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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.

V. R. Kotamarthi et al.

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Response to reviewer comments on "Modeling of trace gases from the 1998 North Central Mexico forest fire plume, as measured over Phoenix"

General Response: Note: The full text of this response along with the all the figures included in a pdf file can be obtained from http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/response.pdf

We wish to thank the two anonymous reviewers for their extensive and often exhausting comments. Since the comments made by both the reviewers are similar and reviewer one had a number of specific comments, we have addressed the general concerns of both the reviewers at the start and addressed specific questions of reviewer 1, which

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include all the comments made by reviewer 2. The general theme of the comments being that there is not enough data to support the modeling and the paper is not comprehensive enough. First, it needs to be clarified that this is not a paper discussing forest fires in general. Our focus is more on what if any are the consequences of a forest fire plume mixing with an urban mix of atmospheric pollutants. We have not cited many references as there are not many papers published regarding this aspect. The references cited by reviewer 2 are now included in the introduction. As for a general discussion of forest fires and their measurements, it would be inappropriate, as the Phoenix measurements are not designed for measuring forest fires. As for the evidence for a forest fire derived smoke plume over Phoenix: the primary problem is having no measurements from aircraft for the 20th, when the smoke plume was visually 'observed' by the participating DOE field experimental scientists and resulted in the cancellation of G-1 based measurements for the day. This eyewitness account is our primary strong evidence for the plume. The chemical and meteorological data was analyzed for supporting evidence and when a reliable supporting factor was found, we showed it in the paper. a) TOMS: That there were forest fires in southern and western Mexico during this period is beyond any doubt. The TOMS figures are mainly used to locate the source of the forest fires and show the approximate time period over which the plumes from these fires approached Southwestern USA and in particular the Phoenix, AZ area. As explained by Torres et al. (1998) the aerosol optical depth data is assembled using 2 days worth of data and the date of the aerosol optical depth file from NASA signifies the day on which a 'majority' of the data is obtained. There is also a question of the local 'overpass' time and the grid size of the data (1 degree x 1 degree). Given all these variables to use the TOMS maps for anything other than a qualitative description of the event would be farfetched. The following are maps generated from six days of TOMS AOD data. The first figure1a (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure1a.pdf) starts on May 18, figure 1b (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure1b.pdf) is for May 19th, followed by figure 1c

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(http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure1c.pdf)for May 20th, figure 1d (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure41d.pdf) for May 21st,figure 1e (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure1e.pdf) is for May 22rd and figure1d for May 23rd (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure1f.pdf) The location of Phoenix is marked with a triangle in the portion of the map corresponding to Arizona in the southwest corner of the US map. It can be qualitatively inferred from the figure that the aerosol optical thickness for 18 and 19th is higher to the south of the US border and the optical depth over the southwestern USA starts increasing from the 20th and 21st, 22nd and 23rd show increased optical depths. As per eyewitness account the primary plume over Phoenix was observed on the 20th of May and measurements made on the 21st as explained in the paper continue to show the effect of the smoke plume. The fact that TOMS shows a plume over Phoenix on the 21st is not surprising given the fact that the local 'overpass' occurs around noon and the event was observed later in the day on the 20th. It is obvious from the above figures that May 18 and 19th didn't have any of the forest fires influence over Phoenix and May 20th/ 21st and possible 22nd have indications of increased optical depths. These TOMS pictures (in a different color scheme and for the entire globe) are available readily on the TOMS website. I don't see the point in reproducing them in the paper. b) Back Trajectories: The trajectory plots shown here are for a few days before and after the incident we modeled. Before 20th the flow was predominantly from the west both in the lower and upper atmosphere. Figure 2a (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure2a.pdf)shows the back trajectories reaching Phoenix on the 17th, figure 2b for the trajectories reaching on the 19th (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure2b.pdf) , figure 2c shows trajectories reaching on the 21st (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure2c.pdf) and figure 2d shows the trajectories for the 22nd (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure2d.pdf). The 21st and 22nd trajectories show

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flow from the southeast in the PBL and from the southwest and over the ocean in the upper atmosphere. The modeled day 20th as shown in the paper has flow from the south and central portions of Mexico into Phoenix and upper layers have a westerly flow over the oceans.

c) CH3Cl: As for CH3Cl: Methyl chloride was measured from canister samples collected for hydrocarbon measurements by Dr. Paul Doskey of Argonne based on a request for constraining the model. Normally about 6 samples were collected at the top of the hours during the time periods when G-1 was operating. Data on CH3Cl for all the dates and times the canisters were sampled for CH3Cl is sown in figure 3(http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure3.pdf). It is very obvious from the figure that there is no question the CH3Cl is higher on May 20th compared to all other days at the Usery Pass surface site. In fact, the idea for the paper originated after we started looking at the CH3Cl data.

