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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric O_3 over Indonesia during biomass burning events measured with GOME (Global Ozone Monitoring Experiment) and compared with trajectory analysis" *by* A. Ladstätter-Weißenmayer et al.

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General comments :

This paper presents the tropospheric ozone columns enhancement observed by GOME over Indonesia in September 1997 compared to the records from September 1998. The paper is written as follows. After a brief introduction, comes the description of the methodology and tools before discussing the results themselves based on the trajectory and the photochemical models. According to the introduction, the goal of the paper is to qualitatively and quantitatively understand the meteorological and anthropogenic contributions that led to the enormous increase of tropospheric ozone over Indonesia. Then the authors make use of the trajectory model Traj.x and of a photochemical model BRAPHO to achieve this goal.

Criticism: First, the paper misses reliable references and/or validation proof for these models before trusting in the results. Finally the conclusion is that the El Nino conditions in 1997 leading to extreme dryness and uncontrolled fires over Indonesia are responsible for such an enhancement. This is clearly not a new result deserving a publication focused only on that. Besides, the net production of ozone calculated with the BRAPHO model is very similar to the calculation made by Levine et al., 1999 as acknowledged by the authors. However, the authors mention an ozone enhancement of the same order of magnitude over the Indian Ocean between 10 and 20°S. As far as I know this broad maximum is clearly a new result that deserves further highlights. If such observations were reliable the present paper would be much better if focused on the explanation for such a broad enhancement that has not been reported before. The concluding remarks claiming that the mixing of air masses containing NOx from lightning over the Congo basin with air masses containing volatile organic compounds from biomass burning is responsible for such high tropospheric ozone columns then deserves further arguments and proofs. Giving them would make an excellent paper. I recommend the publication after major revisions. I would strongly suggest focusing and rebuilding the study more on the explanation and quantification of the broad ozone maximum over the Indian Ocean than on the enhancement over Indonesia only.

Answer: We thank the author for his fair comment on our study. His suggestions contributed to improvements of this publication and will certainly meliorate further studies on this issue.

Before taking stand to the comments we take the opportunity to correct a false statement made by the anonymous author:

“Besides, the net production of ozone calculated with the BRAPHO model is very similar to the calculation made by Levine et al., 1999 as acknowledged by the authors.“

Levine (1999) calculated the amount of carbon, NOx and particulate matter released by the Indonesian fires based on estimations on the burnt area. According to emission databases, the amount of ozone being produced by this event was derived. No model studies were applied.

Concerning the ozone increase due to mixing of different air masses:

The scope of this study is to analyse the distribution of tropospheric ozone being generated by biomass burning over Indonesia qualitatively and quantitatively. In addition the ozone maximum over the Indian Ocean can be explained by mixing of air masses originating from lightning over the Congo Basin and from biomass burning over Indonesia. A quantification of the ozone amount being generated by this process is behind the scope of this analysis since our work relies on the trajectory and chemistry models.

Now we will continue by answering the specific comments:

Specific comments:

Annotation: (1) The introduction is quite poor to describe the general context and to express the motivation and the interest for such a study. It gives too often too old references. The authors should seriously consider rewriting the introduction with the “new” goal of the paper and its content clearly presented. Many references concerning the previous studies on the 1997 ozone anomaly because of the wild fires are missing such as Duncan et al., JGR 2003, Kita et al., Atmos. Env., 2000, Hauglustaine et al., GRL 1999 for example.

Answer: For better readability, this annotation is divided into two steps (1.1) to (1.2).

Annotation: (1.1) Poor introduction describing the general context and to express the motivation of this study:

Answer: The objective of this study is to get insight into the meteorological and an-

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thropogenic conditions that lead to the enormous increase of tropospheric ozone during September 1997 over Indonesia. An additional focus is put onto the tropospheric ozone maximum located over the Indian Ocean between 10 and 20°S. According to our analysis, this ozone maximum is not directly caused by emissions from biomass burning over Indonesia. We claim, that this ozone amount is caused by the mixing of VOC enriched air masses released over Indonesia with NOx enriched air masses emerging the global lightning maximum over the African Congo Basin.

