Authors' reply to comments on the revision of "Development of a plume-in-grid model for industrial point and volume sources: application to power plant and refinery sources in the Paris region"

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We thank the anonymous referees for reading the manuscript attentively and giving helpful comments to improve the manuscript.

1 Reply to the anonymous referee #1's comments

Referee #1's main comment 1:

1. Section 4.3 should be clarified. First, the name of the sensitivity run "PinG-injection" is not ideal, because the base PinG simulation also uses an injection approach.

   Our response: The name has been corrected as "PinG-SizeOnly".

2. Second, the zones discussed in the text are not clear in the Figures, and the results in Figure 5 do not appear to easily fit into radial zones. I recommend labeling the zones in the figures and zooming into the region of interest in the figures (here and elsewhere) so that smaller scale features can be seen.

   Our response: Figures 4, 5 and 6 (now Figures 4, 6, 7 and 8) have been corrected following the recommendation of the referee. The zones have been labeled according to distance from the source and we now zoom slightly onto the region where the sources are located.

3. Third, it is stated that earlier injection into grid cells leads to lower concentrations due to greater dispersal into neighboring grid cells in the Eulerian model (p. 5879, lines 22-25). Is this because the column injection approach used here would tend to underestimate horizontal transport by injecting all puff mass into the horizontal cell where its centroid lies even if part of the puff overlaps a neighboring cell? Also, I would think that early injection into the grid could increase (rather than decrease) concentrations by enhancing mixing to the surface.

   Our response: The injection method was incorrectly stated in the manuscript. The integrated injection has been used instead of the column injection. The manuscript has been corrected as follows:

   "The Gaussian puff model formulation uses similarity theory for the parameterization of the Gaussian standard deviations and the integrated injection for the injection method. In the integrated injection method, puff mass is distributed over the grid cells covered by the puff horizontal and vertical extents (here, $2\sigma_y$ and $2\sigma_z$)."

   The early injection into the grid could increase concentrations by enhancing mixing to the surface in case where the puffs remain aloft. In the case of the two industrial sources simulated here, vertical dispersion of the puffs is sufficiently significant (e.g., see Figure 5) that the puff material reaches the surface and, therefore, horizontal dilution tends to govern the differences in concentrations here.

For vertical mixing, the figure below presents vertical cross-sections of the differences in the mean concentrations of NO_x between the PinG and PinG-SizeOnly simulations. The concentrations of NO_x at the Grandpuits refinery are calculated using the integrated injection for both simulations. They are lower in the PinG simulation than in the PinG-SizeOnly simulation between the surface and about 500 meter altitude at the source. The greater concentrations in the PinG simulation west of the source result from upwind diffusion (prevailing winds are from the west).

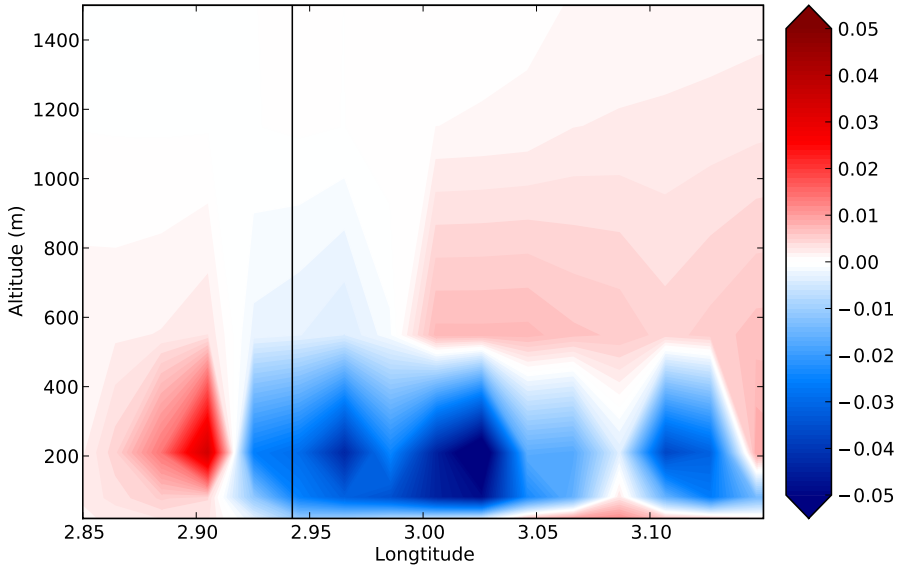

Figure 1: Vertical cross-sections of the differences (PinG - PinG-SizeOnly) in the mean concentrations of NO_x (ppb). The black line corresponds to the longitude of the Grandpuits refinery.

