radical reservoirs, i.e., HO_y=OH+peroxy+2H₂O₂+2CH₃OOH+HNO₂+HNO₄). This has also been confirmed by the measurements of peroxides and CH₃OOH of Cohan et al. (1999) in aged convective outflows in the tropical Pacific. Conversely, HO_x greater than the model predictions for high NO regimes (500–600 pptv) have been measured in biomass burning plumes encountered over the western Pacific equatorial region (Folkins et al., 1997). Similarly, unexpected high OH concentrations have been measured over the pristine

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Amazon forest (Lelieveld et al., 2008). It has been proposed that natural isoprene oxidation recycles OH efficiently in low NO_x air through reactions of organic peroxy radicals.

The Institute of Environmental Physics of the University of Bremen (IUP-UB) participated in the AMMA measurement campaign taking place during the wet monsoon season in August 2006. IUP-UB contributed with the measurement of the total sum of peroxy radicals (RO₂^{*}=HO₂+RO₂) onboard of the German DLR-Falcon. This was part of a suite of measurements from aircraft, ground stations and satellites to study the composition and the oxidative capacity of air masses over West Africa with special focus on the impact of MCS within the monsoon period (Reeves et al., 2009). In addition, biomass burning plumes were observed during the measurement campaign.

Six DLR-Falcon flights were conducted from Ouagadougou in Burkina Faso (12.15° N 1.30° W) in the period from 1 to 18 August 2006. An overview of the aircraft campaign and details of the flights are described in Reeves et al. (2009).

2 Experimental

RO₂^{*} was measured by using an airborne PERCA (PEroxy Radical Chemical Amplifier) instrument, with a double detector, i.e., a dual channel airborne peroxy radical chemical amplifier (DUALER). Briefly, the DUALER utilises the simultaneous detection of the NO₂ mixing ratios by two identical reactors using a common sampling line. NO and CO are alternatively added to the reactors in such a way that the chemical conversion of RO₂^{*} into NO₂ in a chain reaction takes place in one reactor (amplification mode) while the other measures the NO₂ background, i.e., the NO₂ present in the air sampled plus the NO₂ originating from all other sources up to the detector such as reaction of NO with O₃, the decomposition of PAN, etc. (background mode). The modes of the reactors are switched every 60 s. NO₂ is measured by detecting the light emitted from the chemiluminiscent reaction of NO₂ with a solution of Luminol. Provided that the length of the chain reaction for the reactors (CL: chain length) is known, the difference

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in the signal of the detectors (amplification-background) can be related quantitatively to the RO_2^* content in the air sampled. More details about the instrument principle, characteristics, performance und characterisation are provided elsewhere (Kartal et al., 2009).

5 2.1 Critical aspects of the measurement of peroxy radicals

In previous studies it has been shown that the CL of the PERCA decreases with the pressure (Kartal et al., 2009). For this reason the DUALER is maintained at a constant pressure, lower than the ambient, during the measurement in the aircraft and the corresponding CL is determined in the laboratory. In order to cover the range of the pressure levels during the flights but still keeping a reasonable detection limit, 200 mbar was the pressure selected for the AMMA campaign. A pressure gradient $\Delta P = P_{\text{ambient}} - P_{\text{DUALER}}$ of 70 mbar is required for successful measurement. This limits somewhat the amount of RO_2^* data available during the flights for the characterisation of the MCS outflow which were often carried out at altitudes corresponding to pressure levels lower than 250 mbar.

Concerning the vertical profiles of trace gases taken by the DLR-Falcon, it is important to note that the aircraft normally remains at each single pressure level only for a few minutes. This normally leads to poor statistics of the RO_2^* measurement signals, especially in the case of unexpected variations in the concentrations.

Another critical aspect of the DUALER measurements during AMMA is related to the NO_2 calibrations of the detectors. The concentration of the NO_2 gas cylinder built in the DUALER for calibrations was not stable during the campaign. This is attributed to the high temperatures and humidity reached in the aircraft prior to the flight, which possibly caused that the wall losses at the pressure regulator and gas tubing change along the day in spite of relatively long flushing. This resulted in inconsistent in-flight NO_2 calibration of the detectors, which could only be calibrated before and after the measurement flight. To overcome these issues and to monitor potential changes in the sensitivity of the detector during the flight, a mathematical method based on the

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ozone concentrations measured simultaneously on board the DLR- Falcon has been developed for the interpretation of results and is described in more detail elsewhere (Kartal et al., 2009).

2.2 Data from other instrumentation

The analyses of the air masses undertaken in this study used measurements of other constituents and parameters made aboard the DLR- Falcon. These data are provided by the DLR-Institute of Atmospheric Physics. O₃ and CO₂ were measured using UV and IR absorption techniques, respectively (Schlager et al., 1997; Schulte et al., 1997). CO was detected with an UV fluorescence instrument (Gerbig et al., 1996). Reactive nitrogen compounds (NO, NO_y) were measured using two O₃/NO-chemiluminescence detectors and a gold converter for reduction of higher oxidized NO_y compounds to NO (Schlager et al., 1997; Ziereis et al., 2000). Formaldehyde, HCHO, was detected by a fluorometric technique (Hantzsch reaction). Characteristics of the instruments (accuracies, detection limits) are summarized in Reeves et al. (2009).

2.3 Supporting calculations

Clusters of back trajectories have been launched in the vicinity of the aircraft location to gain additional information about the origin of the air masses sampled within the case studies. The trajectory calculations were made using the FLEXTRA model (Stohl et al., 1995, 1998). In order to account for uncertainties in the meteorological data, these trajectory clusters were released from a volume of 0.6×0.6 degrees (horizontally)×1000 m (vertically)×1 h (time) centred at the aircraft position and time of the corresponding measurements. All back trajectories are followed for 72 h. The trajectory density, i.e., the number of trajectories passing a certain volume of air, has been additionally determined. For the analysis of AMMA data the trajectory density bases on a grid of 0.25°×0.25°×100 m. Subsequently the trajectory densities are normalised to 1.

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Furthermore, the Lagrangian Boxmodel BRAPHO (BRemen Atmospheric PHOtochemical model) (Meyer-Arnek, 2005) has been used to analyse the photochemical evolution of the probed trace gases. BRAPHO calculates the chemistry within a closed volume of air, on the basis of the Master Chemical Mechanism (MCM) (Saunders et al., 2003). For the present work, a simplified photochemistry (see Appendix A) was used for the estimation of the local O₃ production under the measurement conditions. The simulations are initialised with the corresponding measurements taken on board the DLR-Falcon.