Annotation: (1.2) Too old references:

Answer: The suggested references to the work of Duncan et al., 2003, Kita et al., 2000 and Hauglustaine et al., 1999 are now cited in our introduction. All these studies are strongly linked to wildfires over Indonesia and give insight into:

- 3D-modelling aspects and sensitivity studies performed by applying the MOZART model (Hauglustaine et al., 1999) - Increase of total ozone over Indonesia during the October of El Nino years (Kita et al., 2000) - The impact of the Indonesia wildfires of 1997 on the tropospheric chemistry: Application of the GEOS-CHEM model on the Indonesian wildfire episode and analysis of the transport of air masses and radiative forcing resulting from gaseous and particle releases (Duncan et al., 2003).

All these above studies are to be seen as being complementary with respect to our study, although their individual aim differs from our work.

Nevertheless, references to the above publication is now included.

Kita, K., M. Fujiwara, S. Kawakami, Total ozone increase associated with forest fires over the Indonesian region and its relation to the El Nino-Southern oscillation, Atm. Env., 34, 2681-2690, 2000

Duncan B. N., R. V. Martin, A. C. Staudt, R. Yevich, and J. A. Logan, Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, J. Geophys. Res., 108 (D2), 4100, doi:10.1029/2002JD002378, 2003.

Duncan B. N., I. Bey, M. Chin, L. J. Mickley, T. D. Fairlie, R. V. Martin, H. Matsueda, Indonesian wildfires of 1997: Impact on tropospheric chemistry, J. Geophys. Res., 108 (D15), 4458, doi:10.1029/2002JD003195, 2003.

Hauglustaine, D. A., G. P. Brasseur, and J. S. Levine, A sensitivity simulation of tropospheric ozone changes due to the 1997 Indonesian fire emissions, Geophys. Res. Lett., 26(21), 3305-3308, 1999

Annotation: (2) The organization of the paper in terms of two distinct paragraphs: Methodology and results seems not very appropriate. It often leads to repetitions.

Answer: Splitting methodology and results into two sections is one way to structure this study and to our mind this is the best way. This structure offers the best readability since the reader learns about data and models (chapter “methodology and tools”) and – as a second step - gets insight into the results obtained by applying the methods presented before. We are of the opinion that the amount of repetitions resulting from our approach is tolerable.

Annotation: (3) Figure 1 is definitely too small. I suggest having two figures (one for 1997 and one for 1998) if necessary. The size of the maps should be the same as the ones in Fig. 7.

Answer: This reorganisation as suggested by the author of the annotations enhances the focus on the deduction of tropospheric ozone retrieved from GOME measurements during September 1997.

Therefore we implemented the proposed changes and organised figure 1 as follows:

Fig. 1 a): Tropospheric ozone (GOME) (Sep. 1997) (map as large as in Fig. 7) Fig. 1 b): Tropospheric NO2 (GOME) (Sep. 1997) Fig. 1 c): Formaldehyde column (GOME) (Sep. 1997)

Fig. 2 a): Tropospheric ozone (GOME) (Sep. 1998) Fig. 2 b): Tropospheric NO2 (GOME) (Sep. 1998) Fig. 2 c): Formaldehyde column (GOME) (Sep. 1998) (all figures equal in size)

Annotation: (4) Page 3110, line 2: The sentence is confusing. The SHADOZ network has been set up in 1998. The Java measurements from 1997 are not included in the SHADOZ database. Besides, the proper references are Kita et al., 2000 and Fujiwara et al., 2000 (both in Atmosph. Env.), not Fujiwara et al., 2003. The text and the legend of Figure 2 should make it clearer.

Answer: Ozone soundings performed at this location prior to 7th of January 1998 are not subject to the SHADOZ database. The reference has been corrected in section 2.1. In addition the sentence was slightly adjusted.

The correct citation for the 1997 ozone sonde data (which is now given in chapter 2.1 “Trace gas columns retrieved from GOME”) is:

K. Kita, M. Fujuwara, S. Kawakami, Total ozone increase associated with forest fires over the Indonesian region and its relation to the El Niño-Southern oscillation, Atmospheric Environment, Volume 34, Issue 17, p. 2681-2690, 2000.

Annotion: (5) Page 3110, line 16-20: The paragraph is not very clear. What such a multiannual climatology would be applied to?

Old Version: 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.

