Anonymous Referee #2

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General Comments:

This paper describes an investigation of the contribution of flare emissions to local and regional air quality in Mexico. The relevance of this study extends beyond this specific application since flare emissions are poorly understood and contribute significant uncertainty to air quality modeling in areas with intensive petrochemical operations. The introduction does a good job describing the relevance of this work, reviewing previous studies on flame emissions, and giving background on the Tula area and its emission activities. While Sections 1 and 2 read well and are clearly presented the later results sections are rather disjointed, rambling, and difficult to follow. Also, gramatical and word choice errors are more prevalent in these latter sections. There are many parts of the paper that should be clarified and there are many technical corrections that are needed. I think that if these questions are addressed and the text is clarified in the indicated sections along with thorough editing, the paper may be acceptable for publication.

Specific Comments:

1) The study uses a CFD combustion model to compute emissions from the Tula flares. By necessity, the set up of this model is largely prescribed with a single constant wind profile and gas stream. The composition fractions of the gas stream are specified (page 15184) but the mass flux is not given or described as to how it was derived. I would think that given the difficulties in calculating emissions that agree with other estimates, particularly for soot, that some sensitivity model runs should be made where winds and gas components and fluxes are varied. Perhaps this is part of the other paper that is repeated mentioned; if so, this should be mentioned along with a clear description of what the other paper is about and how in compliments this paper.

With respect to the mass flux, please see second paragraph of reply to point 2.

Regarding Referee's comments concerning the sensitivity tests, a paragraph is added to section 2.1.1 pg15185ln22 in order to clarify the difference between these works. It reads: "In that paper, the influence of wind field, gas composition and fuel mass flow on the emission rate of combustion by-products is being investigated. In addition, the Gas Research Institute chemical mechanism of natural gas, GRI3.0, that includes NOx formation is being considered for the chemistry of hydrocarbons. Results of that paper, serve as the basis for the combustion simulation of this work. For instance, the chemical mechanism of Lutz (Lutz et. al, 1988) and Leung (Leung et al, 19991) were tested for C1-C3 hydrocarbon combustion; however, a better performance and numerical stability was obtained with the Glassman mechanism. Improvement of the oxidation term of the soot model and a 3D set up are part of that paper as well."

2) The description of the emission calculations for the slices in Section 3.1.1 is quite confusing. For example, I don't understand why there is inflow and outflow along the slice. Isn't the flow through the slice? Also, what are the quadrants? Perhaps another diagram would help me visualize the process. I am also confused about how the emissions from the

single modeled flare are scaled to the three flares. In Section 3.1.2 (pg15189) the IMPei estimates for the 3 flares is used to extrapolate the model calculations of one flare to all 3. It is not described how this is done, but from the values given it seem that the model is taken to represent F1. However, on pg15191 a similar extrapolation for soot seems to be inconsistent. If the model is assumed to represent F1 as for SO2 then the total for the 3 flares would be about 0.88 g/s which is about an order of magnitude greater. It seems that in this case the modeled flare is assumed to represent F3. Please explain and justify these calculations.

With respect to the emission calculations for the slices we removed the paragraphs between ln23 and ln10, and added the following paragraph to section 3.1.1on pg15187ln23. "Buoyancy and air entrainment vortices are relevant for open flames, particularly in the near field. Thus, the flow field is different outside and inside the plume. The velocity field is captured in a slice at a particular height of the plume. As a result of air entrainment, wind vectors not aligned with the mean flow field of the plume (Fig. SM3 of supplementary material). In this sense, we refer to inflow when the direction of the velocity vector with respect to the slice is greater than 180 degrees and less than 360 degrees. We refer to outflow when the direction of the velocity vector with respect to the slice is greater than 0 and less than 180 degrees. The angle is calculated for each point along the slice taking a Cartesian coordinate system as a reference. Thus, we refer to a quadrant in this context, where outflow corresponds to quadrant I and II and inflow to quadrant III and IV. Data within 2 standard deviations of a variable's profile are used in order to account for artificial spread of the plume. Total flow through the slice is obtained as the difference of outflow minus inflow. Finally, the result is integrated numerically".