3 Results

Air masses of different photochemical activity were sampled during the AMMA measurement campaign. In addition to measurement uncertainty, there are many physical and chemical factors which control the variations of the RO₂* observed, e.g. the different composition of air masses transported from different source regions, the variations in the actinic radiation fields, possible loss reactions in the presence of clouds etc. Within the same pressure level, chemical episodes with different balance between controlling variables can often be distinguished. Generally, change of pollution plume type is indicated by short term variations of NO_x, NO_y, CO₂ and CO concentrations. As a surrogate for the volatile organic compounds (VOC), HCHO was measured by the instrument payload on the DLR-Falcon. The HCHO data coupled with the variations of CO and CO₂ can be used to identify the arrival of air masses of different origins and likely different loading of VOCs.

In Fig. 1 the trace gases measured at the 260 mbar pressure level during the DLR-Falcon flight on the 15 August 2006 are shown demonstrating the chemical complexity of air masses observed during AMMA. The objective of the flight was to probe the outflow of a MCS over northern Benin. The DLR-Falcon remained at the same pressure level between 15:02 and 15:52 h. While j_{NO₂} does not indicate significant variations in the cloud cover, the trace gas concentrations change considerably. Between 15:20 and

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15:30 h CO sharply increases up to 135 ppbv while NO and NO_y rise above the background level and show high variability up to 1.2 and 2.2 ppbv respectively. RO₂^{*} generally show high variability and are anticorrelated to NO variations as expected according to the known chemistry, i.e., the formation of RO₂^{*} via reaction of CO (and/or VOC) with OH in the presence of H₂O, and the depletion in the presence of NO_x by loss reactions. However, RO₂^{*} occasionally follow quite closely (see Fig. 1) the NO behaviour. These simultaneous increases in CO, CO₂, NO/NO_y, RO₂^{*} and O₃ are indicative of local photochemical activity being triggered by fresh emissions of potential radical precursors together with the emission and/or transport of NO_x and other pollutants. This leads to an O₃ production in the presence of radicals. Box model calculations initialised with the conditions observed at 15:25 h indicate that the radical chemistry in the air mass is responsible for an O₃ production of approximately 1.7 ppb h⁻¹.

The focus of this manuscript is the analysis of RO₂^{*} for selected episodes and behaviours observed during the AMMA. Specifically convective outflow from clouds, biomass burning plumes and the vertical distributions observed above Ouagadougou were selected for study.

3.1 RO₂^{*} measurements within convective episodes

Data obtained during the first flight on the 15 August 2006 are depicted in plate 2. The objective of the flight was to measure the outflow of a MCS located over Mali after passing Ouagadougou the evening before. The DLR-Falcon reached this outflow area at the 287 mbar pressure level. The NO_y and NO_x data measured during the flight (Fig. 2b) show the typical signature of high variable concentrations within this kind of convective outflow. CO remains at 100–110 ppbv, about 20 ppbv higher than outside the outflow area. As mentioned in Sect. 2.1, RO₂^{*} measurements are only available for pressures >250 mbar.

Figure 2c focuses on the RO₂^{*} results, which on the average remain about 10 pptv. The increase of RO₂^{*} with NO up to the 0.5 ppb maximum (Fig. 3), and the similarity of the RO₂^{*} and NO variation patterns are remarkable. Rather it is expected that

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RO_2^* decrease as NO increases because the sink reactions of radicals with NO_x are gaining in importance. The observed behaviour thus indicates the presence of a radical precursor being emitted simultaneously with NO. This could be a peroxy radical source, either transported by convection from another region and/or locally produced.

5 O_3 remains practically constant at 45 ppbv for the whole period.

The convective injection of peroxides has been suggested to be a source of HO_2 radicals (Prather et al., 1997; Cohan et al., 1999; Jaeglé et al., 1997; Faloon et al., 2000). The convective pumping of carbonyl compounds like formaldehyde, acetaldehyde or acetone may also be responsible for local production of RO_2^* . Faloon et al. (2000), already reported on unexpected HO_2 values in different masses in the upper troposphere (at or above 6 km) in the presence of $100 < \text{NO} < 500$ pptv. These authors interpret the model underestimation of the HO_2 as a possible unmeasured HO_x source that happened to be spatially correlated with NO_x , independent of its origin. A similar underestimation has also been reported both for HO_2 and $\text{HO}_2 + \text{RO}_2$ measurements performed during the TRACE-P campaign by using Peroxy radical Chemical Ionization Mass Spectroscopy (PerCIMS) (Cantrell et al., 2003b). Similarly, acetone seems to explain most of the missing HO_x in models for $\text{H}_2\text{O} < 25$ to 30 ppm in the upper troposphere over the Pacific (McKeen et al., 1997).

20 The available DLR-Falcon measurements of HCHO at this pressure level (Fig. 2c) though presenting similarities with the NO and NO_y pattern are not enough to make any conclusive interpretation. On the other hand H_2O remained around 1000 ppm at the 287 mbar pressure level. However, as the NO to NO_2 ratio is relatively high (between 7 and 9) if acetone would be present, it should be quite effective in the production of HO_x , as the competing reaction of the formation of peroxyacetyl nitrate (PAN) is favoured by low NO to NO_2 ratios.

25 Temporal variations in humidity confirm local inhomogeneity caused by the convection at this pressure level. Increases in humidity at the 10 km flight altitude indicate air masses being uplifted by convection (Fig. 4). This has an effect in the local chemistry which might be responsible for the variability of the NO_x and RO_2^* in situ values. In

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order to obtain a representative picture of the photoactivity of the air mass within the MCS outflow and at this pressure level as a whole, the O_3 production rate was estimated for three different cases by initialising the box model with the measurements at 10:20, 10:23 and 10:25 h. These simulations lead to O_3 production rates of 5.4, 1.5 and 0.2 ppb h^{-1} , respectively. These values are in reasonable agreement with the simulations in MCS outflow reported by Schlager et al. (2009). In this work, O_3 production rates between 4 and 8 ppb day^{-1} are obtained assuming different VOC and dilution patterns in the MCS outflow.

Another example of convective outflow is given by the flight on the 4 August 2006. At the 315 mbar pressure level the DLR-Falcon sampled the outflow of the MCS which has passed over Niamey the day before (Fig. 5). The aircraft remained within the MCS outflow till approximately 09:50 h while keeping the pressure level till circa 10:20 h. This is clearly confirmed by the variation of the absolute humidity with a clear decrease in the values when leaving the outflow zone (Fig. 6). NO and NO_y are highly variable reaching 2.5 and 1.2 ppbv, respectively, at the beginning of the level around 09:15 h. At this point RO_2^* remain undetectable till NO decreases down to the 0.2–0.5 ppbv level around 09:30 h. The increase in the NO/NO_y ratio from 0.04 up to 0.5 indicates the presence of relatively fresh emissions. Similarly to the 15 August, as the NO/NO_2 ratio remains relatively high (around 7) most of the time, if acetone which has a lifetime of several days were simultaneously transported with NO , the production of radicals would be favoured. This might explain the variability of the RO_2^* mixing ratios, occasionally following the NO variations and reaching up to 40 pptv. The error bars depicted in Fig. 5b correspond to the statistical error of the 20 s averages. The total error of the RO_2^* 20 s averages is estimated to be within 45%. As mentioned in the Sect. 2.1 and described more in detail elsewhere (Kartal et al., 2009), the total RO_2^* error partly depends on the quality and variability of the O_3 measurements and therefore varies for any particular measurement interval. The presence of clouds likely introduces some additional data variability. The role of clouds as sink of radicals has been often discussed in the literature but still requires further clarification.

