Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-926-AC1, 2019

Atmospheric Chemistry and Physics
Open Access
Discussions

Interactive comment on “Upper tropospheric CH_4 and CO affected by the South Asian summer monsoon during OMO” *by* Laura Tomsche et al.

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Received and published: 17 January 2019

-Reviewer 1: The EMAC model, on the other hand, has a much coarser resolution of 2.8x2.8 degrees and 90 levels. Unfortunately the EMAC simulation used is not described in detail, leaving open some important questions: Is EMAC used in an offline CTM mode? If this is the case, what is the model then driven by? Or, in other words: Do both models, the Lagrangian as well as the Eulerian model “see” the same background atmosphere? What kind of vertical velocity was used for the EMAC simulation? Another important point of course would be the initialization of the model, the length of the simulation and whether a certain spin-up time was necessary. Since during the analysis of the data results from both models were used simultaneously (e.g. footprints and emission data) or observations of tracers obviously transported upward by convection are compared to distributions modified by vertical transport in EMAC, a more detailed description of the model setup would be very helpful. A very interesting diagnostic in this context would e.g. be the vertical transport time of tracers emitted from the surface to reach the 200 hPa level in EMAC.

Author: We thank the reviewer for pointing out the lack of information regarding the model simulation. Here additional details are given, also added to manuscript.

Authors changes in manuscript: P5-6 L30-9: The EMAC model was not run in an offline CTM mode, as the radiation calculations were based on simulated GHGs concentrations. Nevertheless, the model was weakly nudged towards ECMWF ERA Interim data (Jeuken et al., 1996) and therefore reproduced very similar dynamics to the ECMWF model (although not binary identical). The simulation is an extension of simulation RC1SD-base-10 (Jöckel et al. 2016) so to cover the full OMO campaign. Few changes to the original simulation have been applied (i.e. increased South Asia SO2 emissions and reduced lightning NOx), as described in Lelieveld et al. (2018). Although the simulation is the continuation of a well evaluated experiment, the simulation was running from March 1st, 2015 so to give time to the SO2 and NOx to balance to the new emissions (i.e. 4 months spin up time). Only the data from July and August 2015, which covers the field campaign is actually used. The EMAC model is a hydrostatic model and the convective transport is parameterized (Ouwersloot et al. 2015, Tost et al. 2006). Indication of the vertical transport time in EMAC can be found in Krol et al. (2018), where also a comparison with model of similar complexity is shown.

-Reviewer 1: The derivation of threshold values for CO and CH4 to distinguish between the inside of the monsoon anticyclone and the outside by using vertical profiles for NH and SH background and AMA leads to the question, why profiles over Egypt are considered as influenced by AMA and profiles over Cyprus are not. At least a look at the figures showing the different AMA modes (figures 18 to 21) would lead to a different expectation. But this is just judged by visual measure (and only on 204 hPa), so if there are distinct differences between profiles at these locations, the authors would be well advised to please show them. Since the classification of profiles influences the threshold values, this question may be quite important for the further analyses.

Author: In the classification of the profiles used for the calculation of the Northern hemisphere background and AMA-influenced air masses, respectively, and not only the geographical location but also the meteorological context have been accounted for. The profiles over Egypt were sampled during the second double anticyclone mode, with the westerly part of the anticyclone extending over Egypt. Profiles over Paphos were obtained over a longer period, representing background conditions but partly also AMA-influenced air masses. We calculated the NH background with and without profiles over Paphos. For profiles only over Oberpaffenhofen and Etna the average CH4 mixing ratio is 1871.2±9.2 ppbv and for profiles over Oberpfafffenhofen, Etna, and Paphos the CH4 average is 1863.4±14.0 ppbv. Thus the profiles with and without Paphos profiles agree within their standard deviation. Due to a better statistics, we used the NH background profile including profiles over Paphos.,

Authors changes in manuscript: P7 L5-7: As observed, the CO and CH4 profiles measured during OMO indicate different altitude distributions depending on the geographical location and partly also on the meteorological situation, especially for Paphos and Egypt. Profiles over Egypt were measured when the AMA extended over this region. Profiles over Paphos were sampled during periods with and without the AMA being positioned over Cyprus.

-Reviewer 1: The observations shown for the case study for flight 19 indicate a highly structured CO and CH4 distribution in the vicinity of the AMA boundary region. The distributions simulated by EMAC matches the observations only very roughly. In particular the CH4 values are underestimated significantly. By looking at the horizontal and vertical distributions one gets the impression that the vertical transport of the model is probably too weak. This may have several reasons: First, the vertical velocity may be too slow, e. g. the processes leading to strong updraft (namely convection) are too weak or insufficiently parameterized, or second, the numerical horizontal diffusion implied by the coarse grid resolution dampens the strong updraft plumes (approximately above 500 K). Adding horizontal wind as contour lines to the cross sections could shed some light on this problem. The included lines of potential temperature already point into this direction.

Author: Indeed, the referee is correct in mentioning a possible too low transport of methane and carbon monoxide as a reason for underestimation in the upper troposphere. As shown by Krol et al. (2018), EMAC seems to have a weaker transport of surface tracers than other models. Both reasons suggested by the referee are possible, and it is difficult (if not impossible) to really distinguish the real reason for the underestimation of the transport. Nevertheless we would like to notice that for the comparison of CO with the model, the results are in line with other literature studies at such resolution (e.g. Baret et al., 2016). Horizontal wind components are added to the cross sections in figures 9-12, in detail: eastward wind component in cross sections along a longitude and northward wind component in cross sections along a latitude.

