

Tropospheric O₃ over Indonesia during biomass burning events measured with GOME (Global Ozone Monitoring Experiment) and compared with trajectory analysis

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Received: 26 January 2005 – Accepted: 15 April 2005 – Published: 19 May 2005

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Tropospheric ozone columns of up to 50 DU were observed by GOME (Global Ozone Monitoring Experiment) above Indonesia in September 1997, while only background amounts were measured in September 1998. The Traj.x trajectory model along with BRemen's Atmospheric PHOtochemical model (BRAPHO) were used to investigate the higher than average ozone columns above Indonesia. The transport analysis reveals that biomass burning over central Africa and northern Australia does not significantly influence ozone columns over Indonesia in September 1997. El Niño conditions, leading to extreme dryness and uncontrolled fires in Indonesia, produced ozone precursors, which are initially only slowly advected westwards to the central Indian Ocean. Joint transport and chemistry modelling was able to reproduce the spatial distribution and amounts of ozone, NO₂ and formaldehyde columns over Indonesia. The chemistry modelling shows a net production of 3.1 Tg of ozone produced by biomass burning in Indonesia in September 1997. Transport analysis further reveals that ozone columns over the Indian Ocean, between 10 and 20° S can be accounted for by the mixing of air masses containing NO_x from lightning over the Congo Basin with air masses containing volatile organic compounds from biomass burning.

1. Introduction

Biomass burning, often initiated in order to clear land for agricultural purposes, has been identified as a significant source of trace gases. These emissions affect the atmosphere at local, regional and global scales (Crutzen et al., 1979; Seiler and Crutzen, 1980; Crutzen and Andreae, 1990; Levine et al., 1995; Levine, 1999). Along with particulates, the major gases produced by biomass burning include carbon dioxide (CO₂), carbon monoxide (CO), methane (CH₄), formaldehyde (HCHO), oxides of nitrogen (NO_x = NO+NO₂) and ammonia (NH₃). The combination of CH₄, CO, nonmethane hydrocarbons (NMHC) and NO_x in the atmosphere leads to the photochemical produc-

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tion of tropospheric O₃. Some of these species, CO₂, CH₄, and O₃ are also greenhouse gases that contribute to global warming. Ozone also affects the abundance of important atmospheric oxidants such as OH and peroxy radicals (Atkinson, 2000). The lifetime of these ozone precursors and of ozone itself (Oltmans et al., 1998) is sufficiently long that they can be transported hundreds or even thousands of kilometres away from their source before being chemically destroyed or deposited.

In 1997/98 a smog blanket of 3 million km² covered a large area of Indonesia and affected the health of 75 million people in 6 countries. At the same time extensive biomass burning was occurring in Indonesia. It was caused by an extreme dryness due to the strongest El Niño conditions ever recorded (CTI, Cold Tongue Index of 2.2) (Folland and Parker, 1995). Biomass burning was also occurring in Africa and Australia and could potentially contribute to the smog over Indonesia through transport.

The objective of this study is to qualitatively and quantitatively understand the meteorological and anthropogenic contributions that led to the enormous increase of tropospheric O₃ in September 1997 as compared to September 1998 over Indonesia.

Because of the large extent of the fires over Indonesia and potential influences from air masses originating over Africa and Australia, in situ measurements are insufficient to provide adequate explanation of this large scale event. GOME (Global Ozone Monitoring Experiment) measurements of tropospheric ozone and ozone precursors have wide spatial and temporal coverage. In conjunction with chemical and transport models these data can be used to assess their origin of this event. For this study tropospheric columns of ozone, NO₂, and formaldehyde were retrieved for the Indonesian region. Possible contribution to the ozone column due to transport of air masses affected by biomass burning in Africa, Australia and Indonesian region are qualitatively evaluated using the trajectory model Traj.x. Biomass burning locations were identified from Along the Track Scanning Radiometer (ATSR) data collected over the same time period. A quantitative view of the impact of the Indonesian fires alone during September 1997 was evaluated with the chemical boxmodel BRAPHO (BREMen's Atmospheric PHOtochemical boxmodel) for a large number of trajectories.

2. Methodology and tools

2.1. Trace gas columns retrieved from GOME

GOME was launched in April 1995 onboard the European Research Satellite (ERS)-2 into a near-sun-synchronous orbit at a mean altitude of 795 km. The descending node crosses the equator every 2800 km at 10:30 a.m. local time. GOME is a nadir-scanning double-monochromator measuring the sun-light scattered from the Earth's atmosphere and/or reflected by its surface in the wavelength region of 240 to 790 nm at a moderate spectral resolution of 0.17 to 0.33 nm. Total ground coverage is obtained within 3 days at the equator by a 960 km across track swath (4.5 s forward scan, 1.5 s back scan).

The solar irradiance is measured daily.

Details of the overall scientific objectives of GOME, the instrument concept, and some scientific results are reported elsewhere (Burrows et al., 1999, 2000). In addition, a number of studies have been published focusing on the retrieval of tropospheric NO₂ (Leue et al., 2001; Richter and Burrows, 2002; Martin et al., 2002) and HCHO (Ladstätter-Weißmayer et al., 1998; Chance et al., 2000; Thomas et al., 1998; Palmer et al., 2003) from GOME data. The ozone columns used in this study are based on lv2-v3.0-data which were processed by the GOME Data Processor (GDP) at the German Remote Sensing Data Center (DFD) (DLR, 1996).

As GOME is a nadir viewing instrument, both tropospheric and stratospheric absorptions contribute to the measured signal. While this is not critical for HCHO which has a very small stratospheric column, the stratospheric O₃ and NO₂ columns can not be neglected in the analysis. Therefore, the Tropospheric Excess Method (TEM) was used to derive the tropospheric columns of O₃ (Ladstätter-Weißmayer et al., 2004) and of NO₂ (Richter and Burrows, 2002).

