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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling of trace gases from the 1998 North Central Mexico forest fire smoke plume, as measured over Phoenix" *by* V. R. Kotamarthi et al.

Anonymous Referee #1

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General Comments

This paper studies the trace gas composition and photochemical production of ozone (depending on H2O, soot, NOx, initial O3) in a smoke layer transported from North Central Mexico to Phoenix.

The paper addresses scientific questions relevant to the scope of ACP. However, the data base used in the paper is rather old (1998), and novel ideas or concepts of the paper are not pointed out clearly by the authors. The paper consists of many discrepancies and is not convincing from the data material and performed simulations. The paper is partly not well organized and the section with results is extremely short (1

page). The paper can perhaps be improved to be publishable in ACPD after a major revision, which however in this case would need an extensive effort from the authors.

Specific Comments

- No proper credit to related work:

The introduction to the topic is rather short (first 10 lines). A brief overview on previous experiments (model simulations) studying the trace gas composition in smoke layers and their chemical evolution during transport is missing. The reader doesn't get the feeling that the authors read the corresponding literature. The authors don't give proper credit to related work and don't identify their own new/original contribution clearly.

- The data material indicating the presence of a forest fire plume is rather confusing and not convincing:

Fig. 1a-b (TOMS) indicate influence from forest fires over the Phoenix area on 19 May however not on 20 May! Contrarily, measurements from the surface sites are shown from 20 May as representative for the forest fire event (Fig. 2-3): Add TOMS pictures and back trajectories for the whole period of interest 18-22 May for a more distinct separation of days with and without forest fire influence.

It is mentioned that CH3Cl values are enhanced on May 20 but not on other days of the week. Why is CH3Cl not enhanced on May 21, the day that was selected from the aircraft measurements as impacted by forest fire emissions? Or on May 19 (TOMS)?

Fig. 2: The mean from May 15 - June 15 shows the typical diurnal variation on a sunny day. How would the mean distribution for cloudy (rainy) days look like? Is there a difference to May 20? What about time series of UV, do they show unusual values on May 20 in comparison to the May-June period? Instead of Fig. 2-3 perhaps show time series of the different trace gases and UV for the whole May-June time period for the two selected sites (forest fire event more distinct visible?).

Fig. 4c (MHP) shows low (not high!) mixing ratios in the air mass influence by forest

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fire emissions (mentioned in the paper that high mixing ratios are typical for forest fire emissions).

It is mentioned that “an appreciable increase in NOx was measured on the day influenced by forest fire emissions”. Does this not indicate that the source is fresh (local)? The life time of NOx in the boundary layer is only a few hours and not 5 days (transport time from Mexico to Phoenix). It is suggested that NOx is only slowly converted to HNO3 due to the decrease in photochemical activity (smoke). However, is the time period in this case not too long?

The CO units in Fig. 2b are probably not correct (up to 1800 ppbv)?

- Scientific methods and assumptions not well justified:

Many of the values used for the model simulations are given without an uncertainty range or any evidence from measurements (e.g. ambient and initial conditions, Table 1 and 2). For more confident in the results, further sensitivity studies including these uncertainties should be performed (see comments below).

The vertical pathway of the plume trajectory must also be considered (change in temperature and humidity) for the box simulations.

- No complete description of calculations:

In Section 3.1 it is mentioned in one sentence that the plume is modeled as an entraining plume, ambient conditions are set to background conditions from measurements in this region and global-scale three dimensional chemical transport model results (MOZART). This step needs much more explanation: Why introduce MOZART, which MOZART results were used, how were the ambient conditions for Phoenix estimated and how representative are they along the 5-day transport pathway?, Does the plume change position in the vertical and how is the entrainment changed? What about humidity and temperature along the pathway?

How large is the dilution rate and how was it estimated?

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In Section 3.2 correlations between CH3Cl and CO, hydrocarbons and NOx from the literature are used. No uncertainties in the estimates are mentioned and if emissions from African savanna fires are representative for Mexican fires. The estimate of the initial CH3Cl conditions (2.2 ppbv), which all estimates refer to, is probably connected to large uncertainties which is not discussed (Fig. 7, left).

- Results of model simulations are not convincing and confusing:

Fig. 10: The model simulates high ozone mixing ratios after 5 days transport. However, the observations in the Phoenix area show very low ozone mixing ratios during the passage of the forest fire plume. How can this disagreement be explained? How do the other model results fit with the presented trace gas measurements from the surface sites and aircraft? If you are confident with your model, these results should fit or not?

During the ICARTT experiment (http://www.al.noaa.gov/ICARTT/) in 2004 the observed layers from forest fire emissions (aged and uplifted) were frequently very dry: In Fig. 9 and 10 perhaps add a run with low humidity and with soot. What is the main difference to the previous simulations?

Fig. 12: Final ozone (and also already after 1 day) seems to be rather independent on the initial ozone mixing ratio. How can this be explained?

- The paper is unbalanced and partly not well organized:

Sections “Introduction” (related work) and “Results” (and Conclusions) are too short.