Authors changes in manuscript: P11 L13-18: The simulated CO pattern, especially the enhanced values over Oman, fits well to the observed CO mixing ratios along the flight track. The EMAC model underestimates CH4 and CO in the upper troposphere. As shown by Krol et al. (2018), EMAC seems to have a weaker transport of surface tracers than other models. There are two potential reasons for that, but it is difficult to distinguish them. First, a too slow vertical velocity, thus the convective updraft is too ineffective, or second, the numerical diffusion implied by the coarse resolution restricts the updraft too strong. Nevertheless we would like to notice that for the comparison of CO with the model, the results are in line with other literature studies at such resolution (e.g. Baret et al., 2016). Horizontal wind components are added to the cross sections in Figures 9-12 (P35-41)

-Reviewer 1: However, although the EMAC distributions may be consistent within the model, these effects may lead to a too small AMA region, when defined by an observational-based CH4 threshold. A dynamical shape of the AMA could be gained by using geopotential height or stream function. In this context I would suggest to add some contour lines to the figures displaying the horizontal CO and CH4 distribution including the threshold values and lower values to give a better visual feedback of the AMA and its position relatively to the flight tracks.

Author: In Figures 7,8, 18-21 and also in Figures 22 and 23 now contour lines are added for the CH4 threshold (1879.8 ppbv) and the CH4 background (1859.4 ppbv) values according to the calculation of the CH4 threshold in section 3.1. In the horizontal CO distribution also the CH4 threshold is added. Now the position of the AMA is easier to identify with respect to the flight tracks.

Authors changes in manuscript: Figures 7,8,(P31-32), 18-21 (P42-45), 22 (P47),23 (P48)

-Reviewer 1: A comparison between footprints of last PBL contact derived from 10 day backward trajectories from FLEXPART and the surface emissions from EMAC could be much more efficient, when footprints would be graphically added to the surface emission charts.

Author: Footprint is now added as white contour lines for the number of particles per grid cell = 2 to the surface emission charts for CH4 and CO (Figures14 and 15).

Authors changes in manuscript: Figure 14,15 (P38 and P 39)

-Reviewer 1: The analysis with respect to the different AMA modes defined by the CH4 distribution of the EMAC simulation leads to very interesting results, which are almost impossible to interpret from the values of table 2 without the knowledge of the flight tracks and the position of the AMA. Probably one could use the distance not to the anticyclonic centers but to the boundaries of the anticyclones.

Author: In Table 2 we add a column for the relative position to the AMA, which is quite descriptive. As most of the flight tracks are in and outside the AMA a more detailed geographical location with respect to the AMA can be realized better in a graphical

way. Thus we added for each flight in the supplement the CH4 threshold (1879.8ppbv) for the AMA-influence and the background value (1859.4ppbv) as contour lines in the EMAC CH4 and CO distributions as already done in the manuscript, e.g. Figure 7 and 8 for flight 19. In these plots the position of the flight track with respect to the AMA is more obvious.

Authors changes in manuscript: Column added in table 2.P46

-Reviewer 1: The last case study focusing on an outflow event tracked with trajectories and probed twice within 4 days seem to give better agreement with EMAC results (again only judged by visual measure). Maybe an additional figure showing observed and simulated tracer distributions would complement this very interesting manuscript.

Author: According to the suggestion of the reviewer the CO and CH4 distributions along the flight track as a time line are added in Figures 24 and 25. The trace gas mixing ratios (observed and simulated) show clear enhancement due to the outflow event for both flights (flight 12/13 and flight 17/18). The outflow regions are marked in grey in the Figures. Additionally, we add in the manuscript the average CO and CH4 mixing ratios calculated from EMAC for the outflow periods for both flights (flight 12/13: CO=112.2±1.2 ppbv and CH4=1891.7±1.2 ppbv and flight 17/18: CO=90.8±3.1 ppbv and CH4=1864.6±5.9 ppbv) for a better comparison with the measured data in the outflow.

Authors changes in manuscript: Figures 24 and 25 added to manuscript P49 P17 L18-22: In the air mass CO and CH4 mixing ratios increased to 117.3±22.2 ppbv and 1893.5±9.8 ppbv, respectively (background: CO=78.6±33.3 ppbv and CH4=1827.4±26.8 ppbv), which can be seen in Figure 24. The second probing of this air mass took place at August 10 (flight 17/18, Figure 23) over the Red Sea yielding mixing ratios of 94.2±6.8 ppbv and 1903.7±19.2 ppbv. This corresponds to the increase at around 12-13 UTC in Figure 25.

P17 L25-28: Comparing the EMAC simulations with the in situ data along the flight tracks (Figure 24 and 25), the trends for the outflow agree. The EMAC average mixing ratios for CO and CH4 are 112.2±1.2 ppbv and CH4=1891.7±1.2 ppbv for flight 12/13 and 90.8±3.1 ppbv and CH4=1864.6±5.9 ppbv for flight 17/18, respectively. Thus also the values agree within their standard deviation beside CH4 in flight 17/18, where the outflow is underestimated by the model.

-Reviewer 2: The goal of this study is not clearly stated. Is it to explore transport pathways inside the anticyclone or in the vicinity? For instance, flight 19 suggests the measurements took place outside the anticyclone based on the boundaries estimated from the model simulations (Fig. 7 & 8). I think it is important to clarify the goal of this study and explain different transport pathways separately.