The old version of this paragraph may be understood falsely. It is now corrected:

New Version: When accounting for air pollution in tropical regions, lightning cannot be omitted since it is an important source of nitrogen oxides especially in the upper troposphere. In order to consider lightning-induced NOx emissions by the trajectory model, their temporal and spatial occurrence has to be evaluated. This is achieved by

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using a lightning climatology which is based on seven years of measurements of the Lighting Imaging Sensor (LIS) and the Optical Transient Detector (OTD) (Christian et al., 2003).

Annotation: (6) Paragraph 2.2: This paragraph would need a reference or further description and validation to argue that this trajectory model is well appropriate for the present study. Is the convection well calculated for example? Besides, it is not clear how the authors discriminate air masses influenced by fires or lightning. Is it based on altitudes criteria only? Further details are necessary in that paragraph. Concerning the box model BRAPHO, it is a shame that the only reference is in German.

Answer: Comparisons to FLEXTRA (Stohl, A. and P. Seibert, Accuracy of trajectories as determined from the conservation of meteorological tracers. Q. J. Roy. Met. Soc. 124, 1465-1484, 1998) have shown that Traj.x performs well in representing the mean atmospheric transport. This intercomparison was performed at IUP Bremen prior to this study.

Small scale processes like convection are extremely problematic to handle. In meteorological models (e. g. the ECMWF model) sub-scale processes like convection are parameterised (e. g. M. Tiedtke, A comprehensive mass flux scheme for cumulus parameterisation in large-scale models, Mon. Wea. Rev., 117, 1779-1800,1989). Within Traj.x convection is not explicitly accounted for. For this reason the large number of trajectories (230.000) was computed to depict the main transport paths of pollution caused by biomass burning and lightning over Africa, Australia and Indonesia.

For the analysis of the transport of air masses originating in a certain regime (lightning respectively biomass burning), trajectories were released close to their potential origin. That implies that trajectories representing biomass burning emissions are released close to the ground (50, 100, 150 and 200hP above the ground), those influenced by lightning are released at altitudes of 500 to 200 hPa. In this altitude region the maximum outflow of NOx out of the anvil takes place (Fehr, 2000).

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Fehr, T., Mesoskalige Modellierung der Produktion und des dreidimensionalen Transports Stickoxiden durch Gewitter, Fakultät für Physik der Ludwig-Maximilians-Universität München, 2000.

The latter information has been added to paragraph 3.1.

Concerning the chemical box model BRAPHO, an English publication became available in the meantime:

Meyer-Arnek, J., Ladstätter-Weißenmayer, A., Richter, A., Wittrock, F. and Burrows, J. P., A study of the trace gas columns of O3, NO2 and HCHO over Africa in September 1997, Faraday Discussions, DOI: 10.1039/b502106p, 2005.

Annotation: (7) Page 3112: I understand the problem in finding detailed trace gas measurements for this period over Indonesia. However, the authors should also consider taking into account the JAL (Japan airlines) measurements made between Japan and Australia in October 1997. See the Matsueda, GRL and JGR, 1999 papers for example. That seems more appropriate than the African measurements. In this paragraph I also don't understand the need of the reference Emmons et al., 2000.

Answer: The data collected during JAL flights between Singapore and Japan during October 1997 impressively underline the strong impact of the Indonesian fires on the CO, CO2 and methane concentrations (especially when compared to the aircraft measurements obtained in 1993 and 1996). When working on this study, we concluded that these measurements may be used for model validation, but not for its initialisation. For the latter, we were looking for a dataset describing the atmospheric composition, including the concentration of CO, methane and some NMHCs. Ideally the measurements applied for box model initialisation should have been performed close to fires but already outside the flames. This ensures that the impact of the fires on the atmosphere is accounted for, but the complicated flame chemistry is avoided. A dataset compliant with our requirements was compiled by Emmons et al. (2000) for savannah fires over Africa. To account for the specific conditions during the 1997 wildfire episode (enrich-

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ment of pollutants, burning peat and forest instead of savannah), the sensitivity study described in our study was perfomed. It finally leads to the initialisation concentrations shown in Table 1. When comparing the CO concentration derived by using our method with CO-measurement obtained from Matsueda (1999), an excellent agreement is to be stated.

During the PACE-5-campaign (Sawa et al., 1999), flights at low altitudes directly over burning area were conducted. CO levels have been between 500 and 3200 ppb. These measurements underline the extremely high CO values during October 1997.