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No obvious correlation with HCHO is observed. This does not exclude however the presence of another source of organic radicals produced from the oxidation of carbon compounds. Vertical transport from the layers below seems to be the most likely explanation of the observed NO. As presented in the summary of the AMMA observations by Reeves et al. (2009), the general pattern in the trace gas concentrations observed along the whole latitude range measured is indicative of the transport by convection to higher altitudes. The pattern comprises two layers of higher mixing ratios, one close to the surface up to 900 hPa and another at higher altitudes. Similar results are recently reported by Ancellet et al. (2008). For NO, given the low mixing ratios measured within the boundary layer close to the surface (Reeves et al., 2009), this transport must be enhanced by NO_x from lightning, which is expected during storm episodes within deep convective clouds.

After leaving the convective zone around 10:00 h, CO increases gradually from 90 to 120 ppb, CO₂ from 374 up to 379 ppmv, NO decreases down to zero and the RO₂^{*} variability diminishes notably (RO₂^{*}: 5–25 pptv).

The box model was initialised with the measurements at 09:30, 09:50 and 10:00 h representing different RO₂^{*}/NO ratios inside the MSC and outside the convection zone. These simulations lead to a local O₃ production rate between 0.5 and 1.77 ppb h⁻¹ in the MCS outflow and 0.17 ppb h⁻¹ outside the convection zone. These local production rates are likewise within the values reported by Schlager et al. (2009).

3.2 Biomass burning plumes

Figure 7 shows the flight tracks on the 4 and 13 August 2006. On both days a vertical profile was taken as the DLR- Falcon reached the coast in Ghana.

The measured RO₂^{*} vertical profiles, based on 20 s and 60 s data averages are shown in Fig. 8. The concentration of other trace gases, especially CO and CO₂ (Figs. 9 and 10) are indicative of the influence of a biomass burning plume at the lower pressure levels (570–650 mbar). At the highest pressure layer, the trace gases CO, CO₂, O₃,

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NO and NO_y reach 260, 379, 70, 0.160 and 2.8 ppbv, and 480, 390 133, 0.25 and 8 ppbv on the 4 and on the 13 August 2006, respectively. Interestingly, RO₂* follow the general pattern, being higher and less variable on the 13 August 2006 (20–25 pptv) than on the 4 August 2006 (5–15 pptv), in spite of the higher NO concentrations and the lower radiation indicated by the lower values of j_{NO₂}, but in agreement with the larger CO and HCHO mixing ratios, the latter reaching 0.6 and 1.16 ppbv on the 4 and on the 13 August 2006, respectively. The NO/NO_y ratio remains quite low in both cases (around 0.04–0.05) indicating aged air masses. However, HCHO and acetone have both primary and secondary sources and can be therefore produced during the oxidation of the BB plume and act as source of peroxy radicals.

The CO satellite pictures in August from the TERRA-MOPITT (<http://eosweb.larc.nasa.gov>; Mari et al., 2008) as well as the HCHO from the ENVISAT-SCIAMACHY (Reeves et al., 2009; Wittrock et al., 2006) instruments indicate clearly the BB region in the proximity of the Gulf of Guinea and between 5° N and 10° S latitude. Mari et al. (2008) distinguish three different periods during the wet season in Africa, affecting the intrusions of southern hemispheric fire plumes in the Northern Hemisphere. These are characterised by advection patterns related to differences in the position and strength of the African Easterly Jet. According to this classification, the 13 August is within the second active phase, characterised by the advection of BB plumes out over the Atlantic in the mid troposphere, while the 4 August corresponds to a break phase in which the pollutants emitted by the fires should be trapped over the continent and accumulate there till they reach convective regions located further north and are injected in the upper troposphere.

The vertical distribution of the trace gases measured on the 4 August is not as marked as on the 13 August which is less homogeneous and presents a clear layer between 500 and 650 mbar of higher concentrations and more photochemical activity. This agrees with the idea of an air mass which has been in more recent contact with BB emissions and is transported at a lower pressure levels to the North as suggested by Mari et al. (2008) during the active phase. And conversely, on the 4 August, as in-

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5 dicated by the CO_2 concentrations, the air mass sampled seems to have been largely mixed vertically, while the effect of the BB emissions have been lost in importance as the air mass aged, in spite of its potential further enrichment with new fire emissions during the trapping over the continent. This also matches the lower photochemical activity observed. The trajectories calculated in the present work confirm this interpretation. Figure 11 shows the projection of the corresponding trajectory densities onto the ground and the longitude-height projection as defined in Sect. 2.3. For each projection the trajectory density is normalised to unity. On the 4 August 2006, the air sampled
10 seems to originate from Nigeria in continental Africa, while at similar altitudes on the 13 August 2006 the air masses originate almost exclusively from the biomass burning area in Cameroon-Congo close to the Gulf of Guinea. According to these trajectories the air in the upper layers is transported from Central Africa.

3.3 Vertical distribution of RO_2^* over Ouagadougou

15 Figures 12 and 13 summarise the RO_2^* vertical profiles obtained on the 11, 13 and 15 August flights. RO_2^* up to 70 pptv with maximum mixing ratios between 400 and 700 mbar are observed. At a first glance the variability of the RO_2^* measurements seem to be large. However, a closer analysis shows that the variability is mostly explained by the presence of clouds, acting as a variable sink of radicals, and by the variability of other controlling trace species like NO_x , CO and VOC (Fig. 14). The former is the case
20 for the measurements taken on the 11 August at 445 and 570 mbar and on 13 August close to the surface, where the low and variable values of j_{NO_2} ($\cong 0.006 \text{ s}^{-1}$) are indicative of the presence of clouds. In addition, variations up to 20 ppbv O_3 , 60 ppbv CO and 0.8 ppbv NO_x are detected within the same pressure level in some of the flights. On the 15 August the NO_y at 920 mbar vary up to 20 ppbv NO_y . This indicates the existence of
25 various vertical layers of different composition and photochemical activity.