The TEM method is based on the assumption that stratospheric O₃ and NO₂ do not vary zonally, and therefore the total columns measured on the same day at the same latitude over a clean air region can be used as an approximation of the stratospheric column over the region of interest. In this study, data from the Pacific region

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(180–190°) were used as reference sector. In addition only GOME data with a cloud cover less than 10% (due to GOME ICFA cloud algorithm) were analysed. The overall analysis then consists of three steps: (a) determination of the total slant column, (b) subtraction of the stratospheric contribution including the tropospheric background yielding the tropospheric excess slant column and (c) division by the air mass factor (AMF) (Rozanov et al., 1997) to obtain the vertical tropospheric excess column. The tropospheric column is then calculated by the addition of the tropospheric background of 21 DU as measured by SHADOZ (Southern Hemisphere Additional Ozonesondes) (Thompson et al., 2003) over Fiji (18.13° S, 178.40° E) for O₃ and of 2.5·10¹⁴ molec cm⁻² for NO₂ (Boersma et al., 2004). The main error sources are longitudinal inhomogeneities in the stratospheric contribution of the species analysed, uncertainties in cloud cover, surface albedo, aerosol loading and the assumed vertical profile of O₃ and NO₂ which are required in calculating the AMF (Richter and Burrows, 2002).

The overall error of the analysis is estimated to be generally in the order of 4 DU for tropospheric O₃ columns (Ladstätter-Weißenmayer et al., 2004) and 1.5·10¹⁵ molec cm⁻² for tropospheric columns of NO₂ (Richter and Burrows, 2002).

HCHO columns are obtained with a detection limit of about 2.5·10¹⁵ molec cm⁻². Since the stratospheric contribution to its total column is negligible, this represents the detection limit for the tropospheric formaldehyde loading.

The tropospheric columns of ozone and NO₂ which were derived by applying the TEM and the total formaldehyde column for September 1997 and September 1998 are shown in Fig. 1. During the September 1997 Indonesian biomass burning episode an increase of monthly mean tropospheric ozone columns of up to 50 DU is detected. The monthly mean tropospheric column of the ozone precursor NO₂ peaked at 2.6·10¹⁵ molec cm⁻² during September 1997. The formaldehyde column, which scales with the abundance of VOC emissions, peaked at 3.0·10¹⁶ molec cm⁻² (see Fig. 1). On 21 September 1997 GOME performed measurements in the region of Borneo. The retrieval of tropospheric formaldehyde from these measurements shows enhanced amounts of this trace gas and has been discussed elsewhere (Ladstätter-

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Weißmayer et al., 1998).

The SHADOZ network reported a maximum tropospheric ozone column of 57 DU over Watukosek, Java (7.57° S, 112.65° E) (see Fig. 2) (Fujiwara et al., 2003) during the same Indonesian pollution episode.

In contrast, in September 1998 the tropospheric ozone columns in the Indonesian region remained below 30 DU which is very close to background conditions.

ATSR, also aboard the ERS-2, measures nighttime infrared radiation emitted by the Earth's surface. Hotspot data from this instrument provided by the European Space Agency (ESA), are identified as fires (Arino et al., 1997). By projecting all hotspots detected during September 1997 and September 1998 onto a global grid of 1.125×1.125°, the fire index, shown in Fig. 3, is generated for both episodes. Biomass burning in Indonesia was much stronger in September 1997 than in September 1998. In contrast, biomass burning over the southern parts of Africa and the northern parts of Australia both show no significant difference in intensity or spatial distribution when comparing September 1997 to September 1998.

When accounting for air pollution in the tropical regions, lightning cannot be omitted since it is an important source of nitrogen oxides especially in the upper troposphere. Due to the fact that its intensity is strongly enhanced in equatorial regions, a multiannual climatology based on the Lightning Imaging Sensor (LIS) and the Optical Transient Detector (OTD) (Christian et al., 2003) is applied.

2.2. Trajectory analysis and chemistry modelling

To determine which air masses contribute biomass burning pollutants to the Indonesian region during this episode, the trajectory model Traj.x, developed at the Institute of Environmental Physics, University of Bremen was used. Traj.x is a kinematic trajectory model which calculates the transport of air parcels along wind fields by applying a fourth-order Runge-Kutta scheme. The meteorological data were taken from the European Centre for Medium-Range Weather Forecasts (ECMWF)'s 40-year reanalysis data set (ERA-40).

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For each day in September 1997 and 1998, trajectories were initiated at 00:00 UT, 06:00 UT, 12:00 UT and 18:00 UT, starting from regions where biomass burning (or lightning, as discussed later) was occurring. All trajectories were released at altitudes of 50, 100, 150 and 200 hPa above ground level.

Altogether about 230 000 trajectories were calculated for air masses originating in biomass burning activity in central Africa, northern Australia, Indonesia and lightning activity over the African Congo Basin. To derive a statistical overview of the transport, a quantity which is referred to as the trajectory density is deduced. This quantity is derived by projecting the current position of all trajectories being considered into a three dimensional grid depending on longitude, latitude and altitude with a resolution of $1.125 \times 1.125^\circ$ and 50 hPa respectively.

By this means the amount of trajectories travelling through each gridcell is determined. High values of the trajectory density denote a large number of trajectories having passed through the regarded volume.

To better compare the transport patterns of air masses released from different processes, the trajectory density is normalised to “1”.

By vertically integrating the trajectory density, a general picture of the spatial transport patterns is obtained.

Regions of high trajectory density qualitatively identify the average paths of air masses influenced by fires or lightning in September 1997. To get a quantitative view on the impact of each regarded emission process on the air chemistry, the chemistry taking place in the different regarded air masses has to be taken into account.

This was achieved by applying the Lagrangian box model BRAPHO (BRemen’s Atmospheric PHOtochemical model) (Meyer-Arnek, 2004). This boxmodel is run on a stochastically chosen subset of the 230 000 trajectories computed to determine the transport pattern of the airmasses. It is based on ASAD (A Self contained Atmospheric chemistry code) (Carver et al., 1997). Lagrangian models describe the chemistry taking place in a closed volume of air which does not undergo any mass exchange with the surroundings.

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The chemistry scheme which was used in this study is based on the very detailed Master Chemical Mechanism (MCM) catalogue version 3.0 (Saunders et al., 2003). In this compilation about 11 000 reactions describe the chemical interactions between about 3500 different species.

As no detailed trace gas measurements were performed during September 1997 over the Indonesian region, the model is initialised by airborne measurements conducted during the TRACE-A-campaign (Transport and Atmospheric Chemistry near the Equator-Atlantic) (Fishman et al., 1996). This campaign took place in September and October 1992 over the southern parts of America, Africa and the Atlantic Ocean. Measurements were partly performed over areas where biomass burning was taking place. By Emmons et al. (2000) all measurements obtained during this campaign were merged into a three dimensional composite. The chemistry model is initialised with averaged composites representing airborne measurements carried out over enhanced burning activity over Africa within the lowermost kilometer. They represent airmasses which recently left the fire, but are already cooled down to environmental temperatures, so that the complicated fire chemistry does not have to be taken into account by the boxmodel.