Author: The goal of the present study is to understand the transport pathways from the source regions into the upper troposphere via the convective uplift into the AMA and further within the UT, especially towards the southern and western areas of the AMA. The transport pathways in the UT include the transport along the edges of the AMA, the circulation in the AMA where air masses are trapped and the transport across the AMA edges, and the outflow out of the anticyclone due to instabilities in the strong circulation. For instance, flight 19 took place outside and at the western edge of the AMA, which is now better visible in Figures 7 and 8 due to addition of a contour line for the CH4 threshold.

Authors changes in manuscript: Adapted Figures 7 and 8 P2-3 L32-3: The measurement campaign OMO (Oxidation Mechanism Observations) took place in July/August 2015 with the German High Altitude and Long range (HALO) research Aircraft, performing flights at altitudes between 11 km and 15 km over the above-mentioned regions to investigate the dynamics and atmospheric chemistry in the upper troposphere over five weeks during the monsoon season.

-Reviewer 2: In introduction, brief background of the Asian monsoon anticyclone and its role in chemical transport in the UTLS region should be mentioned first. Then why

in-situ measurements are so valuable but challenging and limited should be mentioned along with pros and cons of other data sources, including, satellite measurements. The purpose of utilizing two separate models should be emphasized. The key factors of OMO field campaign should be included with proper citations as well. Additionally, the goal of this paper and why this paper is unique compared to previous work should be mentioned clearly.

Author: The introduction is reorganized according to the suggestions of the reviewer. We used the EMAC model simulations to extend our view on trace gas distributions from the regional scale along flight tracks to a global scale, i.e. horizontal and vertical trace gas distributions, and also to separate different AMA modes. With the second model (FLEXPART) we calculated back trajectories to investigate the emission sources and the transport pathways from the source regions, via the convection into the AMA in the upper troposphere and further westward towards the flight tracks. Thus the back trajectories are mainly used for dynamical processes.

Authors changes in manuscript: P1-3 L27-21: The Asian monsoon anticyclone (AMA) is an annual, large-scale weather phenomenon in the upper troposphere and lower stratosphere during the boreal summer. It is enclosed by the westerly subtropical jet in the north and the easterly jet in the south and extends over southern Asia and the Middle East up to the Mediterranean. It is formed by diabatic heating in the South Asian monsoon region (Gill, 1980, Hoskins and Rodwell, 1995). The anticyclone is a strong and nearly closed circulation system, which is variable in strength and location (Hsu and Plumb, 2000, Popovic and Plumb, 2001, Garny and Randel, 2013, Ploeger et al., 2015). The strong winds at its edges act as transport barrier for chemical constituents in the upper troposphere. Stratospheric tracers, like ozone, show generally lower concentrations inside the AMA than outside (Park et al., 2008, Randel and Park, 2006). Tropospheric tracers, like CO and CH4, are uplifted to the upper troposphere by the strong monsoon convection. These chemical constituents can be trapped in the anticyclone, change the atmospheric chemistry in the upper troposphere and lower stratosphere and clearly signify the monsoon influence (Park et al., 2007). The signature of the anticyclone has been identified from different measurement platforms, like satellites and aircrafts. Airborne measurements are rare and limited in time and space but resolve small scales. For example, the in-service airborne projects CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container; e.g. Schuck et al., 2012, Rauthe-Schöch et al., 2016) and IAGOS-MOZAIC (IAGOS (In-service Aircraft for a Global Observing System) and MOZAIC (Measurements of OZone by Airbus In-service aircraft); Barret et al., 2016, Dethof et al., 1999) reported trace gas measurements in the Asian monsoon region. In addition aircraft campaigns investigated the Asian monsoon during the aircraft campaign MINOS (Lelieveld et al., 2002, Scheeren et al., 2003) and the Earth System Model Validation (ESMVal) campaign (Gottschaldt et al., 2017). In contrast, satellite data cover a larger spatial area and can be used for long term measurements, nevertheless they are limited to their overpassing track and they have a coarse resolution. The obscured view from clouds during the South Asian monsoon additionally restricts the satellite view (e. g. Ojha et al., 2016), which requires long-term averaging in time and should be complemented by in situ measurements. Satellite data for different trace gases, like H2O (Park et al., 2004, Randel and Park, 2006), CO (Li et al., 2005, Park et al, 2008) and CH4 (Park et al., 2004), show the vertical and horizontal extension of the AMA and are generally in agreement with model simulations (e.g. Pan et al., 2016, Nützel et al., 2016, Bergman et al., 2013). To improve model outputs and satellite data retrievals, airborne measurements are necessary, A more physically motivated criterion to distinguish between the AMA and its surrounding in the upper troposphere is the potential vorticity (PV) (e.g. Ploeger et al., 2015, Garny and Randel, 2013). In the anticyclone PV values on isentropic surfaces are lower than outside. Therefore, a maximum in the PV gradient can be used to identify the horizontal transport barrier associated with the AMA. However, applying the PV criterion is not straightforward since PV values in the AMA increase during the monsoon season and decrease from the extra-tropics towards the tropics, which limits its usefulness. Nevertheless, it is quite helpful in combination with trace