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3.4 Comparison with other measurements

Comparison with other aircraft measurements made during the campaign is possible on the 16 August 2006 flight, during which the DLR-Falcon and the BAe-146 flew close to each other at two different pressure levels. The flight is described more in detail by Reeves et al. (2009). A detailed quantitative comparison of the radical measurements is planned when the data of the PERCA onboard the BAe-146 become available. A preliminary comparison of the DUALER results with the HO₂ measured by LIF leads to a RO₂/HO₂ 60:40 ratio.

A quantitative comparison with literature results requires a deep analysis of the individual data concerning the chemical composition of air masses, photochemical conditions, characteristics of the different platforms and measurement techniques. It is normally difficult to acquire this detailed information, and even so, the comparison requires a normalisation which is complicated by the presence of too different parameters and variables. However, a simple qualitative comparison is of interest as it provides a first insight in the tropospheric vertical distribution and the range of values which can be expected under similar conditions. The data obtained within AMMA have been compared with the overview given in Cantrell et al. (2003a). These authors reported a general vertical pattern for various measurement campaigns, sorted by latitude and season, with a mid lower troposphere concentration maximum around 3–4 km. As can be seen in Fig. 13, this description is in agreement with the values obtained between 800 and 450 mbar in Ouagadougou and presented in Sect. 3.3.

4 Summary and conclusions

The total sum of peroxy radicals, RO₂^{*}, was successfully measured onboard of the DLR-Falcon instrumented aircraft during the West African monsoon period in summer 2006 by using a DUALER based on chemical amplification and a double reactor-detector system.

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Air masses having different photochemical histories have been investigated. RO_2^* mixing ratios of high variability and up to 50–60 pptv were observed in the outflow of MCS. The variability is partly associated with the presence of clouds, which block and reflect photochemically active ultraviolet radiation. Occasionally simultaneous unexpected increases of NO and RO_2^* are observed, indicating the presence of a radical precursor, which is related to NO emissions. As the NO mixing ratios measured during the period by other aircrafts in the boundary layer are generally lower (Reeves et al., 2009), the NO_x observed must be to a large fraction related to lightning episodes rather than being exclusively convective pumped to upper levels. Thunderstorms associated to MCS might also produce HO_x and radical precursors from VOC decomposition.

It can be concluded that the MCS outflow air masses detected during the measurement campaign are still photochemically active. Simulations using a box model with simplified chemistry indicate local O_3 production rates up to 2 ppb h^{-1} being consistent with other model simulations of MCS outflow within AMMA. The estimation of the global impact of MCS in the O_3 production requires further investigation.

The photochemistry of air masses affected by biomass burning plumes has also been investigated by means of two vertical profiles taken at the coast close to Ghana. The differences in chemical composition agree reasonably with the expected advection patterns related to the different phases of the African Easterly Jet. RO_2^* mixing ratios around 20 pptv and with peak values up to 60 pptv within 400 and 500 mbar are detected, indicating the presence of photochemical active layers.

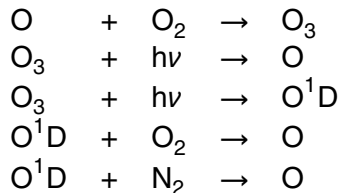
The final case study as part of this campaign was the investigation of vertical profiles measured over Ouagadougou. The data from the AMMA campaign yield a vertical distribution of RO_2^* having maximum mixing ratios between 450 and 700 mbar. This behaviour is similar to literature data taken under similar conditions and provides evidence for the enhanced free radical production in this pressure range. This presumably results from the uplifting of radical precursors from below.

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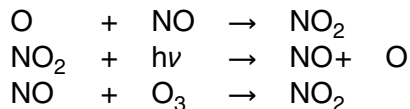
Appendix A

Chemistry of the box model

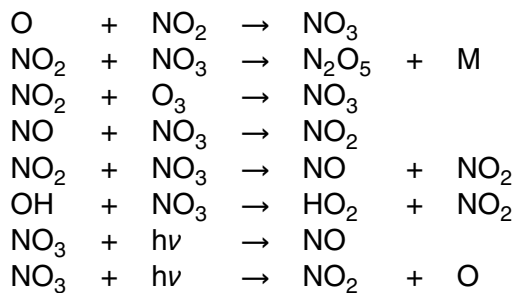
Ozone cycle



NO_x cycle



NO₃ production and decay



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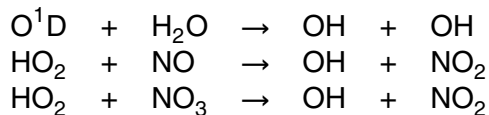
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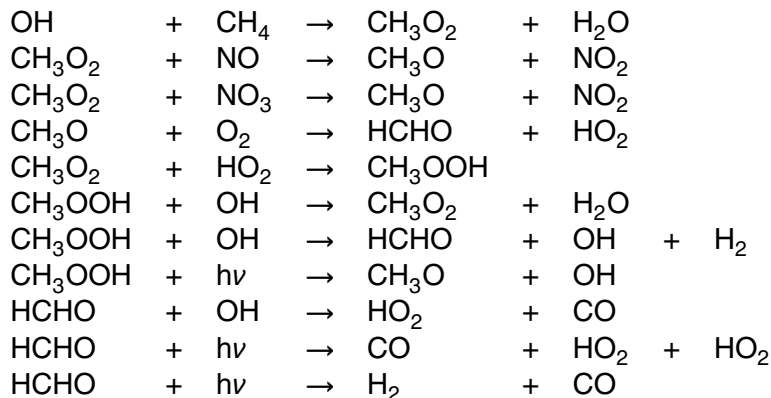
Hydroxyl radical



CO oxidation



Methane oxidation



All reaction rates are according to JPL-publication JPL 06-2 (Sander et al., 2006). The photolysis frequencies have been obtained using PhotoST (Burkert et al., 2003). PhotoST obtains the radiative fluxes from the radiative transfer model SCIATRAN (Rozanov et al., 2002), determines the actinic flux and performs the spectral integration of the product of the actinic flux, the absorption cross section and the quantum yield to derive the photolysis frequencies for all included photolytic reactions. For this study

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it is assumed the presence of cumulus clouds with a cloud top height well below the altitude of the aircraft.

Acknowledgements. We would like to thank all the DLR colleagues and the Falcon crew for their support before, during and after the AMMA measurement campaign. Special thanks also to T. Hamburger for providing maps with trajectories. The DLR-Falcon operation was funded in part by DLR and the European Commission.

5 Based on a French initiative, AMMA was built by an international scientific group and is currently funded by a large number of agencies. It has been the beneficiary of a major financial contribution from the European Community Sixth Framework Research Programme. Detail information on scientific coordination and funding is available on the AMMA International web site at <https://www.amma-eu.org/>.