Applying results from measurements which were performed over the burning African savanna during the TRACE-A-campaign for modelling the Indonesian fires of September 1997 can only be regarded as a first approach in modelling this event. Over Indonesia burning peat contributed most to the air pollution. Since hardly any information about VOC released by peat fires is available, this approach was performed.

During the TRACE-A-campaign the concentrations of about 10 different species have been measured. Only these species (O₃, NO₂, CO, C₂H₂ (ethyne), C₃H₈ (propane), CH₃COCH₃ (acetone), H₂O₂ (hydrogen peroxide), HNO₃ (nitric acid), PAN (peroxyacetyl-nitrate)) have been used to initialise the chemistry model. Consequently the MCM chemistry was reduced to represent the interactions between all of the measured species and of all their oxidation products.

The reduction was performed by extracting the complete oxidation paths of all

species mentioned above and of methane. This procedure lead to a model chemistry consisting of 400 reactions between 133 different species.

To compare the volume mixing ratios (VMR) resulting from the chemistry modelling with trace gas columns retrieved from GOME measurements, the VMR of selected compounds are projected into a three dimensional grid depending on longitude, latitude and pressure (Meyer-Arnek, 2004). These mixing ratios are then integrated vertically to yield vertical columns.

To take into account the diurnal cycle of the photochemistry, only model results obtained for 10:30 local time (GOME's overpass time) are applied to this procedure.

To account for the strong accumulation of fire releases due to extremely low horizontal windspeeds over the Indonesian region during this episode, leading to an enormous increase of initial tracer concentrations, a sensitivity study was performed. At first a base run initialised with the data set as derived from the TRACE-A-composites was performed. In four subsequent runs, the VMR of all considered nonmethane hydrocarbons (NMHC) were increased by a factor of 2, 4, 10 and 20. Methane was increased by 20% over the course of these four additional runs. Initial mixing ratios of NO_x remained unchanged. The 5th model run (initialised with $5 \times [\text{NMHC}]$ and $1.2 \times \text{CH}_4$) shows the best agreement between GOME retrieval and model result for ozone, NO_2 and HCHO. The corresponding chemistry model initialisation is shown in Table 1.

As the impact of biomass burning is considered to be major with respect to all of the species applied for the model initialisation, anthropogenic and biogenic emissions are not considered.

3. Results and discussion

3.1. Trajectory analysis and the origin of the trace gases in the Indonesian region

Three dimensional trajectory densities reveal that air masses originating over Australian or African biomass burning are either convectively lifted up into the upper –

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where these air masses undergo long-range transport – or they are slowly advected at lower altitudes. The projection of the three dimensional trajectory density on the ground is shown in Fig. 4.

The majority of the air masses released from fires over northern Australia is transported in easterly directions, away from the Indonesian region. At lower altitudes (below 700 hPa) a small portion of polluted air masses is directly advected into the Indonesian region.

The same accounts for emissions from biomass burning over Africa. Emissions either stay in the vicinity of the fires at lower altitudes or they are risen up by convection. In the upper troposphere these air masses mostly undergo long-range-transport into easterly directions. Consequently fire releases from biomass burning over Africa arrive over the Southern Central parts of the Indian Ocean, but do not reach Indonesia itself (see Fig. 4).

Kunhikrishnan et al. (2004) investigated the intra-annual distribution of NO_x over the central Indian Ocean (from 10° N to 20° S) by applying the 3-D-global chemistry transport model MATCH-MPIC (Lawrence et al., 2003) driven by meteorological analysis from NCEP/NCAR. The study reveals that NO_x-enriched air masses released over Africa are responsible for the amount of NO₂ derived from GOME measurements.

The assumption that the influence of emissions from Africa and Australia is minor is underlined by the low interannual variability of hotspot data derived from ATSR measurements. The biomass burning activity remained constant for both episodes (September 1997 and September 1998) whereas the corresponding trace gas columns over the Indonesian region experienced extraordinary differences already shown in Fig. 4.

Trajectories of air masses originating over biomass burning in Indonesia indicate that fire emissions rose up slowly and are then gradually spread into westerly directions. Due to weak easterlies in the equatorial region, trajectories travelled only up to 25 degrees within 10 days. During fall 1997 the convective rise of air masses over the Indonesian region was partly prevented by the descending branch of the longitudinal

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Walker circulation (Holton, 1992) related to the extreme El Niño conditions.

This downdraft led to an extreme dryness, lengthening the duration and severity of the biomass burning event. Consequently the combination of meteorological conditions and severe burning resulted in an enormous increase in the concentration of pollutants (both gas phase emissions and aerosols) directly over Indonesia.

While the transport analysis indicates that the unusually high tropospheric O₃, NO₂ and formaldehyde columns observed by GOME directly over Indonesia in September 1997 were produced largely by local events, high columns of ozone and NO₂ over the Indian Ocean between 10 to 20° S (see Fig. 1) cannot be explained by transport of Indonesian fire emissions.

CO can be regarded as a tracer for emissions from biomass burning. Measurements of CO amounts derived from measurements of the MOPITT instrument (Measurement of Pollution In The Troposphere) (Drummond and Mand, 1996; Crawford et al., 2004) performed during September 2002 are shown in Fig. 5. Monthly mean transport patterns of both episodes (September 2002 (MOPITT measurements) and September 1997 (GOME measurements) can reasonable be assumed to be comparable as both studies targeted El Niño conditions. These data reveal that the CO above Indonesia is strongly influenced by fires in Indonesia itself. It also indicates that significant amounts of CO in the Indian Ocean region originates in biomass burning taking place over Africa (Bremer et al., 2004). Although biomass burning emissions in Africa are quite plausible as an explanation of O₃ over the Indian Ocean, poor agreement between the GOME retrieved trace gas columns and model results for the region over the Indian Ocean between 10 and 20° S suggests that some additional source of pollution has still not been accounted for. Particular in the upper troposphere lightning is a globally significant source of NO_x. The total amount of lightning-NO_x is still an issue of ongoing research activity.

While the lifetime of NO_x in the lower troposphere is too short to survive transport from Africa to the Indian Ocean (τ is in the order of hours), its lifetime in the upper troposphere is much longer (τ is in the order of days), (Jaegle et al., 1998; Kunhikrishnan

et al., 2004) and allows its effective transport. Climatological LIS and OTD data state strong lightning activity over the Congo Basin in central Africa (Christian et al., 2003) for September.