gas distributions from in situ and satellite measurements. During the aircraft campaign MINOS the outflow of the AMA was investigated over the eastern Mediterranean basin (Lelieveld et al., 2002, Scheeren et al., 2003), while during the Earth System Model Validation (ESMVal) campaign a single flight was performed from Male/Maldives to Larnaca/Cyprus in September 2012 that intersected the AMA at an altitude of 150 hPa (Gottschaldt et al., 2017). In situ airborne measurements in the region of the Mediterranean, the Arabian Peninsula, and the Arabian Sea during the monsoon season are still limited, even though the AMA impacts these regions either by its extension or via outflow. Here we present results from an aircraft mission, which focuses on the AMA between the Indian Ocean and the Mediterranean. The measurement campaign OMO (Oxidation Mechanism Observations) took place in July/August 2015 with the High Altitude and Long range (HALO) research Aircraft, performing flights at altitudes between 11 km and 15 km over the above-mentioned regions to investigate the dynamics and atmospheric chemistry in the upper troposphere over five weeks during the monsoon season. The present study focuses on the measurements of CH4 and CO, which document long-distance transport of air pollution, as these species have extended lifetimes of 8-9 years (CH4, Lelieveld et al., 2016) and 2-3 months (CO, Xiao et al., 2007). These trace gases can be used to identify emission sources from the surface as they are co-emitted with other pollutants. They have both natural and anthropogenic sources. Major CO sources are anthropogenic and emitted via combustion processes of fossil fuel, biomass, and domestic fuel. Its natural sources are mainly from vegetation and oceans, but they are minor (Pandis and Seinfeld, 2006). CH4 is also emitted by combustion of fossil fuel and biomass (Khalil, 2000). Further sources are rice cultivation and ruminants, but also swamps and flood areas. For wetlands, the uncertainty in CH4 emissions is still a large concern in atmospheric chemical transport models (Bloom et al., 2017, and references there in). In South Asia anthropogenic emissions increase with a growing population and economic development (Rauthe-Schöch et al., 2016, Ohara et al., 2007). The observations of CH4 and CO show zonal and meridional concentration gradients as well as vertical gradients in the upper troposphere, allowing to investigate the extent of the AMA. In order to differentiate background from AMA influenced air masses, we derived a CH4 based threshold. Further, we compared our observations with EMAC model simulations, which extend the view on the trace gas distribution from a regional (along the flight tracks) to a global scale. To study the transport pathways we calculated back trajectories with the Lagrangian particle dispersion model FLEXPART along the flight tracks. With FLEXPART we gained a more detailed insight into the dynamics. We compared the back trajectories with observations of CH4 and CO to distinguish between different transport pathways. Thus we also studied the origin of emissions within South Asia. Finally, we investigated the variability of the AMA over several weeks as the anticyclone changes its position, extent, and strength due to the monsoon dynamics.P3 L15-21: Further, we compared our observations with EMAC model simulations, which extend the view on the trace gas distribution from a regional (along the flight tracks) to a global scale. To study the transport pathways we calculated back trajectories with the Lagrangian particle dispersion model FLEXPART along the flight tracks. With FLEXPART we gained a more detailed insight into the dynamics. We compared the back trajectories with observations of CH4 and CO to distinguish between different transport pathways. Thus we also studied the origin of emissions within South Asia. Finally, we investigated the variability of the AMA over several weeks as the anticyclone changes its position, extent, and strength due to the monsoon dynamics.

-Reviewer2: Abstract of this paper should be a summary of what is shown in this work without including general statements. In the current form, most of the information exists without clearly stating what the goal of this paper is.

Author: The abstract is revised.

Authors changes in manuscript: P1 L6-22: The Asian monsoon anticyclone (AMA) is a yearly recurring phenomenon in the northern hemispheric upper troposphere and lower stratosphere. It is part of the South Asian summer monsoon system, and it has a clearly observable signature due to vertical transport of polluted air masses from the

surface to the upper troposphere by the monsoon convection. We performed in situ measurements of carbon monoxide (CO) and methane (CH4) in the region of monsoon outflow and in background air in the upper troposphere (Mediterranean, Arabian Peninsula, Arabian Sea) by optical absorption spectroscopy on board the High Altitude and Long range (HALO) research aircraft during the OMO (Oxidation Mechanism Observations) mission in summer 2015. We identified the transport pathways and the origin of the trace gases with back trajectories, calculated with the Lagrangian particle dispersion model FLEXPART, and we compared the in situ data with simulations of the atmospheric chemistry general circulation model EMAC. CH4 and CO mixing ratios were found to be enhanced within the AMA, the in situ data increased on average by 72.1 ppbv and 20.1 ppbv, respectively, originating in the South Asian region (Indio-Gangetic Plain, North East India, Bangladesh and Bay of Bengal). It appears that CH4 is an ideal monsoon tracer in the upper troposphere due to its extended lifetime and the strong South Asian emissions. Furthermore, we used the measurements and model results to study the dynamics of the AMA over several weeks during the monsoon season, with an emphasis on the southern and western areas in the upper troposphere. We distinguished four AMA modes based on different meteorological conditions. During one occasion we observed that under the influence of dwindling flow the transport barrier between the anticyclone and its surroundings weakened, expelling air masses from the AMA. The trace gases exhibited a distinct fingerprint of the AMA, and we also found that CH4 accumulated over the course of the OMO campaign.

-Reviewer2: Section 2 (methods) should include general information about OMO field campaign, including its science goal. What other species were measured during the campaign? What were the science questions? Are there any references?

Author: General information about the OMO mission are added in the manuscript in the method part in section 2.1 including references.