Fig 2: there were no clouds on May 20th. As per eyewitness accounts from the participating scientists a thick layer of smoke was observed over Phoenix on that day. Most of the data from the Usery pass was discussed by Gaffney et al., (2002). In particular figure 5 shows the UVB for the entire 3 week period of the experiment and it is very obvious from Figure 5 (a) in Gaffney et al., (2002) that the UVB for day 141 is approximately 30% lower than any other day. I added a reference to the paper and the figure in the text in page 6 line 26, replacing the figure not shown with the above citation.

Fg4c: MHP being lower and not higher on 21st. To address this and other questions related to peroxide, we reanalyzed all the data collected from G-1 over Phoenix. Figure 4 (a) (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure4a.pdf) shows the H2O2 and Fig 4(b) (http://gonzalo.er.anl.gov/POST/acpd-2006-0006-supporting-material/figure4b.pdf) shows the MHP. The data was collected at various altitudes as shown in figure 4 in the paper and over a 2-hr period for each flight. As a result the data spread along the x-axis is limited due to the long 20 day scale used. The H2O2 obviously for day 141 is much higher than the rest of the days. The MHP

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on the other hand doesn't show any trend and actually is higher than H2O2 on several of the days during the early part of June. At this point we have no explanation for this and it seems prudent to drop the MHP figure from our paper. We have removed the figure 4(c) from the discussion. Li et al (1998) have seen increases in MHP in sampled forest fire plumes in the mid to upper troposphere and away from the source areas. Apparently, we don't see this in this data set.

NOX being higher: I agree that local sources of NOx probably will predominate the measured NOx plume at Usery pass. Gaffney et al., (2002) have hypothesized that regular nighttime plumes of high NOx were sampled at the Usery pass and probably is a leading cause for the increased NOx. We didn't suggest that NOx lifetime is of the order of 5 days in the PBL, rather in this particular plume conversions rates are lower due to the smoke layer and will probably retain more NOx than a typical urban PBL plume. The increase in NOX at Usery pass may be lower conversion rates locally and not due to the transport from Mexico. This sentence in the paper is now corrected to say this more clearly.

CO in urban areas does range in the 0.5 to 3 ppm and the units are correct.

-Scientific methods not well justified: This is as claimed in the title a modeling study to understand the behavior of a smoke plume primarily retained in the PBL over a few days. There are uncertainties inherent in this type of modeling activity, the primary idea being to speculate on possible mechanisms and outcomes of such events such that future measurement campaigns can focus on these uncertainties, if necessary. For this to be a valid exercise, there should be certain constraints on the modeling, clear statements of assumptions made and an evaluation of the sensitivity of these assumptions. The key point we are trying to make is that the smoke layer reduces photochemical activity in the PBL and hence the time scales of the some of the typical chemical processes are extended and could have potentially different impacts on receptor regions as compared to a case when the smoke is not present. We have explored the sensitivity of our calculations to the parameters central to this issue. The primary uncertainty