To investigate whether the mixing of air masses influenced by lightning (yielding enhanced NO_x -levels) with air masses released from biomass burning (yielding enhanced VOC levels) may have caused the observed enhancement of ozone, the 3-D-trajectory densities of both emission processes were combined.

Mixing by which ozone is possibly formed is assumed to take place when the trajectory density of air masses influenced by biomass burning (released from biomass burning over Indonesian Africa and Australia) and the trajectory density of air masses influenced by lightning are both nonzero in a certain grid cell.

Applying this criterion reveals that mixing is most intense over the Indian Ocean between 10 and 20° S at altitudes between 600 and 200 hPa.

In this study, mixing is not quantitatively accounted for: If mixing is assumed to take place within a grid cell, the corresponding mixing state is set to unity, otherwise it remains zero.

Integrating this mixing status over pressure and normalise it with the pressure difference between ground and tropopause yields Fig. 6. It shows a good agreement with GOME observations in this area and reveals that mixing is an important process which has to be accounted for when considering the tropospheric trace gas distribution.

3.2. Chemical modelling

The best agreement between modelled and GOME retrieved tropospheric O_3 , NO_2 and formaldehyde columns is achieved using initial NMHC mixing ratios 20 times higher than those given in Table 1 along with a 20% higher methane mixing ratio. The resulting tropospheric columns for this model run are shown in Fig. 7.

When considering the uncertainties of the joint transport and chemistry analysis, the differences between the modelled and retrieved tropospheric columns are to be considered as relatively small.

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Although maxima in tropospheric O₃, NO₂ and formaldehyde columns are well reproduced, the largest remaining discrepancies between the model and GOME results are in the special extent of NO₂ columns and the persistence of elevated formaldehyde columns above Indonesia.

5 The reason why the modelled NO₂ columns cover smaller geographical areas than observed by GOME may be caused by the overestimation of NO₂ loss from modelled air masses through irreversible deposition in the form of HNO₃. Overestimation of HNO₃ deposition may, in turn, arise from an underestimation of OH loss via chemical reactions between OH and NMHC/VOCs. Higher OH concentrations enhance the contribution of the three-body reaction between OH, NO₂ and a collision partner leading to formation of HNO₃. Removal of HNO₃ from modelled air masses through deposition is then revealed as a decrease in the spacial extent of the modelled NO₂ columns.

Unusually short duration of modelled formaldehyde columns may arise from overestimation of the rate of chemical decomposition of formaldehyde. Reducing the formaldehyde lifetime will reduce the distance formaldehyde can be transported and will affect the persistence of formaldehyde columns. Further work concerning the sources of these errors has been carried out and will be published separately. Given that pollution over Indonesia in September 1997 is largely generated by local fires, the total amount of ozone produced by these fires can be estimated by integrating time averaged ozone columns obtained as from BRAPHO.

Using the combined chemistry and transport models with air masses originating only above Indonesian fires and allowing 10 days for chemical processing, an estimated 3.1 Tg of ozone were produced by biomass burning in Indonesia for a month long period beginning 1 September 1997. In an earlier work [Levine \(1999\)](#) estimated that 7.1 Tg of ozone were produced over September and October 1997 (61 days). When only accounting for the ozone production during September and assuming an equal ozone production for each day of this episode, due to [Levine \(1999\)](#) about 3.5 Tg of ozone were generated by the Indonesian fires. This value is in reasonable agreement with the value calculated in our study.

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The difference may be due to the fact that the lifetime of VOC precursors of ozone and of ozone itself is on the order of days to weeks (depending on the humidity of the surrounding air). Allowing the joint transport and chemistry analysis to account for the first 10 days after the release of the regarded air masses leads to an underestimation of the ozone production.

4. Conclusions

GOME measurements show that ozone columns over Indonesia in September 1997 reached up to 50 DU and were higher than the more typical background conditions of surrounding months and higher than values in September 1998. Trajectory analyses and the lack of differences in burning activity in Africa and Australia between 1997 and 1998, suggest that air masses from Africa and northern Australia do not contribute significantly to increased ozone columns over Indonesia, although they do contribute to ozone columns over the Indian Ocean between 10 and 20° S. Trajectory analyses also suggest that air masses from biomass burning in Indonesia itself were only slowly transported away from this region and caused the unusually high ozone columns observed by GOME. Chemical modeling reasonably reproduces O₃, NO₂ and formaldehyde columns observed by GOME in the larger Indonesian region. Based on model results, the amount of ozone produced in a month long period in September 1997 over Indonesia itself is estimated to be 3.1 Tg, most of which is derived from biomass burning in Kalimantan and Sumatra. Enhanced tropospheric O₃ columns over the Indian Ocean cannot be explained by biomass burning alone. Transport models indicate that the spatial distribution of the ozone maximum over the Indian Ocean is likely to be generated by mixing of air masses containing NO_x derived from lightning over the Congo Basin and air masses containing VOCs derived from biomass burning.

Acknowledgements. The authors would like to thank B.-M. Sinnhuber, A.-M. Schmoltnner and T. Custer for fruitful discussions and ECMWF for providing meteorological data. Parts of this work have been funded by the University of Bremen, Germany, the DLR/DARA, the European