Authors changes in manuscript: P3-4 L23-7: The Oxidation Mechanism Observation (OMO) aircraft measurement campaign focused on the self-cleaning capacity of the atmosphere in connection with the Indian summer monsoon. The mission took place in July and August 2015 with flight tracks in the upper troposphere (10-15 km) over the Mediterranean, the Arabian Peninsula, and the Indian Ocean (Figure 1). In South Asia the pollution emissions are growing and during the monsoon season they are uplifted into the upper troposphere. The pollution is partly removed by wet deposition or transformation into soluble gases, or they are involved in air chemistry and transported downwind of the sources. For a broad analysis of the efficiency of the self-cleaning mechanism a large variety of chemical compounds, like CH4, CO, OH, HO2, NOy, SO2, RO2, H2O2, and total peroxides, were measured during the multi-institutional campaign, involving the Max-Planck-Institute for Chemistry, Mainz, the Research Centre Jülich, the German Aerospace Center, the Research Centre Karlsruhe, and the universities of Bremen, Heidelberg, and Wuppertal. The main objectives were the oxidation processes and free radical chemistry, the efficiency of convective cloud transport and wet deposition, as well as long-distance transport of air pollution and impacts on air quality and climate change. The OMO mission comprised 111 flight hours during 17 flights. HALO was based alternately at Paphos (Cyprus) and on Gan (Maldives) with refueling stops at the airport of Bahrain. Further information about OMO can be found in Lelieveld et al. (2018) and on the webpage http://www.halo.dlr.de/science/missions/omo/omo.html.

-Reviewer 2: Section 3.5 (AMA mode) should include discussions of bimodal mode of the monsoon anticyclone shown in Zhang et al. (2002) and Nützel et al. (2016). Also, it should be justified why it is necessary to have four modes instead of two. Is bimodal distribution of the anticyclone wrong?

Author: A short discussion about bimodality of the AMA is now added I section 3.5.

Authors changes in manuscript: P15 L8-17: Zhang et al. (2002) presented a bimodality of the AMA with a center position of the anticyclone over the Iranian or the Tibetan Plateau. During OMO we found both positions, which in line with the bimodality assumption. In contrast, Nützel et al. (2016) reported different center positions of the

AMA in several models, but most of them did not simulate a preferred bimodality. Regarding the eastern anticyclones during the double anticyclones modes, the positions were in-between the Iranian and Tibetan Plateau (first mode) and in the fourth mode over the Tibetan Plateau. Consequently, they do not support a preferred bimodality. In Zhang et al. (2002) and Nützel et al. (2016) the Iranian and the Tibetan mode are further distinguished by parameters, like diabatic heating, rain patterns or areas of convection, which are out of scope in the present study. Here the focus is on the dynamics with respect to the trace gas distributions. The subdivision into four modes represents the dynamics of the AMA over the course of the campaign.

-Reviewer 2: The abstract includes a few general statements, which makes abstract sound rather like introduction. For instance, L9-11 (However: : :expected) can be removed.

Author: L9-11 Sentence is removed.

Authors changes in manuscript: P1, L9-11

-Reviewer 2: P1, L7 – It is connected to -> It is part of the South Asian summer monsoon system Author: This has been changed.

Authors changes in manuscript: P1, L7: It is part of the South Asian summer monsoon system

-Reviewer 2: P1, L17-19 – Are those based on the in-situ measurements?

Author: Yes, these values are representing the in situ data, but also the simulated data show increased mixing ratios with AMA-influence, as mentioned in section 3.4 The AMA during OMO. In situ increase 72.1 ppbv and 20.1 ppbv and EMAC increase 24.0 ppbv and 14.7 ppbv for CH4 and CO, respectively.

Authors changes in manuscript: P1, L14-15: the in situ data increased on average by 72.1 ppbv and 20.1 ppbv, respectively,

-Reviewer 2: P1, L21 – areas within the upper troposphere -> areas in the upper troposphere

Author: This has been changed.

Authors changes in manuscript: P1, L19: areas in the upper troposphere

-Reviewer 2: P2, L3 – Park et al. (2008) might be relevant here.

Author: The reference has been added as it is relevant here.

Authors changes in manuscript: P2, L2: Stratospheric tracers, like ozone, show generally lower concentrations inside the AMA than outside (Park et al., 2008, Randel and Park, 2006).

-Reviewer 2: P2, L4 – within the strong: : :monsoon -> by the strong monsoon convection

Author: This has been changed.

Authors changes in manuscript: P2, L3-: to the upper troposphere by the strong monsoon convection.

-Reviewer 2: P2, L5 – Park et al. (2007) might be relevant here.

Author: The reference has been added as it is relevant here.

Authors changes in manuscript: P2,L5: clearly signify the monsoon influence (Park et al., 2007)

-Reviewer 2: P2, L9 – physical -> physically

Author: This has been changed.

Authors changes in manuscript: P2, L20: A more physically motivated criterion

-Reviewer 2: P2, L17-18 – Full name for CARIBIC and IAGOS-MIZAIC should be provided here as well.

Author: The full names are added.

Authors changes in manuscript: P2, L7-10: CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container; e.g. Schuck et al., 2012, Rauthe-Schöch et al., 2016) and IAGOS-MOZAIC (IAGOS (In-service Aircraft for a Global Observing System) and MOZAIC (Measurements of OZone by Airbus In-service aircraft); Barret et al., 2016, Dethof et al., 1999)

-Reviewer 2: P3, L1- It is also important to mention that there is a big uncertainty in source estimates of methane (Bloom et al., 2017 GMD and references there in).

Author: The information has been added including the reference.

Authors changes in manuscript: P3L9-11: Further sources are rice cultivation and ruminants, but also swamps and flood areas. For wetlands, the uncertainty in CH4 emissions is still a large concern in atmospheric chemical transport models (Bloom et al., 2017, and references there in).

-Reviewer 2: P3, L8 – ‘variability of the AMA’ can be explained more detail here.