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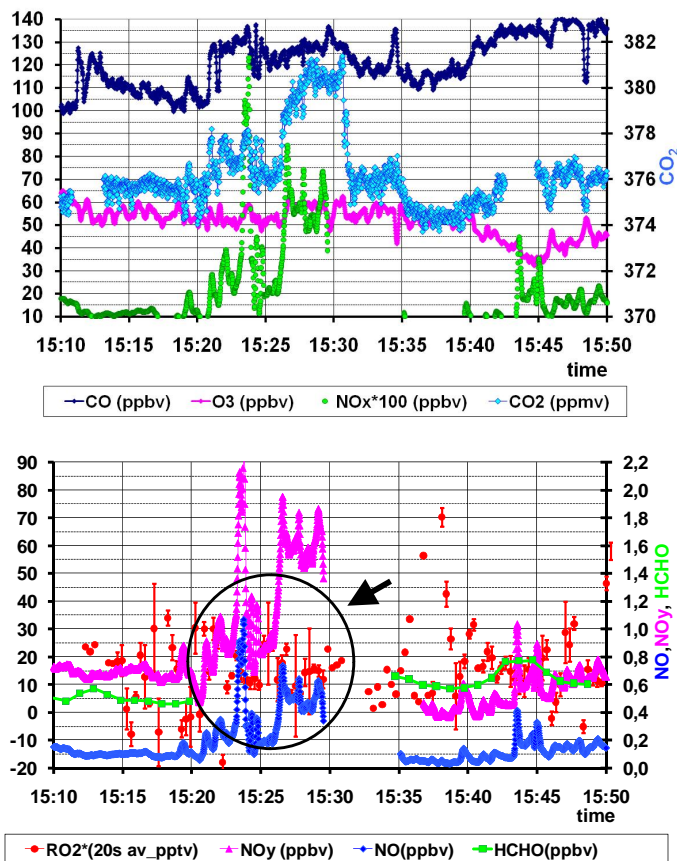
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Fig. 1. Trace gas measurements during crossing of a pollution plume during the DLR-Falcon flight carried out on the 15 August 2006. The measurements were taken at the 260 mbar pressure level. An episode of coincident RO₂* and NO variations is highlighted.

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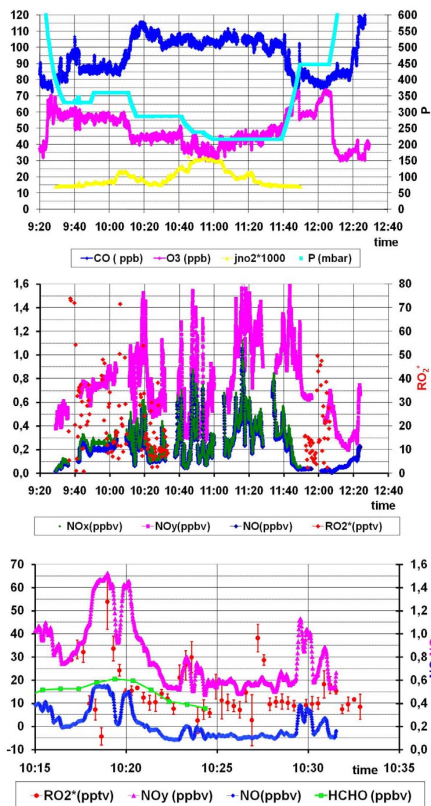
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Fig. 2. Trace gas mixing ratios measured during the DLR-Falcon flight on the 15 August 2006. The bottom plot shows in detail the RO₂* mixing ratios measured at 287 mbar. The depicted error bars only represent the statistical error of the 20 s RO₂* averages.

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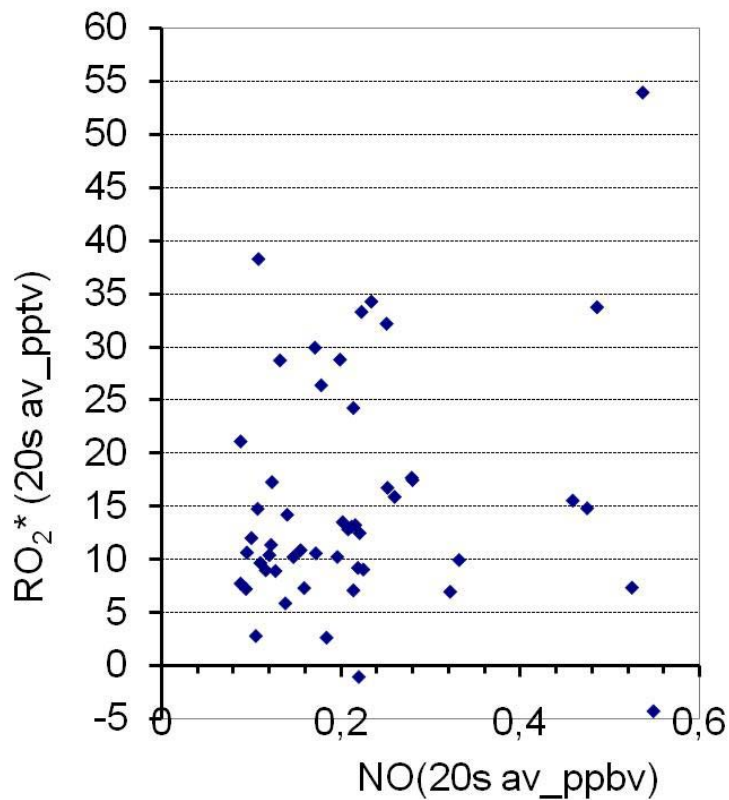
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Fig. 3. RO₂* and NO observed on the 15 August 2006 at the 287 mbar level.