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- Arino, O., Melinotte, J.-M., Rosaz, J.-M., and Monjoux, E.: ESA Fire Product, Proceedings of the 7th ISPRS conference on Physical Measurement and Signatures in Remote Sensing, Courchevel, 1997. [3110](#)
- Atkinson, R.: Atmospheric chemistry of VOCs and NO_x, *Atmos. Environ.*, 34, 2063–2101, 2000. [3107](#)
- Boersma, K. F., Eskes, H. J., and Brinksma, E. J.: Error Analysis for Tropospheric NO₂ Retrieval from Space, *J. Geophys. Res.*, 109, D04311, doi:10.1029/2003JD003962, 2004. [3109](#)
- Bremer, H., Kar, J., Drummond, J. R., Nichitu, F., Zou, J., Liu, J., Gille, J. C., Deeter, M. N., Francis, G., Ziskin, D., and Warner, J.: Spatial and temporal variation of MOPITT CO in Africa and South America: A comparison with SHADOZ ozone and MODIS aerosol, *J. Geophys. Res.*, 109, D12304, doi:10.1029/2003JD004234, 2004. [3115](#)
- Burrows, J. P., Weber, M., Buchwitz, M., Rozanov, V. V., Ladstätter-Weißenmayer, A., Richter, A., de Beek, R., Hoogen, R., Bramstedt, K., Eichmann, K.-U., Eisinger M., and Perner, D.: The Global Ozone Monitoring Experiment (GOME): Mission Concept and First Scientific Results, *J. Atm. Sciences*, 56, 151–175, 1999. [3108](#)
- Burrows, J. P., Richter, A., Weber, M., Eichmann, K.-U., Bramstedt, K., Ladstätter-Weißenmayer, A., Wittrock, F., Eisinger M., and Hild, L.: Satellite observations of tropospheric and stratospheric gases in Chemistry and Radiation Changes in the Ozone Layer, Kluwer Academic Publisher, 301–329, 2000. [3108](#)
- Carver, G. D., Brown P. D., and Wild, O.: The ASAD atmospheric chemistry integration package and chemical reaction database, *Comput. Phys. Commun.*, 105, 2, 197–215, 1997. [3111](#)
- Chance, K., Palmer, P., Spurr, R. J. D., Martin, R. V., Kurosu T. P., and Jacob, D. J.: Satellite observations of formaldehyde over North America from GOME, *Geophys. Res. Lett.*, 27(21), 3461–3464, doi:10.1029/2000GL011857, 2000. [3108](#)
- Christian, H. J., Blakeslee, R. J., Boccippio, D. J., Boeck, W. L., Buechler, D. E., Driscoll, K. T., Goodman, S. J., Hall, J. M., Mach, D. M., and Stewart, M. F.: Global frequency and distribu-

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- tion of lightning as observed from space by the Optical Transient Detector, *J. Geophys. Res.*, 108(1), doi:10.1029/2002JD002347, 2003. [3110](#), [3116](#)
- Crawford, J. H., Heald, C. L., Fuelberg, H. E., Morse, D. M., Sachse, G. W., Emmons, L. K., Gille, J. C., Edward, D. P., Deeter, M. N., Chen, G., Olson, J. R., Connors, V. S., Kittaka, C., and Hamlin, A. J.: Relationship between Measurements of Pollution in the Troposphere (MOPITT) and in situ observations of CO based on a large-scale feature sampled during TRACE-P, *J. Geophys. Res.*, 109, D15S04, doi:10.1029/2003JD004308, 2004. [3115](#)
- Crutzen, P. J., Heidt, L. E., Krasnec, J. P., Pollock, W. H., and Seiler, W.: Biomass burning as a source of atmospheric gases CO, H₂, N₂O, NO, CH₃Cl and COS, *Nature*, 282, 253–256, 1979. [3106](#)
- Crutzen, P. J. and Andreae, M. O.: Biomass burning in the tropics: Impact on atmospheric chemistry and biochemical cycles, *Science*, 250, 1669–1677, 1990. [3106](#)
- GOME level 1 to 2 Algorithms Description, Deutsches Zentrum für Luft- und Raumfahrt (DLR), Oberpfaffenhofen, Germany, Tech. Rep. ER-TN-DLR-GO-0025, Iss./Rev.2/A, DLR/SAO, 1996. [3108](#)
- Drummond, J. R. and Mand, G. S.: The measurement of pollution in the Troposphere (MOPITT) instrument: Overall performances and calibration requirements, *J. Atmos. Ocean. Tech.*, 13, 314–320, 1996. [3115](#)
- Emmons, L. K., Hauglustaine, D. A., Müller, J. F., Carroll, M. A., Brasseur, G. P., Brunner, D., Staehelin, J., Thouret, V., and Marengo, A.: Data composites of airborne observations of tropospheric ozone and its precursors, *J. Geophys. Res.*, 105(16), 20 497–20 538, doi:10.1029/2000JD900232, 2000. [3112](#)
- Fishman, J., Hoell, J. M., Bendura, R. D., McNeal R. J., and Kirchhoff, V. W. J. H.: NASA GTE TRACE A Experiment (September–October 1992): Overview, *J. Geophys. Res.*, 101(19), 23 865–23 879, 1996. [3112](#)
- Fujiwara, M., Tomikawa, Y., Kita, K., Kondo, Y., Komala, N., Saraspriya, S., Manik, T., Suropto, A., Kawakami, S., Ogawa, T., Kelana, E., Suhardi, B., Harijono, S. W. B., Kudsy, M., Sribimawati, T., and Yamanaka, M. D.: Ozone-sonde observations in the Indonesian maritime continent: A case study on ozone rich layer in the equatorial upper troposphere, *Atmos. Environ.*, 37, 353–362, 2003. [3110](#)
- Folland, C. K. and Parker, D. E.: Correction of instrumental biases in historical sea surface temperature data, *Quart. J. Roy. Meteor. Soc.*, 121, 319–367, 1995. [3107](#)
- Holton, J. R.: *An Introduction to Dynamic Meteorology*, Academic Press Inc., 1992. [3115](#)