Author: A more detailed explanation is now added. Authors changes in manuscript: P4:L20-21: Finally, we investigated the variability of the AMA over several weeks as the anticyclone changes its position, extent, and strength due to the monsoon dynamics.

-Reviewer 2: P4, L8 (section 2.2) – I assume the trajectory calculations are done backward. Where is the initialization location?

Author: We calculated back trajectories and the initializations are along the flight tracks. The subtitle for section 2.3 is now “FLEXPART back trajectories”.

Authors changes in manuscript: P5 L8: 2.3 FLEXPART back trajectories

-Reviewer 2: P5, L11 (section 2.4) – The reason why MODIS cloud top pressure is used is missing. Is this used as convective proxy?

Author: Yes it is used as a proxy for the location of convection to compare the region with the calculated updraft of the back trajectories. This information is added in the actual section 2.5.

Authors changes in manuscript: P6 L23-24: Cloud top pressure information is used as a proxy for convection. We compared the location of the convective clouds with the location of the uplift of the back trajectories simulated by FLEXPART. The cloud top pressure data are collected from the MODIS instrument on board of AQUA

-Reviewer 2: P5, L28 – I would like to know if there are any in-situ measurements of methane and if so how the mixing ratios compare with them even over different regions in different season.

Author: Yes, there are other in situ profiles. Lelieveld et al. (2002) measured profiles over the Mediterranean in summer 2001 during MINOS. They have observed enhanced CH4 and CO values in the UT, especially during stronger influence from the AMA in the UT with CH4 mixing ratios up to ca. 1890 ppbv. Bergamaschi et al. (2013) presented CH4 profiles over the pacific in dependence of the latitude observed in 2009. The CH4 mixing ratios decrease from the northern hemisphere to the southern hemisphere. The highest values are reported for the lower troposphere in the northern hemisphere (around 1882 ppbv). In the UT CH4 increases towards the tropics to around 1800 ppbv.

Authors changes in manuscript: P7 L10: is now the position for the authors answer in the manuscript.

-Reviewer 2: P5, L30 – I have tried to find CO observations from satellite in Randel and Park (2006) but they seem to have used only ozone and water vapor.

Author: The reference was wrong and the right one is Park et al. (2007).

Authors changes in manuscript: P7,L12: Park et al. (2007) used CO observations from satellites and wind fields

-Reviewer 2: P5, L31 – to identified -> to identify

Author: This has been changed.

Authors changes in manuscript: P7 L13: to identify monsoon influenced

-Reviewer 2: P6, L8 -12 – This paragraph is not convincing to me without supporting material or references

Author: The paragraph is rewritten including supporting material and references.

Authors changes in manuscript: P7 L22-26: The observed CH4 increase with height can be explained by the global circulation. . In the boundary layer CH4 mixing ratios are influenced by turbulent mixing close to emission sources or by horizontal advection in remote places (Saito et al., 2013). At the surface the air at Gan is influenced by wind from southern directions with low CH4 mixing ratios originating from the southern Indian Ocean. High altitude advection leads to interhemispheric transport (Saito et al., 2013) thus to transfer of higher CH4 mixing ratios from the NH into the SH, which have been convectively uplifted from the boundary layer.

-Reviewer 2: P6, L18 – This is in consistent -> This is consistent

Author: This has been changed.

Authors changes in manuscript: P8 L1: This is consistent with the observed upper tropospheric increase of CO and CH4 in the NH background profiles

-Reviewer 2: L6, L20-22 – Do the mixing ratios of CO in the upper troposphere agree as well?

Author: Park et al., 2008 reported CO MR in the UT (10-15km) of around 100ppbv in the AMA and 65-90 ppbv outside. We measured in 10-14km around 74.0±15.2 ppbv and outside of 71.2±10.0 ppbv. Park et al., 2008 defined the AMA by a CO threshold opposite to our CH4 approach and in our profiles inside and outside events are included, only separated by their location, which leads to a smaller difference in the CO mixing ratios for background and AMA-influence in the UT.

Authors changes in manuscript: P8 L1-5-6: CO mixing ratios in the upper troposphere inside the AMA (around 100 ppbv in 10-15 km) in comparison to air outside the AMA (65-90 ppbv in 10-15 km).

Reviewer 2: P6, L30-31- Does this problem prevented from using the measurement or only degraded the data quality of CO measurements? Author: This problem only degraded the data quality of CO measurements.

Authors changes in manuscript: P8 L14-15: is now the position for the authors answer in the manuscript.

-Reviewer 2: P7, Eq. (1) – I think this threshold is somewhat subjective. At least it should be mentioned that this might introduce uncertainty in the analyses and also how sensitive the results are depending on the threshold values.

Author: The threshold is a simple tool to distinguish between background air masses and air masses influenced by the monsoon. It is based on in situ measurements and it is subjectively chosen, however its application to the in situ data showed a reasonable differentiation. The threshold itself was not applied to the EMAC data along the flight tracks and in the histograms (Figures 16 and 17) as the model underestimated the in situ measurements. The in situ CO and the EMAC data are distinguished into AMA-influence and background according to the time, when the in situ CH4 was above or below the CH4 threshold. Nevertheless the CH4 threshold is represented in the EMAC horizontal trace gas distributions as a contour line for a better orientation of the AMA position. Consequently the analyses depend on the threshold. A change in the absolute value would increase or decrease the region which we assumed to be influenced by the monsoon.