4. Fourth, in this section and in the following section, the authors discuss NO_x and SO_2 in comparing the mixing and transport of pollutants in the different configurations. An inert tracer like CO would be more useful for understanding mixing and transport differences than the reactive species chosen.

   Our response: We have examined the impact of the different configurations on the concentration of CO. However, the differences in the concentration of CO are less significant than those of NO_x and SO_2 because of the much lower emission rate of CO than that of NO_x or SO_2 from the two point sources (10 times less).

5. Fifth, the percent change in concentration due to the change in injection method is given, but the magnitude of this change in relation to the overall impact of using PinG compared with not using PinG is not discussed.

   Our response: The following discussion has been included in the revised manuscript.

   "An indicator, I, which quantifies the influence of the time criterion on the pollutant concentrations was calculated using the following equation.

$$I = \frac{C_{PinG} - C_{PinG-SizeOnly}}{C_{PinG} - C_{Reference}} \tag{1}$$

   where C is the concentration in grid cells. Very low C values lead to high I values and can mislead the analysis. Therefore, a threshold value for $(C_{PinG} - C_{Reference})$ is applied (0.1

ppb or μg m^{-3}). As expected, the indicator I shows similar patterns as those displayed in Figure 7 with negative values near the source (zone 1 typical of low wind speed conditions) and positive values farther downwind (zone 2 typical of high wind speed conditions), as depicted in Figure 10 for NO_x. The magnitude of I is a measure of the effect of the injection criterion compared to the effect of the use of a PinG treatment. It appears that the latter effect is dominant as the former is typically 20 % or less of the latter as shown in Figure 10; similar results were obtained for other species (not shown).

Nevertheless, the effect of the injection criterion is not negligible and the use of the size criterion is recommended. However, it should be considered that trajectory uncertainties become large with long injection times and an upper limit of 3 h for the injection time has been recommended for simulations using very coarse grids (Korsakissok and Mallet, 2010b)."

Referee #1's main comment 2:

1. Section 4.4 focuses on understanding the impact of horizontal grid resolution but the authors changed the injection criteria along with the grid resolution (p. 5881, lines 27-28). Therefore it is hard to know how much of the differences presented are due to the grid resolution change versus the injection criteria change.

   Our response: The sensitivity simulations for the injection criteria presented in Section 4.3 were performed using the same injection criteria to the sensitivity simulations for the grid size presented in Section 4.5 (previously 4.4). Therefore, we have used the results of the simulations in Section 4.4 (previously 4.3) for the comparison instead of the results of the simulations in Section 4.2. We have added the following text to clarify.

   " For consistency in the injection criteria, the results of the simulations with the puff size injection criterion presented in Section 4.4 are used when the sensitivity simulations for the horizontal grid size are compared to the simulations with the fine grid resolution. "

2. Also, this section refers to grid resolution criteria and transport of pollutants in units of meters and km, but the grid resolutions for this study are all provided in degrees. I recommend providing an estimate of the grid resolution in km so that discussions of this type are clear.

   Our response: The following text has been added in the revised manuscript.

   " In this study, the resolution of 0.10° corresponds to 7.3 km (east to west domain-mean value) × 11.1 km (south to north) and that of 0.02° corresponds to 1.5 km (east to west domain-mean value) × 2.2 km (south to north)."

3. Also, the authors indicate that the PinG treatment has a greater impact at finer resolution due to issues related to grid-cell averaging (p. 5882, lines 9-10). Does this model have subgrid sampling capabilities that could be used to eliminate grid-cell averaging effects in the comparison?