Slight adjustments to the paper have been made in chapter 2.2 “Trajectory analysis and chemistry modelling”.

Matusueda, H. and Hisayuki, Y. I., Aircraft measurements of trace gases between Japan and Singapore in October of 1993, 1996 and 1997, Geophys. Res. Lett., 26, 16, p. 2413-2416, 1999.

Sawa, Y., Matsueda, H., Tsutsumi, Y., Jensen, J. B., Inoue, H. Y. and Makino, Y., Tropospheric carbon monoxide and hydrogen measurements over Kalimatan in Indonesia and northern Australia during October 1997, Geophys. Res. Lett., 26, 10, p. 1389-1392, 1999.

Annotation: (8) Page 3113, end of the paragraph: The authors should make the text clearer whether or not the BRAPHO model is able to take into account the production of NO2 by lightning for example.

Answer: BRAPHO is a tropospheric chemistry box model. Consequently the model has no capability to estimate the production of NOx due to lightning. Nevertheless the impact of lightning on the tropospheric chemistry can be evaluated by initialising BRAPHO with “post-lightning” NOx loadings and following the species’ chemical evolution along trajectories emerging regions of high lightning intensity. The chemistry model initialisation relies on assumptions published by Crawford et al. (2000). A num-

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ber of further studies dealing with this subject is available. In February/March 2005 the TROCCINOX campaign aiming in estimate the amount of lightning produced NOx has been conducted in Brazil. At that time, our study was already submitted to ACPD.

Regions being strongly influenced by lightning are identified by applying the 7-years-OTD/LIS climatology (Christian, 2000). 7 years of global lightning measurements are combined to a climatology on flash frequencies. Maximum lightning frequency is found of the Congo Basin.

These statements have been made clearer in the text by performing slight changes.

In Meyer-Arnek et al. (2005) BRAPHO was applied to model the impact of lightning of the tropospheric chemistry.

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 distribution of lightning as observed from space by the Optical Transient Detector, J. Geophys. Res., 108, 1, doi:10.1029/2002JD002347, 2003.

Crawford, J., D. Davis, J. Olson, G. Chen, S. Liu, H. Fuelberg, J. Hannan, Y. Kondo, B. Anderson, G. Gregory, G. Sachse, R. Talbot, A. Viggiano, B. Heikes, J. Snow, H. Singh, D. Blake, Evolution and chemical consequences of lightning-produced NOx observed in the North Atlantic upper troposphere, J. Geophys. Res., 105, 15, p. 19795-19805, 2000.

Annotation: (9) Page 3114, paragraph Results and Discussion: General comments and details: Globally this section is not very well organized. Many paragraphs of this section should be written in the introduction like the one starting line14 and the one starting line 28 for example. Line 24, I think it is Figure 1 or 2 instead of Figure 4. Paragraph starting line 19: This statement has been already mentioned. It is actually a strong argument. It would be valuable if the authors could further investigate the differences between 1997 and 1998 (in terms of circulation, lightning activity, etc.). It is explained

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in the previous section that trajectories are launched between the ground level and 200 hPa above (approximately 800 hPa then). Given that only statement and the figure 4, it seems difficult (or it is not convincing at least) to discuss the vertical distribution of the particles. Maybe Figure 4 is not the appropriate way to present and highlight the information. Page 3116, lines 12-13: This sentence clearly needs a reference to a figure. Is it Figure 6? This last paragraph deserves much more detailed to be convincing. I don't clearly see the good agreement with the GOME observations. That is the most interesting part of the paper but unfortunately that is the least investigated.

Answer: For better of readability, the annotation (9) is divided into annotations (9.1) to (9.6).

Annotation: (9.1) Many paragraphs of this section should be written in the introduction like the one starting line14 and the one starting line 28 for example (page 3114).

Answer: The paragraph you mentioned is called "Results and Discussion". The aim of this paragraph is to present results obtained in our study and to discuss them by placing them into a more general background. To our mind the optimum is not always met by address all additional relevant studies in the introduction.

Annotation: (9.2) Page 3114, line 24: I think it is Figure 1 or 2 instead of Figure 4.

Answer: The sentence you addressed was misleading and was slightly changed in the manuscript.

Annotation: (9.3) Page 3114, paragraph starting line 19: This statement has been already mentioned. It is actually a strong argument. It would be valuable if the authors could further investigate the differences between 1997 and 1998 (in terms of circulation? lightning activity? etc.)