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- Jaegle, L., Wang, Y., Weinheimer, A. J., Ridley, B. A., Campos, T. L., Sachse, G. W., and Hagen, D. E.: Sources and chemistry of NO_x in the upper troposphere over the United States, *Geophys. Res. Lett.*, 25(10), 1709–1712, doi:10.1029/98GL00041, 1998. [3115](#)
- 5 Kunhikrishnan, T., Lawrence, M. G., von Kuhlmann, R., Richter, A., Ladstätter-Weißmayer, A., and Burrows, J. P.: Semiannual NO₂ plumes during the monsoon transition periods over the central Indian Ocean, *Geophys. Res. Lett.*, 31, L08110, doi:10.1029/2003GL019269, 2004. [3114](#), [3115](#)
- Ladstätter-Weißmayer, A., Burrows, J. P., and Perner, D.: Biomass burning over Indonesia as observed by GOME, *Earth Obs. Quart.*, 58, 28–29, 1998. [3108](#), [3109](#)
- 10 Ladstätter-Weißmayer, A., Meyer-Arnek, J., Schlemm, A., and Burrows, J. P.: Influence of stratospheric airmasses on tropospheric vertical O₃ columns based on GOME (Global Ozone Monitoring Experiment) measurements and backtrajectory calculation over the Pacific, *Atmos. Chem. Phys.*, 4, 903–909, 2004, [SRef-ID: 1680-7324/acp/2004-4-903](#). [3108](#), [3109](#)
- 15 Lawrence, M. G., von Kuhlmann, R., Salzmann, M., and Rasch, P. J.: The balance of effects of deep convective mixing on tropospheric ozone, *Geophys. Res. Lett.*, 30, 1940, doi:10.1029/2003GL017644, 2003. [3114](#)
- Leue, C., Wenig, M., Wagner, T., Klimm, O., Platt, U., and Jähne, B.: Quantitative analysis of NO_x emissions from GOME satellite image sequences, *J. Geophys. Res.*, 106(6), 5493–5505, doi:10.1029/2000JD900572, 2001. [3108](#)
- 20 Levine, J. S., Cofer, W. R., Cahoon, D. R., and Winstead, E. L.: Biomass burning: A Driver for Global Change, *Environ. Sci. Technol.*, 29, 3, 120A–125A, 1995. [3106](#)
- Levine, J. S.: The 1997 fires in Kalimantan and Sumatra, Indonesia: Gaseous and particulate emissions, *Geophys. Res. Lett.*, 26(7), 815–818, doi:10.1029/1999GL900067, 1999. [3106](#), [3117](#)
- 25 Martin, R. V., Chance, K., Jacob, D. J., Kurosu, T. P., Spurr, R. J. D., Bucsel, E., Gleason, J. F., Palmer, P. I., Bey, I., Fiore, A. M., Li, Q., Yantosca, R. M., and Koelmeijer, R. B. A.: An improved retrieval of tropospheric nitrogen dioxide from GOME, *J. Geophys. Res.*, 107(20), doi:10.1029/2001JD001027, 2002. [3108](#)
- 30 Meyer-Arnek, J.: Qualitative und quantitative Analyse einer Episode erhöhter Spurengassäulen im September 1997, Institute for Environmental Physics, University of Bremen, Bremen, 2004. [3111](#), [3113](#)
- Oltmans, S. J., Lefohn, A. S., Scheel, H. E., Harris, J. M., Levy II, H., Galbally, I. E., Brunke,

**Tropospheric O₃ over
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- E.-G., Meyer, C. P., Lathrop, J. A., Johnson, B. J., Shadwick, D. S., Cuevas, E., Schmidlin, F. J., Tarasick, D. W., Claude, H., Kerr, J. B., Uchino, O., and Mohnen, V.: Trends of Ozone in the Troposphere, *Geophys. Res. Lett.*, 25(2), 139–142, 1998. [3107](#)
- Palmer, P. I., Jacob, D. J., Fiore, A. M., Martin, R. V., Chance, K., and Kurosu, T. P.: Mapping isoprene emissions over North America using formaldehyde columns observations from space, *J. Geophys. Res.*, 108(6), 4180, doi:10.1029/2002JD002153, 2003. [3108](#)
- Richter, A. and Burrows, J. P.: Retrieval of tropospheric NO₂ from GOME measurements, *Adv. Space Res.*, 29(11), 1673–1683, 2002. [3108](#), [3109](#)
- Rozanov, V., Diebel, D., Spurr, R. J. D., and Burrows, J. P.: GOMETRAN: A radiative transfer model for the satellite project GOME – the plance parallel version, *J. Geophys. Res.*, 102(14), 16 683–16 695, doi:10.1029/96JD01535, 1997. [3109](#)
- Saunders, S. M., Jenkin, M. E., and Derwent, R. G.: Protocol for the development of the Master Chemical Mechanism, MCM v3, *Atmos. Chem. Phys.*, 3, 161–180, 2003, [SRef-ID: 1680-7324/acp/2003-3-161](#). [3112](#)
- Seiler, W. and Crutzen, P. J.: Estimates of gross and net fluxes of carbon between the biosphere and the atmosphere from biomass burning, *Climatic Change*, 2, 207–247, 1980. [3106](#)
- Thomas, W., Hegels, E., Slijkhuis, S., Spurr, R. J. D., and Chance, K.: Detection of biomass burning combustion products in Southeast Asia from backscatter data taken by the GOME spectrometer, *Geophys. Res. Lett.*, 25(9), 1317–1320, doi:10.1029/98GL01087, 1998. [3108](#)
- Thompson, A. M., Witte, J. C., McPeters, R. D., Oltmans, S. J., Schmidlin, F. J., Logan, J. A., Fujiwara, M., Kirchhoff, V. W. J. H., Posny, F., Coetzee, G. J. R., Hoegger, B., Kawakami, S., Ogawa, T., Johnson, B. J., Vömel, H., and Labow, G.: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology. 1. Comparison with Total Ozone Mapping Spectrometer (TOMS) and ground-based measurements, *J. Geophys. Res.*, 108(2), doi:10.1029/2001JD000967, 2003. [3109](#)

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et al.**Table 1.** Chemical initialisation of the photochemical boxmodel for modelling the chemistry of airmasses being released from biomass burning over Indonesia.

Specie	VMR [ppb]
O ₃	72.000
CO	1285.000
CH ₄	2028.000
NO	0.970
CH ₃ OOH	16.685
HCHO	6.907
C ₂ H ₆	31.958
C ₂ H ₂	19.360
C ₃ H ₈	4.267

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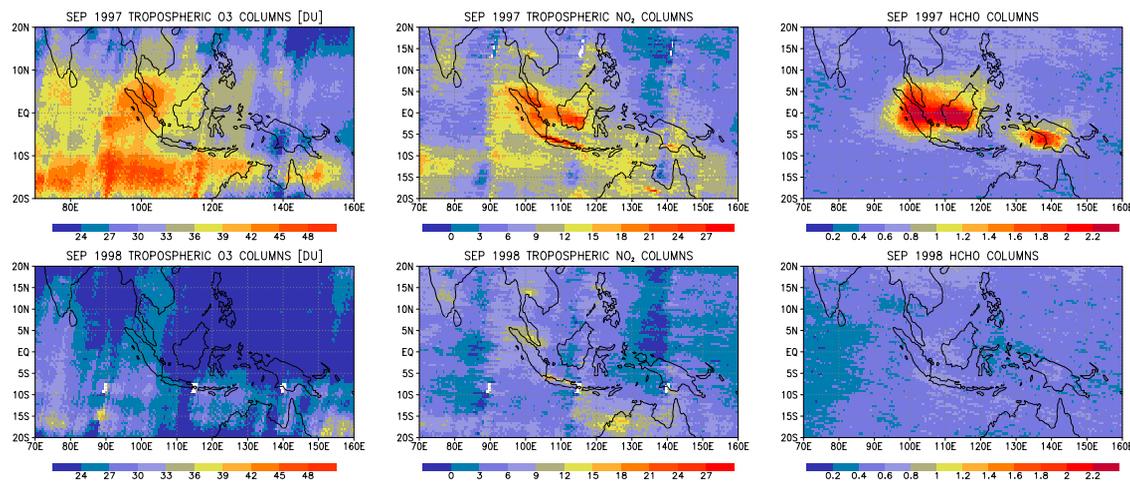


Fig. 1. Monthly mean tropospheric vertical columns of O₃, NO₂ and formaldehyde (given in DU (Dobson Units), 10¹⁴ molecules cm⁻² respectively 10¹⁶ molecules cm⁻²) as retrieved from GOME measurements performed in September 1997 (upper row) and September 1998 (lower row).