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is the PBL is ‘dilution’ as people dealing with urban air quality are well aware, the initial conditions of the plume and concentration of oxidants and their precursors. Our primary focus is as a result dealing with the dilution rates and initial conditions. The CH3Cl measurement at Phoenix provided a strong constraint for generating a profile for the initial conditions, when dilution rate was defined based on expected horizontal mixing rates in the PBL. The dilution rate was set using an initial size of 90 km for the plume and then assuming a growth rate for the plume width as a function of scale-dependent diffusion rate defined from literature as 7.0E08 m2/sec (Walton, 1972) . The width of the plume at anytime is calculated according to Gelinas and Walton (1974). The entrainment factor is factor is defined as the ratio of plume width to diffusion rate and used for calculating the mixing between the plume and ambient air. Since dilution is primarily the single factor controlling the CH3Cl , the final measured CH3Cl is a ‘strong’ and ‘binding’ constraint on both the initial concentration of CH3Cl if the dilution rates are correct. Since there is no independent control available for determining the dilution rates, the time evolution of CH3Cl is considered here to provide a constraint on the dilution rates and its initial concentrations. This fact is critical for the rest of the paper as it is used to derive the initial concentrations of oxidants and their precursors based on available measurements. This is unlike an upper tropospheric trajectory calculation in the sense that mixing plays an important role in the PBL and an ‘air-parcel’ has an integral conserved entity, as modeled in the upper troposphere/lower stratosphere, doesn’t really exist in the lower atmosphere beyond an hour. I don’t see the point in analyzing the uncertainty of every initial condition used unless there is anything tat would contribute to enhancing or changing the message of the paper i.e. ‘soot layer leads to lower photochemical activity’ and changes the evolution of ozone in the plume as a function of time. Some sensitivity runs were conducted for VOC sensitivity by changing the initial CH4 used for calibration and the effects on our computed final results were found to be minimal. Lot of the stuff that is being asked here is what any conscientious researcher would do and putting every calculation made into the paper would make this into a really lengthy paper in comparison to the points we are trying to

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make. Vertical path of the plume: The plume is considered to be occupying the entire PBL. There is no vertical profile in the plume, it is just a box going along the surface.

-No Complete description of the calculations:

This is not correct. Please read section 3.1 paragraph one again. It tells the reader what the initial size of the plume is and how the dilution rates were set (look up the references Gelinas and Walton for how to do it and Gifford for calculating the dilution rates in the PBL). These are standard and well known methods and there is no need to explain over again what these are. However, we are adding some additional text to describe the mixing rate used for the plume. Again, there is some confusion here of relating this to some upper air 'air-parcel', this is not an air-parcel in the traditional upper troposphere sense. The size of the plume is represented in figure 6 and tells the reader the final width of the plume is 500km. I don't understand the point about ambient conditions. They are listed in Table 2. Where is the mystery? The path traveled is a few grid points in MOZART, there is not much variability over these 3 grid points or over the few days to bother changing the ambient conditions every hour. The model ambient conditions are clearly stated. The water vapor concentrations are listed in figure 9, 10 and 13 and as explained in Section 4 Results, paragraph 1, they are held constant at these values for various sensitivity runs discussed in the model.

Fig 10: Figure 10 represents the plume as it would enter Phoenix and there is no mixing from local emissions. It would be very easy to include high fresh NO (the simulation stops early in the morning) from local sources and titrate the ozone out of the model. That is not the point of the paper, we are not trying to reproduce the observed results in an urban plume. The point is that the plume is coming into the urban area not with its peak ozone, but almost 10-15 ppbv lower than the peak ozone produced during its history. About using lower water vapor: 1000 ppm in the PBL is quite low and we have the case represented in our studies. This again is not upper tropospheric plume, it is entirely in the PBL. Figure 12: Yes, much of the ozone production seems to be occurring in the first few days. As the VOCs and NOX in the plume decrease rapidly in

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the plume the potential for producing ozone later in the plume history decreases. The NOX/VOC conditions determine the ozone that can equilibrate with the mix. It happens to be nearly the same with differing ozone initial conditions is not surprising, given the large initial concentrations of NOx/VOC into the plume. -The paper is not well organized. There are more details in the paper in sections were the data is not presented before or to explain the field measurement campaign. On one hand there is a complaint that there is not enough explanation of some items and where there is explanation it is deemed to be very extensive. The results and conclusions describe and summarize the results. As long as it conveys the summary of the paper and describes the results with the use of the figures, we see no point in adding extra words. This doesn't have to be lengthy paper for presenting the work and explaining our conclusions. Technical Corrections: Avoid mentioning Phoenix: If we remove Phoenix, then we have to say source/receptor etc. to make sense and don't see the need to make the change. In the rest of response, only substantial comments are addressed. Corrections related to the style (or usage of English) where a change doesn't materially affect what we want to say is not addressed. The text was reviewed by a technical editor and most of the corrections to English suggested seem to be the personal preference of the referee.

Comments on Line 7 to Line15: Corrected. 1. Introduction: 2. Line 19 ; Changed. Page 3229: This would add to the text for no particular reason. Any one reading ACPD would know how ozone is produced.

Line 4: change 'during the year 1998" to May and June 1998: done Line 4: Phoenix, AZ add long/lat - added 8-13: No changes made. Lines 19-23: Doesn't improve the presentation or clear any confusion. Choice of presentation style. No changes made.