In Section 1 and 2 a lot of information concerning project, sites, platforms, data, selected days is repeated several times. These sections should be reduced and repetitions avoided (condense in Section 2).

Technical Corrections

Abstract:

(avoid repetitions: Mexico (source region) and Phoenix mentioned in three successive

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sentences)

Page 3228, line 6-7: change “influenced by transport” to influenced by long-range transport of emissions

Line 7: cut “in central and southern Mexico” (source region mentioned in next sentence)

Line 9: change “origin of the air” to origin of the polluted air mass

Line 11: change “photochemistry over a five-day” to photochemistry over the five-day

Line 11: cut “from the source region to Phoenix” (pathway already mentioned in last sentence)

Line 15: change “shown to be minimal.” to shown to be minimal after 5 days since emission release.

1. Introduction:

Line 19: “in the immediate vicinity”: not only immediate vicinity, also mention that long-range transport of forest fire emissions is important (case described in the paper)

Page 3229, line 1: “The impact of such forest fires on ozone mixing ratios”: describe also connection (chem. reactions in words) between forest fire emissions (ozone pre-cursors) and ozone

Line 4: change “during the year 1998” to in May and June 1998

Line 4: change “over Phoenix” to over the Phoenix area

Line 4: “Phoenix, Arizona”: add longitude and latitude

Line 4: change “to sample air” to to sample a polluted air mass

Line 5-7: cut “of an extensive six-week study conducted in May and June, in and around Phoenix, as a part” (already mentioned above)

Line 8-13: “Measurements of the flow (Doskey et al., 2000)”: too much detail, move

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this part into Section 2 (however avoid repetitions)

Line 19-23: Do not list all the single days (not important for this paper since different location): compress all sentences to “Peppler et al. (2000) reported an increase in ozone (above 80 ppb), large particles (>10 nm) and aerosol single scattering albedos at the SGP site on days impacted by the forest fire event.”

Line 23: change “It is” to However, it is

Line 26: “highly polluted urban areas”: mention which (Houston...?)

Line 26: change “are directly” to are located directly

Line 26-27: “During May..”: cut sentence, already mentioned in line 14-15

Line 27: “Here we..”: begin with new paragraph

Page 3230, line 2: “Figures 1a...: don’t begin with a new paragraph

Line 7: “The TOMS data..: too detailed, move to figure legend

Line 10: “The aerosol index..”: don’t begin with a new paragraph

Line 10-11: “do not necessarily represent smoke aerosols”: what else could the source be?

Line 16-22: “Measurements made on... support our conclusions”: move all to section 2 (however avoid repetitions) and replace by: “In the next section we combine surface and airborne measurements in the Phoenix area to study the impact of this North Central Mexico forest fire plume on atmospheric chemistry in the area of interest.”

2. Measurements over Phoenix of the North Central Mexico forest fire plume

Line 27: change “Usury” to Usery

Line 27: change “center of the Phoenix” to center of Phoenix

Line 27: “surface site at Usery Pass”: add longitude, latitude and altitude, remote?

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Page 3231, line 2: http-link to Phoenix 1998 data doesn't work. Add a table with instrument techniques and accuracy, detection limit...

Line 4: "The focus here is on two days in May 1998...": Add here that the days with forest fire influence was 20 and 21 May, however for comparison also the day after the pollution event (22 May) and the average from 15 May to 12 June were analysed and presented.

Line 5-6: "in and around Phoenix.": add "In addition, measurements made by the ADEQ at its Central Phoenix site, also known as the Phoenix Supersite, will be used. This site is located in the city center (?)... and measures O3, CO, PM10...

Line 7-8: "20 May...Usery..": Move sentence to line 23 (separate the Usery and Phoenix site), avoid repetitions

Line 7: "CH3Cl": write out methyl chloride. Mention that it is a tracer for biomass burning and give some references.

Line 8-10: "The Central Phoenix site...": compress to "The Central Phoenix site showed remarkable differences on 20 May versus other days in the selected May and June period."

Line 14-15: "uncharacteristic of a urban source dominated by local pollution": could this shape (broad and flat) also be characteristic of a cloudy (rainy) day?

Line 15-16: cut "This observation suggests that the ozone..." (this statement is too early)

Line 19-20: "Interestingly, an appreciable increase in NOx...": why not shown?

Line 20-21: "This additional NOx...": can fresh emissions from local sources be excluded?

Line 25: "The ultraviolet light...": was UV exceptional low on 20 May in comparison to all other days (15 May to 12 June, show in plot)?

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Line 27: change “on 21 May and 22 May.” to on 21 May (forest fire event) and 22 May (no forest fire event).

Line 29: change “poor visibility” to poor visibility due to the presence of the forest fire plume (?)

Page 3232, line 7-8: “The water mixing ratios...”: explanation for the high/low humidity on 21 and 22 May?

Line 8-10: “The mixing ratios for methyl...”: “similar different” not correct, since MHP is elevated on 22 May in comparison to 21 May (Fig. 4c). Explanation? Doesn’t agree with observations of Lee et al.?