Authors changes in manuscript: P8 L22: In situ CH4 mixing ratios P8 L25-29: Further evaluation depends on the CH4 threshold and thus the results are sensitive to it. Nevertheless also other compounds measured during OMO showed the isolation of the anticyclone in the UT (Lelieveld et al., 2018) which confirms the usefulness of

CH4. With a change in the absolute value the region which is supposed to be AMA-influenced will be either larger or smaller, thus the edge of the anticyclone would be differently defined but the whole dynamical process is not significantly changing.

-Reviewer 2: P7, L28 – Does the difference between Scheeren et al. (2003) and this study agrees with the values in Zimmermann et al. (2018) quantitatively?

Author: Zimmermann et al. (2018) calculated a CH4 mixing ratio of 1781 ppbv for the upper troposphere between 2000 and 2006. The CH4 values in Scheeren at al. (2003) are 1819±26 ppbv for North America/North Atlantic origin and 1882±21 oobv for South Asia origin. The value in Zimmermann et al. (2018) is a global average over seven years in contrast to the values of Scheeren et al. (2203), which represent only one summer month of northern hemispheric origin, thus not accounting for the lower southern hemispheric CH4 mixing ratios. Zimmermann et al. (2018) increased the CH4 mixing ratio due to additional CH4 emissions starting in 2007 up to 1815 ppbv for 2015. In this study the CH4 mixing ratio is in average 1866.4±43.0 ppbv.

Authors changes in manuscript: P9 L21: is now the position for the authors answer in the manuscript.

-Reviewer 2: P8, L13 – cloud top height pressure -> cloud top pressure (also in P10, L23) Author: This has been changed.

Authors changes in manuscript: P10 L6: cloud top pressure P12 L23: cloud top pressure

-Reviewer 2: P8, L16-17 – This sentence should be revised for clarity.

Author: The sentence is rewritten.

Authors changes in manuscript: P11 L9-10: Matches were generally found over the Bay of Bengal, the Indo-Gangetic Plain, Bangladesh, the north easern region of India, and Myanmar. During the days when the back trajectories passed over central India, convection occurred also in this area, but the cloud top pressure was at a lower altitude than the height of the trajectories.

-Reviewer 2: P8, L34 – high pressure -> anticyclonic

Author: This has been changed.

Authors changes in manuscript: P11 L27: the anticyclonic circulation

-Reviewer 2: P9, L13 (Figs. 7 & 8) – Here, it looks like the flight path is outside the anticyclone based on the model simulations. The high values from the flight almost should be at the center of the anticyclone. I am not sure how to understand those comparisons.

Author: We added contour lines for the CH4 threshold (1879.8 ppbv) and the CH4 background (1859.4 ppbv) value in Figure 7 and 8 according to the suggestion of reviewer 1. The flight track crosses the edge of the AMA with higher mixing ratios inside the AMA, which can be seen in the measured and the modeled data. The difference between the in situ and simulated values show that on a regional scale the model is not able to reproduce the reality with respect to the absolute values.

Authors changes in manuscript: Figure 7 and 8(P31, P32) (P11 L8-9: is now the position for the authors answer in the manuscript.)

-Reviewer 2: P15, L29 – Instead of ‘these transport’ describe specific transport processes here

Author: A detailed description of the transport processes is added.

Authors changes in manuscript: P1 L19-21: In the present work, we address the transport pathways, including the convective transport from the boundary layer into the UT, the circulation in the AMA, the transport at and across the edges of the AMA, associated with outflow events and further transport in the UT partly in connection with the jet streams.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-926, 2018.

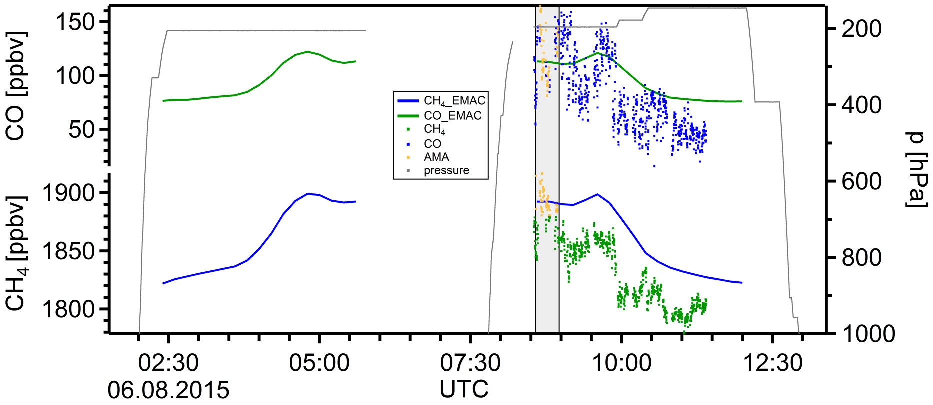

Fig. 1. Figure 24: Flight 12/13 (August 6, 2015) in situ CH4 and CO data and EMAC results along the flight track, as well as the flight altitude. The AMA is colour coded by CH4>1879.8 ppbv. Outflow region is