Line 23: No change Line 26: Highly polluted urban areas : "Houston" added to paper Line 10-11: "do not necessarily represent" : Background inorganic and secondary organic aerosols.

Line 27: Change Usury to Usery: done Line 27: "surface site at Usery Pass" : add

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longitude and latitude - done 3321: Link is corrected. The additional information requested is beyond the scope of the paper. See Gaffney et al. (2002) for some of the methods. Line 4: “add that Ĕ” Done Line 5-6 : add details on supersite : Done Line 7: CH3Cl - spell out CH3Cl and add citation for it being a biomass burning tracer: Added a reference to CH3Cl emissions from biomass burning. Line 14-15: No. We know there was a smoke plume that day. Line 19-20: The increased NOx is here being claimed to be a result of lower photochemical activity and from local sources, not longrange transport. Line 25: UV plot - discussed above in the general response. Line 27: yes Line 29: Yes. Due to forest fire smoke. Page 3232, Line 7-8: The forest fire plume seems to have lot more water vapor than the usual dry Phoenix air. The forest fires are known to cause increase in water vapor from the heat of the fire. Line 8-10 : See the general response section. This figure is now deleted from the text of the paper. Line 19-20: see the general response above. Adding all these to the paper will not add any information to the paper. Line 21: Added Page 3233: Yes, the back trajectories were calculated from Phoenix starting at various altitudes as described. Only the PBL trajectories were used for the calculation described in the text. The entire PBL was modeled as one box, there are now profiles of temperature and humidity in this box. Line 19: Added Line 21: corrected to correspond to the text. Line 22-25: Look at the general response section above. 3.2 Initial conditions - changed Page 3234: Oceans are probably the primary source of CH3Cl, giving about 620-700 pptv of continental background concentration. It has a lifetime of approximately 1.5 years in the troposphere, much longer than CO lifetimes. Line 11: See the general discussion. Line 12-13: No really. CH3Cl has a long lifetime and is nearly inert. The change in concentration we are calculating is primarily based on dilution and a few hours more or less is not going to change this number drastically. Line15-19: Much of the vegetation over Central Mexico is grass and brush and this is the only data set which has an approximately similar profile. Line 20-22: As explained in the general discussion this is a fairly robust estimate in the absence of direct measurements. Line 23: Uncertainty ranges: These numbers were generated from the papers cited in the manuscript. The uncertainties are similar to

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the measurements. There is no method for making independent uncertainty estimates for this case. Line 24: Look at section 3.7 line 24-25. All the conditions are for PBL. Page 3236 : added Results section comments: Line 17: Look at section 3.7 line 24-25. All the conditions are for PBL Line 24: Changed Line 24-25: Corrected Line 26-27: Done Line 8/Page 3237: The behavior of the plume would be between that shown for 1000 ppm no soot and 9000 ppm no soot. Line 15: after 5 days. Added to text. Line 17: after 2 days. Added to text. Page 3238: This is the background conditions along regions of northwestern Mexico and southern Arizona, fairly remote and no extensive anthropogenic sources of NOx.

Conclusions: Additional discussion on uncertainties added. Line 25-26: methyhdroperoxide. Dropped from manuscript. See the general response. Table 2: Toluene added to the table. Table 3: The initial conditions were based on measurements near forest fires from literature. However we have shown that the final ozone in the plume is fairly insensitive to this IC for ozone. Figures : Adding ATSR pictures: This is not really central to the topic of the paper. Fig 1; add other TOMs pictures: see the general response Fig 2b: CO 1.8 ppm is not uncommon in urban areas. Fig 2c: corrected Fig 3: corrected Fig 4a-d: The resolution of the data and the flight plan doesn't provide such detail. Fig 4a: corrected Fig 5: See the general response above. Figure 10: We didn't simulate any NOx mixing into the plume in the Phoenix area. Additional fresh NOx in the early morning could titrate much of the ozone and keep it low during the subsequent hours as a result of the smoke layer. Figure 12: See the general response above.

Reference: Gelinas, R. J. and J. J. Walton (1974): Dynamic-Kinetic evolution of a single plume of interacting species. J. Atmos. Sci, 31, 1807-1813.

Walton, J. J. (1973): Scale-dependent diffusion. J. Applied Meteor., 12, 547-549.

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 3227, 2006.