Regarding the scaling of the combustion model results please see reply to point 2 of Referee 1 for an explanation of the scaling procedure. To clarify this point, next paragraph was added to Section 3.1.1 pg15187ln18: "The purpose of considering a single flare for the combustion simulation of this work relies on computing time and the scarcity of feed composition information. The aim is to represent, as far as possible, the total feed stream that is flared at Tula Refinery. This allows to model one representative flare, instead of three separate flares, and at the same time to simplify the assumption of gas composition. In this sense, the total mass flow used in the combustion simulation is based on the information provided by the emission inventory of IMP (IMPei) which reports 4.65 Kg/s. However, it was decided to slightly increase the total mass flow in order to represent flow variation at the refinery, without losing generality with respect to IMPei information. For this reason, the total mass flow rate for the combustion simulation was set to 5.3 Kg/s. Finally, the emission rates obtained with the combustion model are scaled to each of the three flares in order to compare with IMPei."

With respect to comment about the total soot rate, yes, F3 was taken as the flare with maximum emission rate.

3) The description of the WRF-Chem setup in Section 2.2.1 (pg15186) raises some questions about data assimilation (FDDA). They state that FDDA is used for the 2 coarse grids only for the first 24 hours. However, in Section 3.3.1 (pg15196) they credit FDDA for the good agreement of the model with observations on March 23 which is after the initial

24 hours. Also, in the conclusions (pg15204) there is a statement about the importance of analysis nudging for the whole period. First, why is FDDA used only for the first 24 hours? Which 24 hours, during the 2 day spin-up or on March 22? Is analysis nudging using the 1 degree FNL only? Clearly the meteorology simulation could be improved by using obsgrid to reanalyze with observations or using observation nudging and apply FDDA for the entire simulation period. In fact, Fast et al. (2009) is cited and noted that they used observation nudging and reported reasonable predictions. Why not follow this previous work?

We used FDDA for the first 24 hours of the spin-up period with 1 degree FNL. Preliminary results showed better agreement of the timing of the modeled plume with respect to the timing of the observations on 23 March, as well as better performance statistics. Fig SM4 of the supplementary material shows the difference between these two cases. In addition, we would like to add that even though not shown, we also obtained the time series for RAMA stations and the peak of 23 March and 25 March were better reproduced when nudging only the first 24 hours of the spin-up period.

As Referee suggest we used Objective Analysis to reanalyze meteorology with observations. We used NCEP ADP global surface wind and upper-air observations, corresponding to NCAR archives ds464.0 and ds353.4 respectively. In addition, observations of MILAGRO campaign and of RAMA surface stations and MODIS data for land-use were also included for OBSGRID. Several simulations with nudging in all the simulation period were performed. Basically, we applied grid nudging; grid/surface nudging; grid/surface/observation nudging with different nudging coefficients in two different configurations. The first configuration applied nudging to domains 1 and 2, whereas second configuration applied nudging in all domains. Results showed that Multi-scale FDDA had the best performance statistics. However, we noticed degradation in the plume dynamics if using either observation or grid nudging only, even though the performance statistics showed relatively good values. In particular, this was more pronounced on 23 March and almost all 25 March. This is shown in Fig. SM5 of the supplementary material. Despite Multi-scale FDDA improved the meteorology simulation, the plume dynamics at the supersites is very similar to the results reported in the manuscript. For this reason, we consider that our first results are reliable enough to support the discussion of this work. Nevertheless, Multi-scale FDDA will be the basis for future simulations regarding the study of flaring plume activities, particularly when including the national Emission Inventory.

4) Another point of clarification: the lowest layer is said to be 50 m. Is this the height of the mass level (layer mid-point) or the full layer? If the lowest mass level is 50 m, this is much higher than is typical. Also, when reporting the meteorology model performance it would be good to also show the model mean biases for all of the evaluated parameters.

It refers to the height of the full layer of the model. The mean biases are included in Table 3 as Referee suggest. A brief discussion is included in Section 3.2

5) Important results of this work are the average contributions of MHR emissions to the concentrations at the supersites which are reported on pg 15198 and in the abstract. However, it is not clear how these are calculated. Please explain.

Please see answer to point 9 by Referee 1

Technical Corrections:

6) Abstract: "IMP" is used before it is defined

Thank you. The acronym is defined in the abstract

7) Pg15182ln2: Capitalize Environmental in EPA.

Thank you. The text is corrected.