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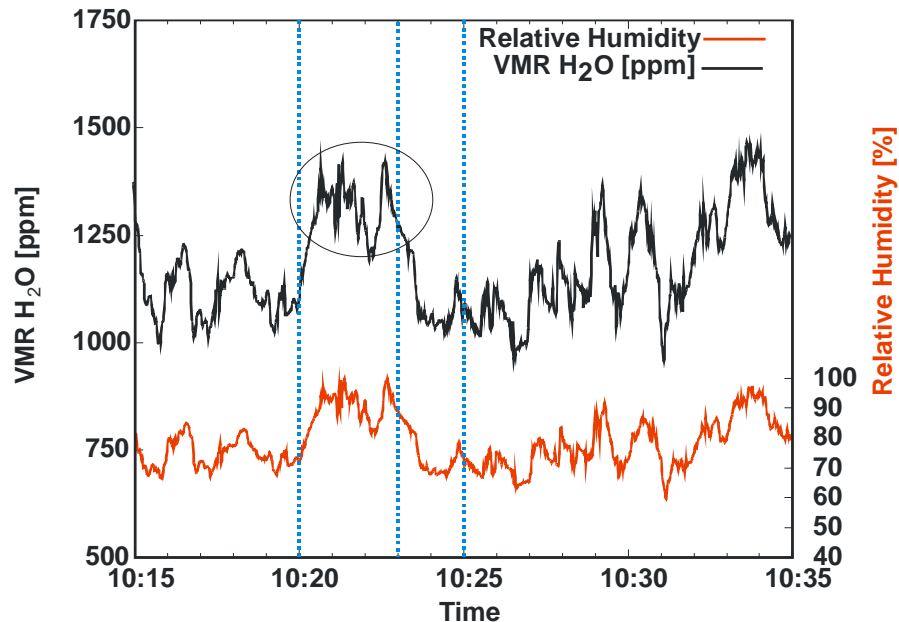
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Fig. 4. Water vapor observed on the 15 August 2006 at the 287 mbar level. The higher humidity values between 10:20 and 10:24 confirm the presence of air masses affected by convection. The time points selected for the box model initialisation are also highlighted.

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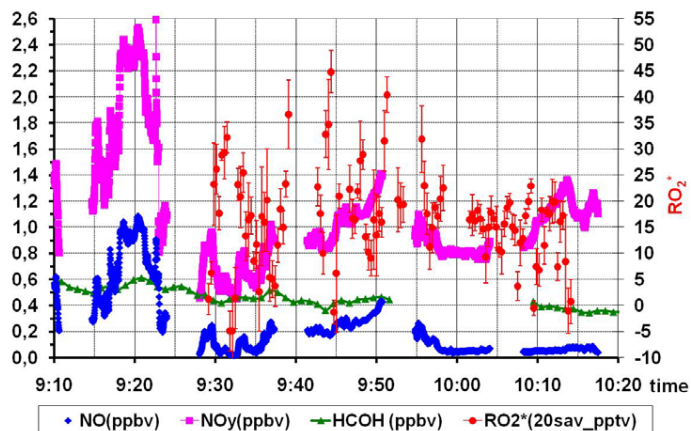
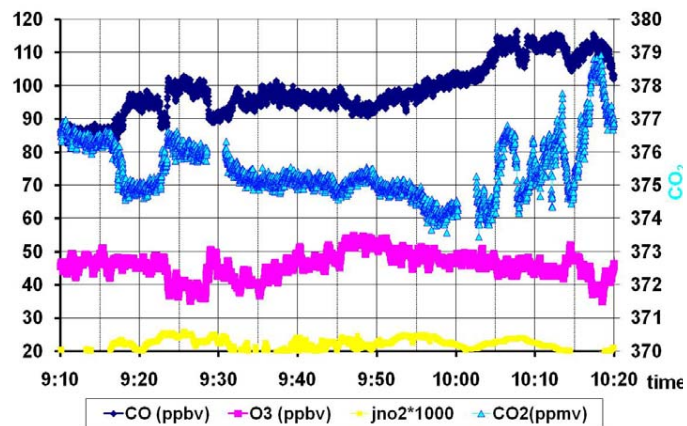
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Fig. 5. Trace gas mixing ratios measured during the DLR-Falcon flight on the 4 August 2006. The outflow of the MSC is reached at the 315 mbar pressure level (09:17 h). The depicted RO_2^* error bars represent only the statistical error of the 20 s averages.

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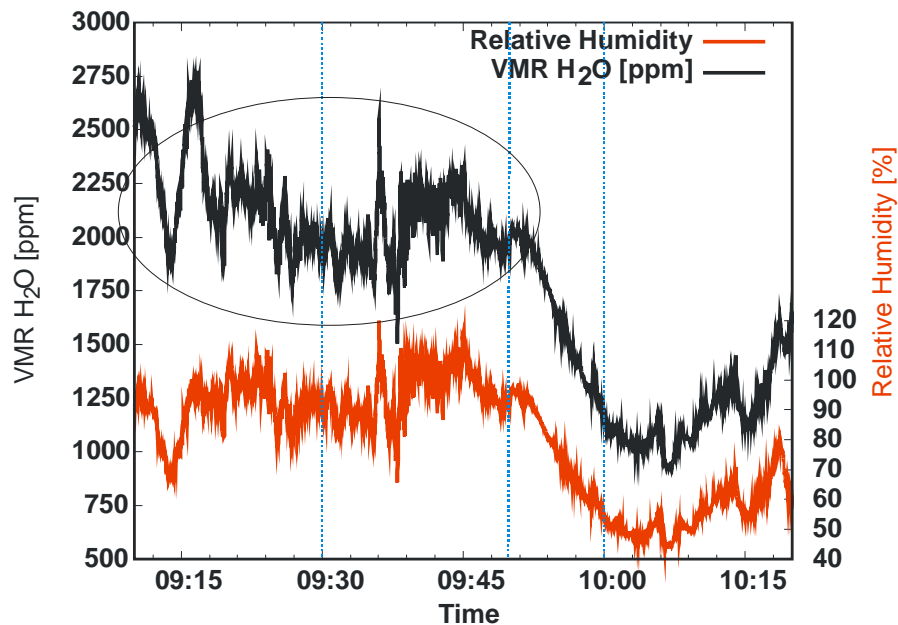
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Fig. 6. Water vapor measured during the DLR Falcon flight on the 4 August 2006. The convection zone is left at 09:50 h. Times selected for the box model initialisation are highlighted.

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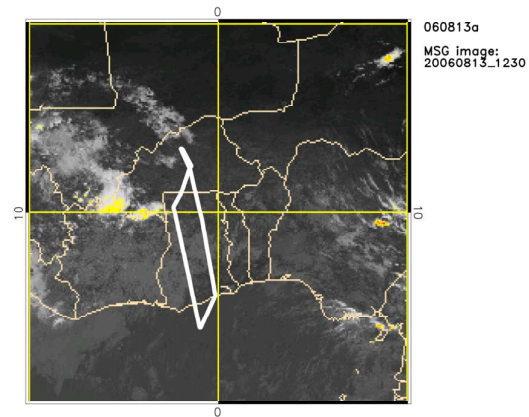
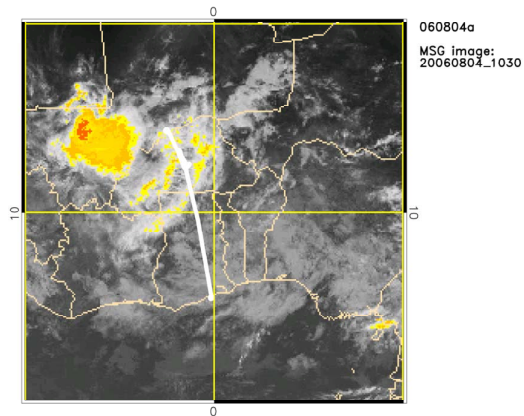


Fig. 7. DLR-Falcon flight tracks on the 4 and the 13 August 2006.