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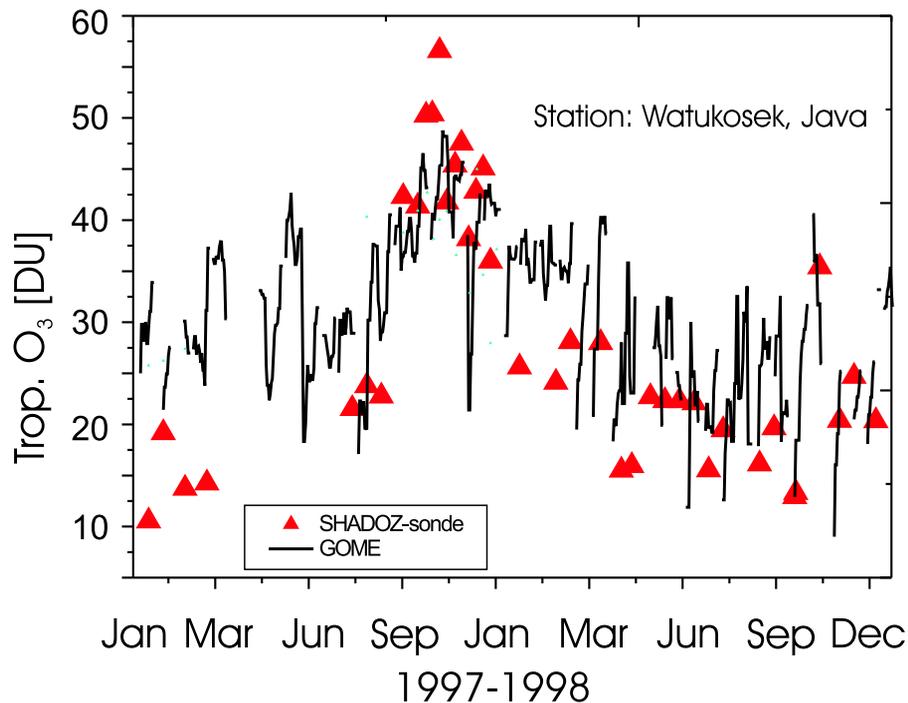
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Fig. 2. Time series of the tropospheric vertical column of ozone over Watukosek, Java, for the time period between 1997 and 1998 measured by both SHADOZ-O₃-sonde and GOME.

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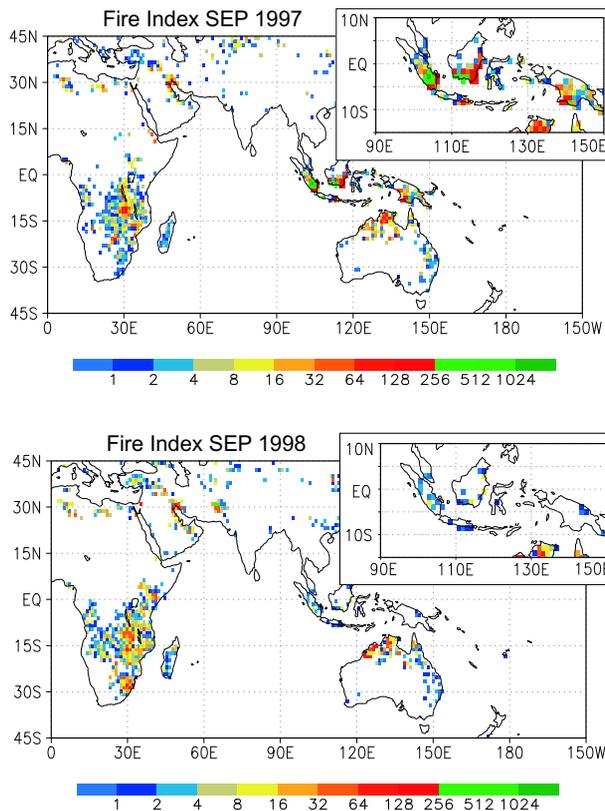


Fig. 3. Fire index as derived from ATSR hotspot data for September 1997 and September 1998.

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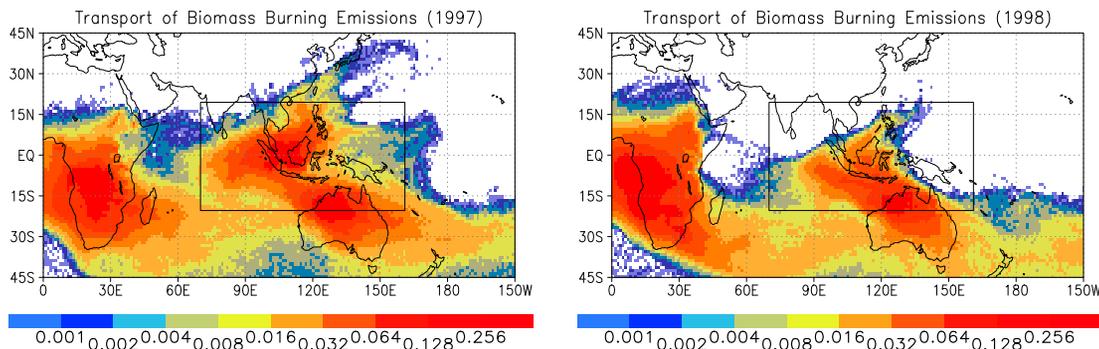


Fig. 4. Trajectory density of air masses being released from biomass burning in Africa, Australia and Indonesia for September 1997 and 1998.