3. Model calculations

Change title to “Model Descriptions and Calculations”

Line 19-20: “calculations were performed... 20 May”: add trajectories for all days within the period 18-22 May for more convincing results (separating days with and without forest fire influence)

Line 21: “(MM5 V.3)”: give some references

Page 3233, line 7-9: Back trajectories were calculated from Phoenix and the results were then plotted in groups of trajectories. Does this mean that back trajectories were started from different altitudes (surface - 300 hPa) over Phoenix? Furthermore, for the box model simulations described later it is essential to also include the vertical pathway of the selected trajectory (change in temperature and humidity impacts chemical reactions?).

Line 19: “Gelinas and Walton (1979)”: 1974 in reference list

Line 21: “green... blue”: not visible in Fig. 6 (black and white)

Line 22-25: “The plume is thus modeled as an entraining plume, ambient conditions

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are set to background conditions from measurements in this region and ... model results (MOZART).": need much more explanation: (1) Why introduce MOZART?, (2) What kind of MOZART results are used: tropical southern Pacific? (3) How can measurements in the Phoenix region be representative for the ambient conditions during the whole transport period? (4) How were the ambient conditions estimated (Table 2)? (5) For which altitude?

3.2 Initial conditions for all key trace gases

Change title to "Initial and ambient conditions for all key trace gases"

Page 3234, line 5: "CH3Cl": also other sources than biomass burning? Long life time like CO?

Line 8-9: "peak values of 900... 20 May... 600... rest of the week": why is CH3Cl not enhanced on 21 May, the day when the aircraft measured the plume?

Line 11: "dilution rates": which dilution rate was used in Fig. 7? How was it estimated?

Line 12-13: "initial conditions, 2.2 ppbv": is this mixing ratio not connected to large uncertainties depending on which hour is selected as start (steep slope to the left in Fig. 7)?

Line 15-19: Are the emission indices used in this paper (from African savanna fires) representative for the burning material in Mexico?

Line 20-22: cut "Thus, we have a fairly robust estimate of the initial conditions... given us confidence in the results based on this model"

Line 23: "Table 1": give some uncertainty ranges

Line 24: "Table 2": how were the ambient conditions estimated? For which altitude? Uncertainty ranges?

Page 3236, line 2-3: "The aerosol concentration... 5000... at surface to 1... at... 10

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km": Reference?

Line 6-7: "Figures 8a and b": also give uncertainty range

4. Results

Line 17: "NOx at 200 pptv in the ambient air": how was ambient NOx estimated?

Line 24: change "Usury" to Usery

Line 24-25: "Gaffney et al., 2001": in reference list 2002

Line 26-27: "and the case with increased H2O mixing ratios": add "and no soot"

Page 3237, line 8: How do the results look like if a dry layer (1000 ppm) with soot is simulated?

Line 15: "The final difference...": add after how many days

Line 17: "is about 15 ppbv"; add after how many days

Page 3238, line 6-7: "continental rural background... 200 pptv... NOx": for which altitude? Is this value not too low?

5. Conclusions

Rather short. More discussion about uncertainties can be included.

Line 25-26: "methylhydroperoxide": no increase seen in Fig. 4c

Table 1: How is the initial ozone mixing ratio in the plume estimated? Use the same order of the trace gases as in Table 2. Are not also humidity and temperature important model parameters (include in Table 1 and 2)?

Table 2: Is Toluene missing?

Figures:

Before Figure 1 you could add a map showing the fires (ATSR hot spots) over Mexico

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for May 1998: see http://dup.esrin.esa.int/ionia/wfa

Fig. 1: add also TOMS pictures for 18, 21 and 22 May and the position of Phoenix. Add longitude und latitude in the map. Explain in the text why high AOD values are observed mainly over the ocean.

Fig. 2b: check the unit of the CO-axis (values up to 1800 ppbv are not realistic). May 20 has not exceptional CO mixing ratios in comparison to average night values in Phoenix. In the legend, change “ozone” to CO.

Fig. 2c: In the legend, change “ozone” to PM10.

Fig. 3: Add the day of the observations.

Fig. 4a-d: the difference between the two days is probably better visible in a vertical profile plot (mixing ratio versus altitude). Is the vertical extension of the plume visible?

Fig. 4a: “triangles”: diamonds? “21 May (black line and circles)”: grey line and diamonds

Fig. 5: In addition, it would be useful to have trajectories for the whole period of interest: 18-22 May. Change “800 mb” to 850 mb (used in the text) Reason for wave pattern of surface trajectories? Backward trajectories for which time period? Cut two last sentences in the legend.

Fig. 10: The model simulates high ozone mixing ratios after 5 days transport. However, the observations in the Phoenix area show very low ozone mixing ratios during the passage of the forest fire plume. How can these discrepancies be explained?

Fig. 12: Final ozone (and also already after 1 day) seems to be rather independent on the initial ozone mixing ratio. How can this be explained?

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