8) Pg15182Ln24: What is meant by "soot radiation"?

It refers to the continuum radiation from soot particles generated in the flame (Kim et al. 2001). Soot particles enhance heat transfer rates due to the continuum radiation in the visible and infrared regions of the wavelength spectrum (Farias, et al., 1998) and can decrease flame temperature (Wang et al., 2005).

9) Pg15183ln24: What is "Favre-averaged"?

When solving numerically the Navier-Stokes equations in constant density flows, the Reynolds averages are applied to the instantaneous balance equations in order to obtain the expressions for the mean quantities. This consists in splitting a variable into a mean and a fluctuating component. If this procedure is applied to a compressible flow, it introduces an unclosed correlation between the variable under consideration and the density. To avoid this, for flows with large density changes as occur in combustion, the Favre averages are preferred. In contrast to Reynolds averages, Favre averaged quantities are weighted by the instantaneous density. It allows the continuity equation to have an exact form and eliminates double correlations involving density fluctuations from the turbulent fluxes. Formally, the resulting expressions for the balance equations of the mean quantities have the same structure as the conventional averages but with simplified terms (Bilger, 1975; Poinsot and Veynante, 2005; Peters 2000).

10) Pg15185ln2: Should the gamma be in "the nucleation (alphaï˘A ˇngamma)" ?

Thank you. Delta has to be in place of gamma. For clarity the name of either process is added below the corresponding term in the equation as appears in the original paper by Moss et al., (1995). Therefore, expressions in parentheses of pg15185ln2 and pg15185ln3 are removed. They now read as follows:

$$\frac{d\rho_s f_v}{dt} = \underbrace{\gamma n}_{surface\ growth} + \underbrace{\delta}_{nucleation} - \left(\frac{36\pi}{{\rho_s}^2}\right)^{1/3} n^{1/3}(\rho_s f_v)^{2/3} R_{ox}$$

$$\frac{d}{dt}\left(\frac{n}{N_o}\right) = \underbrace{\alpha}_{nucleation} - \underbrace{\beta\left(\frac{n}{N_o}\right)^2}_{coagulation}$$

11) Pb15186ln3: Why: "in sub-grid scales"? ": : :different aerosol and photolysis schemes"; different from what?

Thank you. The corresponding paragraph introduces ambiguity. It is changed to "subgrid-scale". As for the aerosols and photolysis schemes, we meant several options to choose at run time. This is corrected in the manuscript as follows: "It uses the same transport scheme, the same grid, the same physics schemes for sub-grid scale transport, and the same time step of the meteorological component. Aside from the gas-phase chemistry mechanisms, it includes several aerosol modules and photolysis schemes."

12) In several places the word "specie" is used (which means coined money) when "species" is intended

Thank you. The manuscript is modified with the correct word.

13) Pg15188: "The initial and boundary conditions are the default values computed by the model." What are these default values and how were they "computed by the model". Also, since no emissions are used other than from the MHR, it is not clear how the ICs and BCs should be set.

The corresponding paragraph in the manuscript was meant to refer to the ideal profile of atmospheric species provided by WRF-Chem to start the simulation. The word default was used in a similar context of the experimental design of Zhang et al. (2009). Specifically it refers to the modeling option of WRF-Chem (chem_in_opt). These initial and boundary conditions are used as the initial background concentrations at the start of the model run (Zhang et al. 2009) in order to set the chemical state of the model

To clarify this point, the following paragraph is added to pg15187ln7: "The chemical boundary conditions are set with the ideal profile provided by WRF-Chem. It consists of idealized, northern hemispheric, mid-latitude, clean environmental profiles of trace gases based upon the results from the NOAA Aeronomy Lab Regional Oxidant Model (NALROM) (Tuccella et al., 2012; Grell et al., 2005)."

14) Pg15189ln19: Should be "about a g/s"

Thank you. This line is corrected in the manuscript.