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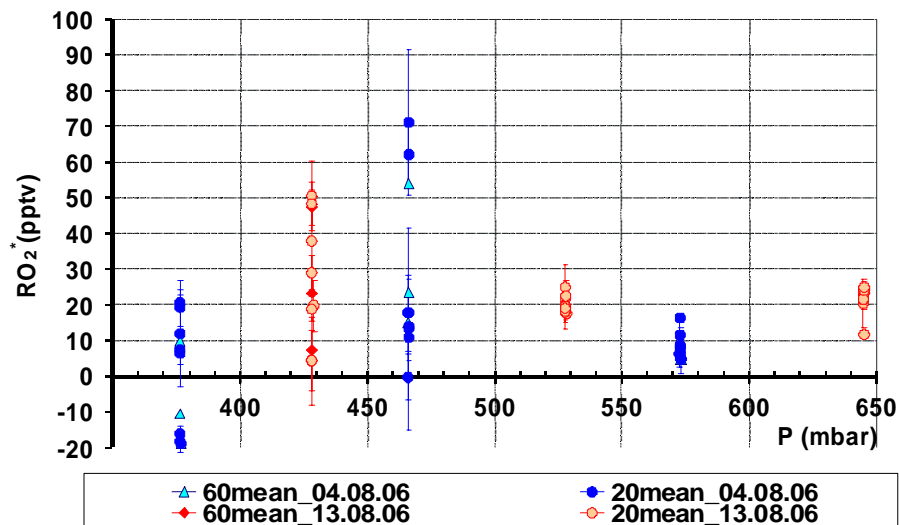
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Fig. 8. Peroxy radical mixing ratios measured on the 4 and 13 August 2006 coastal vertical profiles. The statistical error of the corresponding 60 s and 20 s averages is depicted.

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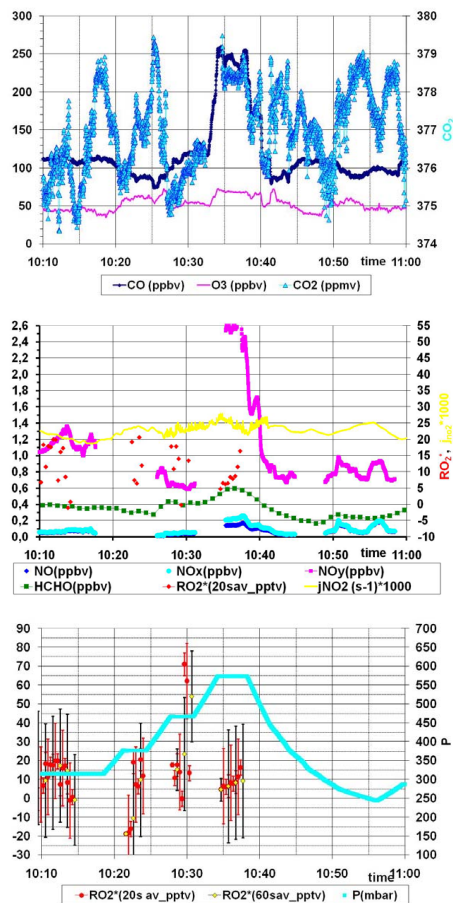
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Fig. 9. Trace gas mixing ratios measured during the coastal vertical profile on the 4 August 2006. The statistical error of the 20 s and 60 s RO₂* averages is also depicted.

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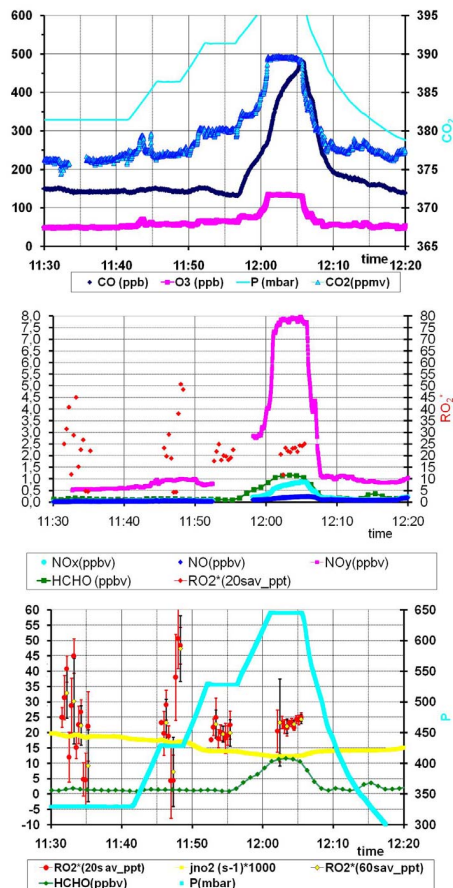
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Fig. 10. Trace gas mixing ratios measured during the coastal vertical profile on the 13 August 2006. The statistical error of the 20 s and 60 s RO₂* averages is also depicted.

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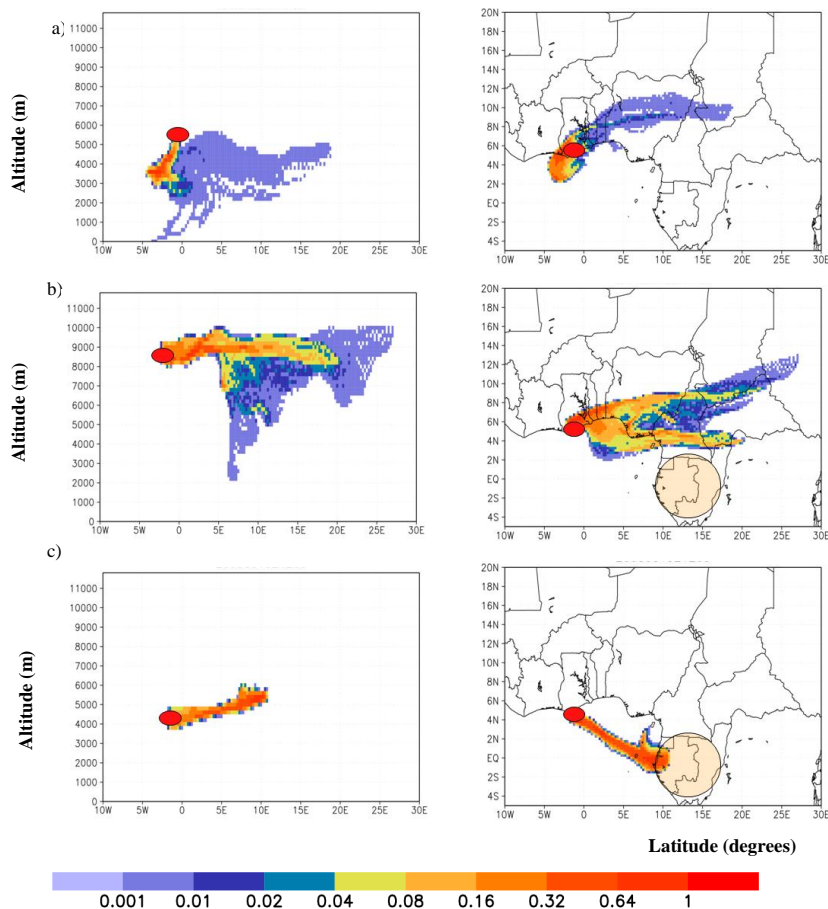
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Fig. 11. Trajectory densities calculated for the vertical profile measured on the **(a)** 4 August 2006 at 10:30 h, **(b)** 13 August 2006 at 11:40 h and **(c)** 13 August 2006 at 12:05 h. The BB area according to satellite data is highlighted.