Answer: Technically: This is clearly a recurrence of a statement already made on page 3110 in line 13. At this position it was made to emphasise the fact that the intensity of emissions outside Indonesia was (nearly) equal for both years.

When conducting further studies, they will have to concentrate on differences between 1997 and 1998 in terms of circulation and source strength. The latter certainly has to include differences in lightning activity.

Annotation: (9.4) Page 3114: It is explained in the previous section that trajectories are launched between the ground level and 200 hPa above (approximately 800 hPa then). Given that only statement and the figure 4, it seems difficult (or it is not convincing at least) to discuss the vertical distribution of the particles.

Answer: The projection of the three-dimensional trajectory density into the latitude-longitude-plane only gives insight into the horizontal transport pattern. To also visualise the vertical distribution, the longitude-altitude-projection of the trajectory density is to be included in our publication. To avoid up to three additional diagrams, we decided not to show this data in our study.

Annotation: (9.5) Page 3116, lines 12-13: This sentence clearly needs a reference to a figure. Is it Figure 6?

Answer: The reason, not to mention the reference to Figure 6 in this paragraph is, that the computation of the mixing (which shown in Figure 6) is explained in the following two paragraphs.

Annotation: (9.6) This last paragraph deserves much more detailed to be convincing. I don't clearly see the good agreement with the GOME observations. That is the most interesting part of the paper but unfortunately that is the least investigated.

Answer: When considering tropospheric ozone derived by applying the tropospheric excess method (as discussed in chapter 2.1), ozone columns southern of about 25°S are already disturbed by longitudinal inhomogeneities.

Mixing of different air masses was analysed in more detail than described in the publication. In our study transport is always treated as trajectory density. In case that non-zero trajectory densities originating in different source (biomass burning, lightning) are found in the same grid cell, mixing is expected to have taken place. Mixing of VOC and NOx enriched air masses is very likely to lead to ozone production at the particular position. The mixing of air masses released by biomass burning over Indonesia with air masses from lightning over the Congo Basin is shown in Fig. 8 (right). It is likely to explain the increased tropospheric ozone between 10 and 20° S eastern of 90° E. Accounting for mixing of air masses released from biomass burning and lightning over Africa leads to a very smooth distribution with respect to longitude (see Fig. 8 (left)).

When regarding the transport of emissions from the African continent onto the Indian Ocean, it has to be taken into account that this transport pattern is shifted southwards (compared to satellite observations) due to erroneous meteorological wind fields. This was discussed in Meyer-Arnek, 2005.

Conclusively, the enhanced ozone values over the Indian Ocean eastern of 90° E are due to mixing of air masses released from Indonesia with air masses released from lightning over the Congo Basin.

Meyer-Arnek, J., Ladstätter-Weißenmayer, A., Richter, A., Wittrock, F. and Burrows, J. P., A study of the trace gas columns of O3, NO2 and HCHO over Africa in September 1997, Faraday Discussions, DOI: 10.1039/b502106p, 2005.

Annotation: (10) Paragraph 3.2 Chemical modeling: I think the disagreement between modeled ozone and observed from GOME in the 10-20°S latitudinal band (Figure 7) should be further discussed. The agreement with Levine (1999) should be presented more like a validation of the BRAPHO model, not like a very new result.

Answer: The focus of this paper is shifted towards this question. Adjustments in the abstract and the introduction have been performed.

Annotation: (11) Last detail: is it fair to start the calculations on the 1st of September? According to Duncan et al. (2003), Indonesian wildfires of 1997 occurred mainly from the second week of September to the first week of November.

Answer: Our study relies on burning activity monitored by the ATSR instrument. This data reveals that enhanced burning activity (over Borneo) was already detected in the second half of August. Analysis from the TOMS instrument reveal that the increase of the aerosol index started during August 1997 (see Kita et al., 1999). Over Sumatra severe burning activity was first detected on 11th of September 1997. Compared to other years (e. g. 1998) burning activity was enhanced throughout August to October. The TOMS aerosol index shows total recovery to the pre-burning state not before the beginning of November 1997. Nevertheless - to our mind - it is reasonable to conduct a study starting on 1st and ending 30th of September 1997.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 3105, 2005.