15) Pg15190lns11-19: This paragraph is not very clear. For example, it is not clear how resolution effects the eddies and how the eddies affect the SO2 concentrations. Is higher resolution expected to increase or decrease SO2 concentrations downwind? Also, the term "crosswind" is confusing since this is a 2-d simulation and crosswind usually means at right angle to the mean flow. "This imply to lower hydrogen sulfide concentration." should be "This implies that hydrogen sulfide concentration should be lower. "

Coarse regions on a grid can promote artificial dispersion of the plume. Small-scale structures are better resolved as the mesh is refined. For instance, turbulent eddies smaller than the plume radius enhances turbulent diffusion (Zhang and Ghoniem, 1993). This can affect local concentration of species within the plume. Thus, artificial spreading can be reduced with higher resolution near the plume edge. In this work, artificial spreading of the plume can occur downwind, since the refined region of the mesh is on the flame. For this reason, we expect that higher resolution downstream diminish SO2 concentrations.

The paragraph in Section 3.1.2 on pg15190ln11 to ln19 is modified as follows: "The results also suggest to increase simulation time and to refine the mesh downwind the combustion plume, since coarse regions on a grid can promote artificial dispersion of the plume. Small-scale structures are better resolved as the mesh is refined. For instance, turbulent eddies smaller than the plume radius enhances turbulent diffusion (Zhang and Ghoniem, 1993). In this work, artificial spreading of the plume can occur downwind, since the refined region of the mesh is upstream on the flame region. This reflects in a wider profile of the species. For this reason, eddies generated after the ignition of the flame, present relatively high concentration of SO2 and can be slowly dissipated promoting a slower transport by the crosswind flow.
Besides, the composition and velocity of the stream sent to the flare are important, so that our assumptions also contribute to the uncertainty of the estimates. This implies that hydrogen sulfide concentration should be lower. In addition, the 2D domain lacks the dynamics and flame width that can be obtained in a 3D setup. Work is in progress to account for this in both transient and steady state."

With respect to the modification of the sentence "This imply to lower hydrogen sulfide concentration", it is included in the above paragraph.

Regarding the term crosswind, it is used in the manuscript in the context of open flames subjected to the influence of a cross flow. The 2D domain of the combustion simulation is oriented in the Z-X plane. The fuel stream exits in the vertical direction (Z axis) and the cross flow in the horizontal direction (X axis). This gives a right angle between the flow direction of each stream. However, in the context of Gaussian atmospheric dispersion (Turner 1994) the same orientation of the 2D domain of this work, implies that the crosswind would be oriented in the Y axis, corresponding to the Z-X plane of this work. Following the Referee's comment, a brief paragraph is included on pg15184ln11 to clarify this point as follows: "In this work, the term crosswind is used in the context of open flames subjected to the influence of a cross flow of atmospheric air. In this case, the fuel stream exits at right angle with respect to the crossflow"

16) Pg15192ln8: Should "1.07" be "3.37"?

Thank you. This is corrected in the manuscript

17) Pg15192ln9: It would be better to say "is too high" rather than "can be lower". There are many places where new paragraphs are started where they shouldn't be, such as pg15192ln16, pg15197lln21, pg15200 and pg15204ln4. I suggest merging these paragraphs with the previous paragraphs.

Thank you. We changed to "is too high" in the corresponding line and these paragraphs were merged as suggested by the Referee.

18) Pg15194ln10: Insert "did" between "simulation" and "not".

Thank you. It is corrected in the manuscript.

19) Pg15195ln24: "For NOx the same estimate of NO2 and NO: : :" Same as what?

This line refers to the combustion model estimate. The manuscript changed to include a better explanation of the scaling of emissions for the flares as previously mentioned.

20) Pg15196ln22: Should "timing" be "magnitude"?

In this work, we use the word timing to refer to the relative agreement of the time of the day between the observations and model results, at which the maximum concentration occurred when the flaring plume impacted a site. Although the magnitude of the concentration is relatively well captured by the model, there is still an overprediction.

21) Pg15197ln19: "form" should be "from"

Thank you. It is corrected in the manuscript.

22) Pg15197ln23: What is meant by "increment of emissions"?
Thank you. This line is misleading. It is changed to "increment of concentration" in the manuscript.

23) Pg15201ln3: "form" should be "from"

Thank you. It is corrected in the manuscript.

24) The Figures should be improved. Many, particularly figs 1, 6, and 7 are too small and blurry to read. The line plots in figs 3, 4, and 5 could also be expanded with times of the day added to the x-axis. On Fig 7 in would be good to label T0, T1, and T2 and add the location of the TIC.

Figures are modified with Referee's suggestions.

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