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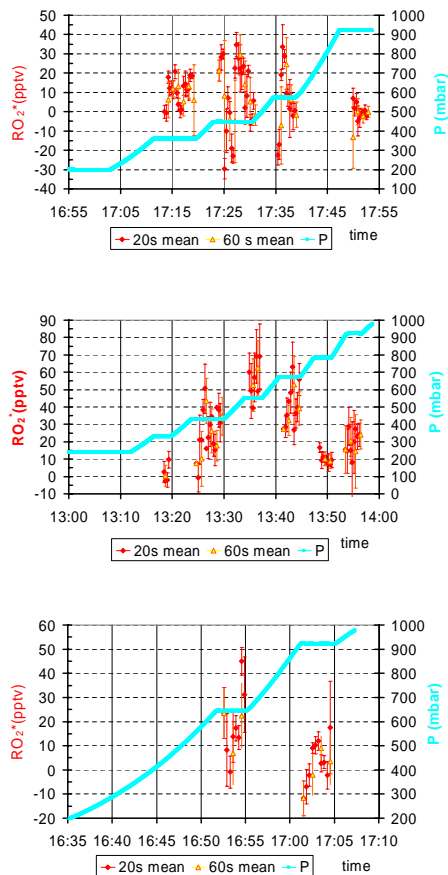
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Fig. 12. RO_2^* mixing ratios measured in vertical profiles taken over Ouagadougou on the (a) 11 August 2006, (b) 13 August 2006 and (c) 15 August 2006.

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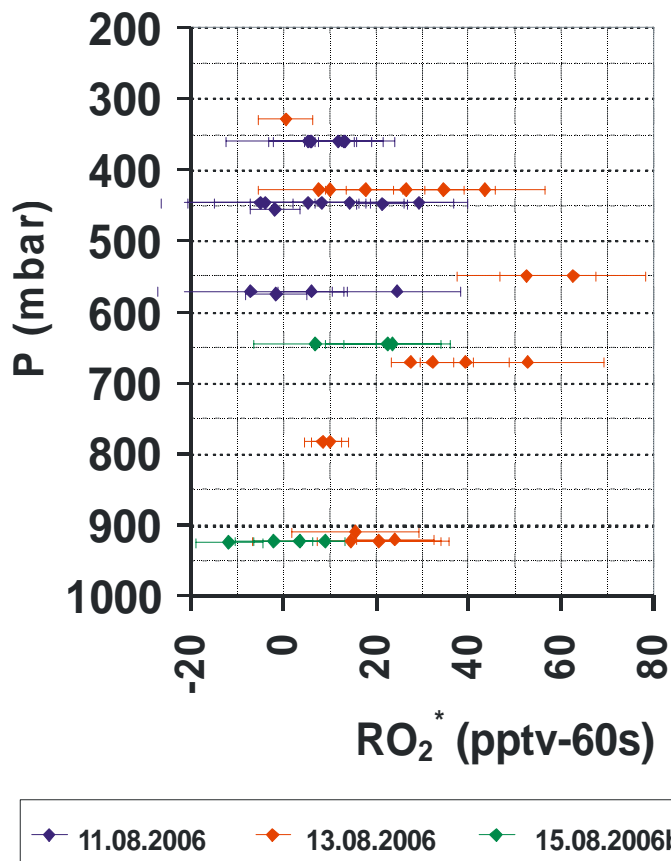
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Fig. 13. Vertical profiles of RO₂^{*} mixing ratios measured over Ouagadougou. The 60 s mean values and the corresponding error bar are depicted. 15 August 2006b refers to the data measured on the flight in the afternoon.

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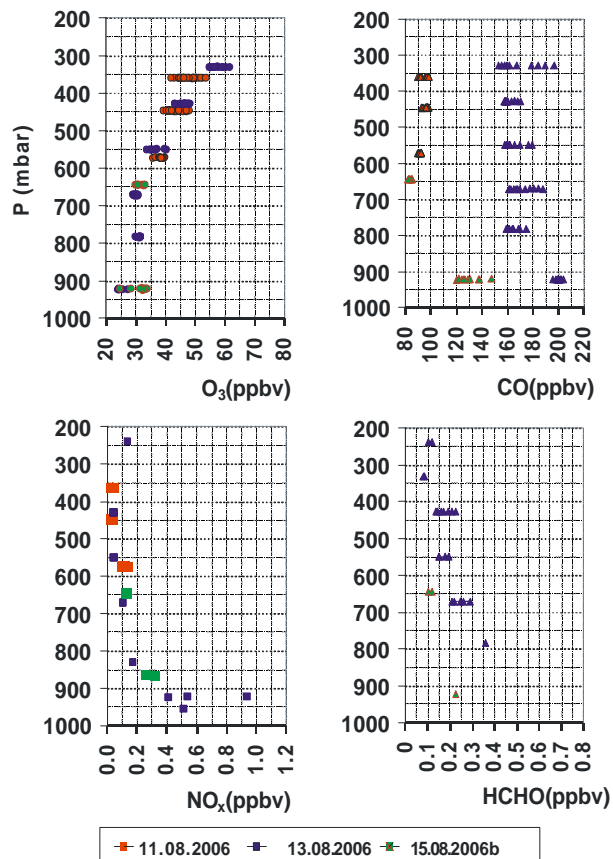
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Fig. 14. Trace gas mixing ratios measured over Ouagadougou on the 11 August 2006, 13 and 15 August 2006. The 15 August 2006b refers to the data of the afternoon flight.

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