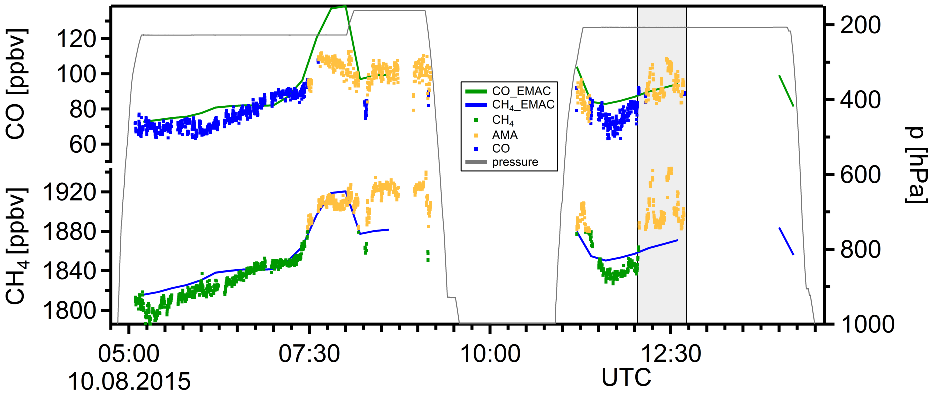

Fig. 2. Figure 25: Flight 17/18 (August 10, 2015) in situ CH4 and CO data and EMAC results along the flight track, as well as the flight altitude. The AMA is colour coded by CH4>1879.8 ppbv. Outflow region is

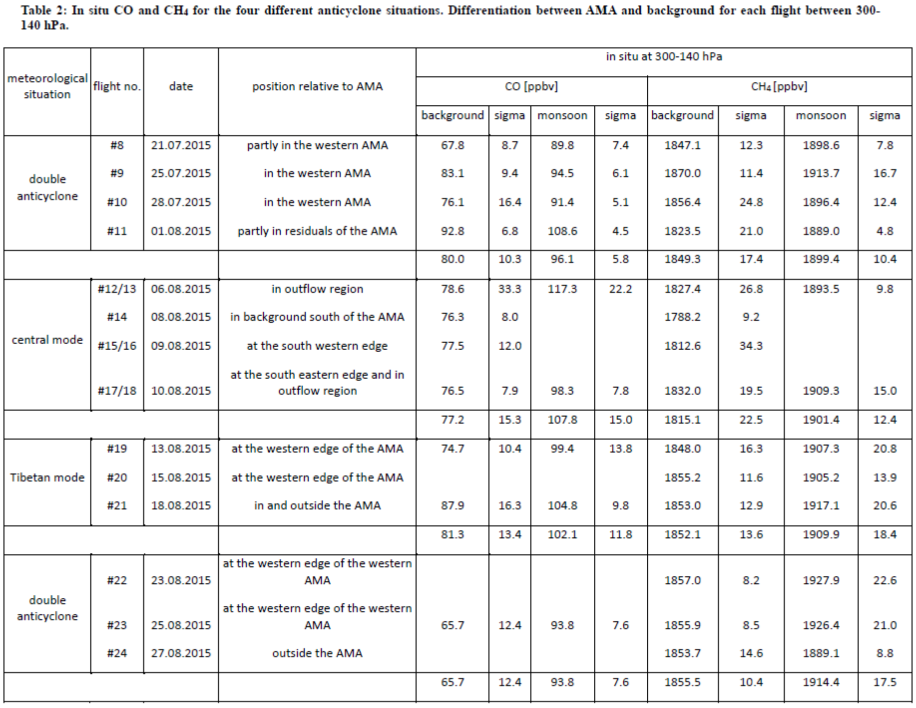

Table 2: In situ CO and CH$_4$ for the four different anticyclone situations. Differentiation between AMA and background for each flight between 300-140 hPa.

meteorological situation	flight no.	date	position relative to AMA	in situ at 300-140 hPa							
				CO [ppbv]				CH$_4$ [ppbv]			
				background	sigma	monsoon	sigma	background	sigma	monsoon	sigma
double anticyclone	#8	21.07.2015	partly in the western AMA	67.8	8.7	89.8	7.4	1847.1	12.3	1898.6	7.8
	#9	25.07.2015	in the western AMA	83.1	9.4	94.5	6.1	1870.0	11.4	1913.7	16.7
	#10	28.07.2015	in the western AMA	76.1	16.4	91.4	5.1	1856.4	24.8	1896.4	12.4
	#11	01.08.2015	partly in residuals of the AMA	92.8	6.8	108.6	4.5	1823.5	21.0	1889.0	4.8
				80.0	10.3	96.1	5.8	1849.3	17.4	1899.4	10.4
central mode	#12/13	06.08.2015	in outflow region	78.6	33.3	117.3	22.2	1827.4	26.8	1893.5	9.8
	#14	08.08.2015	in background south of the AMA	76.3	8.0			1788.2	9.2		
	#15/16	09.08.2015	at the south western edge	77.5	12.0			1812.6	34.3		
	#17/18	10.08.2015	at the south eastern edge and in outflow region	76.5	7.9	98.3	7.8	1832.0	19.5	1909.3	15.0
				77.2	15.3	107.8	15.0	1815.1	22.5	1901.4	12.4
Tibetan mode	#19	13.08.2015	at the western edge of the AMA	74.7	10.4	99.4	13.8	1848.0	16.3	1907.3	20.8
	#20	15.08.2015	at the western edge of the AMA					1855.2	11.6	1905.2	13.9
	#21	18.08.2015	in and outside the AMA	87.9	16.3	104.8	9.8	1853.0	12.9	1917.1	20.6
				81.3	13.4	102.1	11.8	1852.1	13.6	1909.9	18.4
double anticyclone	#22	23.08.2015	at the western edge of the western AMA					1857.0	8.2	1927.9	22.6
	#23	25.08.2015	at the western edge of the western AMA	65.7	12.4	93.8	7.6	1855.9	8.5	1926.4	21.0
	#24	27.08.2015	outside the AMA					1853.7	14.6	1889.1	8.8
				65.7	12.4	93.8	7.6	1855.5	10.4	1914.4	17.5

Fig. 3. Table 2: In situ CO and CH4 for the four different anticyclone situations.