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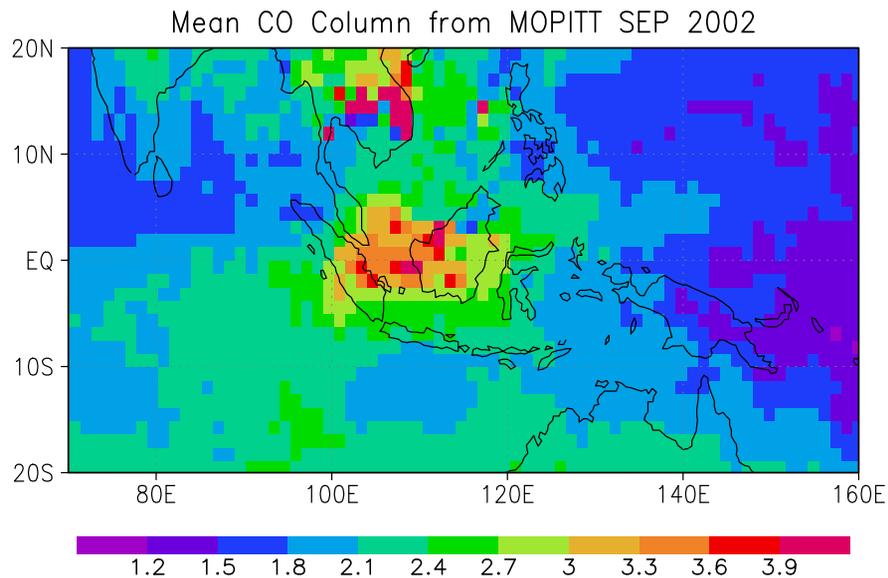


Fig. 5. Total column of CO in 10^{18} molecules cm^{-2} as measured by MOPITT performed in September 2002.

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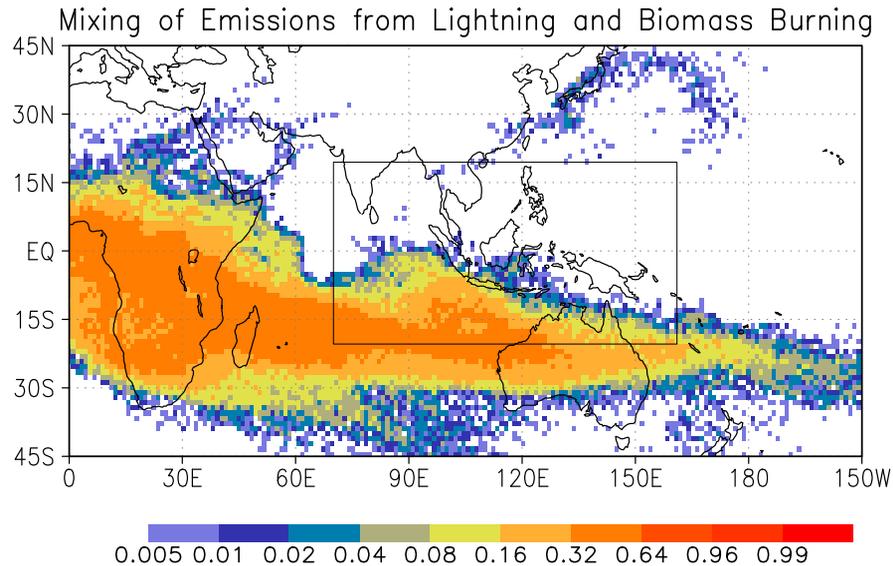
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Fig. 6. Intensity of mixing of air masses influenced by lightning activity over the Congo Basin with air masses emerging from biomass burning regions over Africa or Indonesia. Unit: Fraction of tropospheric bins in which mixing was taking place.

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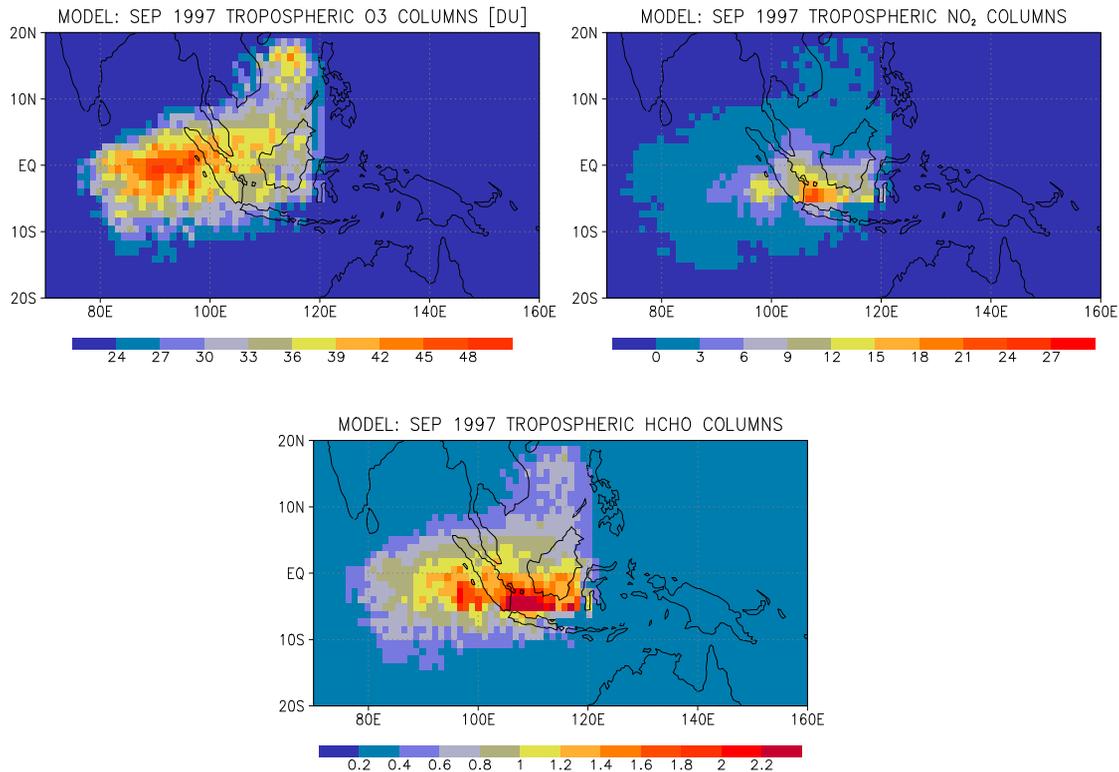
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Fig. 7. Modelled tropospheric column of monthly mean ozone (background of 21 DU is already added) in DU, tropospheric column of NO₂ in 10^{14} molecules cm⁻² (background of $2.5 \cdot 10^{14}$ molecules cm⁻² is already added) and tropospheric formaldehyde column in 10^{16} molecules cm⁻².

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