   Our response: Subgrid sampling has not been implemented yet in this model. Subgrid sampling would be useful to compare to monitoring data at sites impacted by the sources (e.g., Karamchandani et al., 2006), none were available here. When comparing PinG with the Eulerian model, it is common to compare grid-averaged values, in which case subgrid-scale sampling is not needed.

Our responses to the additional comments:

1. The authors should consider softening some of the language in the first paragraph of the introduction. The volume source considered in the current study is roughly the size of a cell in a high resolution (1km) grid simulation, and so pollutants in a standard photochemical run may not experience more dilution than in a PinG simulation. Also, this paragraph implies that standard gridded simulations would have significant errors near sources that would be reduced using PinG techniques. While this may make sense conceptually, the current study and most studies cited never evaluate model predictions with in-plume observations (i.e., they just illustrate the impacts of the PinG treatment). Given the large number of parameters

and algorithms used in PinG models, as well as the lack of fine-scale evaluation, the errors associated with PinG treatments are unclear, and so it should not be assumed that they give better model performance in practice.

Our response: We agree that the volume source could be handled well with a standard Eulerian model with a 1 km grid size; however, in many typical model simulations coarser grid sizes are used (up to 50 km for continental simulations) and grid sizes of about 10 and 2 km are used here. Therefore we have used the following text in the introduction to explain that subgrid-scale modeling is only useful when a grid cell size is much greater than a source size.

" This modeling approach can lead to significant errors for emission sources that have much smaller dimensions than those of the grid cells. "

There have been some evaluations of PinG models with in-plume observations and we have added a summary of those in the revised manuscript as follows.

"The performance of a PinG model (or its subgrid-scale model) has been evaluated with measured in-plume concentrations. Vijayaraghavan et al. (2006) compared concentrations simulated with a subgrid-scale puff model (SCICHEM) used in a PinG model (CMAQ-APT) with aircraft measurements of power plant plumes. Modeled plume excess concentrations (i.e., the differences between the plume and background concentrations) of NO_x and O_3 showed good agreement with measured concentrations about 4 km downwind of the sources. Karamchandani et al. (2006) introduced an approach to calculate subgrid-scale concentrations using a similar PinG model (CMAQ-MADRID-APT) that accounts for uncertainty in the wind direction by calculating the concentration at a receptor site along an arc centered at the source and extending 30° on each side of the receptor site. The source impacting the site was identified based on the SO_2/NO_x plume excess ratio and the backtrajectory. The subgrid-scale concentrations showed variability in SO_2 concentrations within the grid cell of a factor of three and reproduced the plume concentration observed at the receptor site. The Eulerian model (i.e., without PinG treatment) underestimated this observed concentration by a factor of two."

2. In the methods section, it would be good to include some language on how concentrations were processed for the model intercomparisons. For example, were pollutants in non-injected puffs merged with the gridded background concentrations before comparing with the reference simulation (or were in-puff concentrations excluded from the comparison)? Also, are there any issues associated with overlapping puffs independently interacting with background species (e.g., if each overlapping puff has access to NH3 in the background field, one could imagine that NH3 could be depleted in excess of its total amount in some situations, since NH3 is in instantaneous equilibrium with the particles)?

   Our response: All puffs were included in the comparison. To that end, the pollutants were instantly merged for the comparison. The following text has been added in the revised manuscript.

   "For the comparison of the concentrations calculated in the Reference and PinG simulations, the perturbations of concentrations in non-injected puffs in the PinG simulation are taken into account by injecting the puffs in the corresponding grid cells at the moment of the comparison (and solely for the purpose of the comparison). The puff integrated injection method is used for this temporary merging of the puffs into the host model."

   Concerning the potential excess depletion in the background concentrations, it is important that the background concentrations be updated after performing the chemistry for each puff sequentially. For example, if NH_3 in a given grid cell is depleted by aerosol chemistry in a puff, the background concentration of NH_3 decreases. Then, the reduced background concentration is used for aerosol chemistry in the next puff located in that same grid cell (if any). Thus, NH_3 mass is conserved.

3. It would be helpful to add a table with the average altitude of the model grid layers and the emissions for all species from the two PinG sources (possibly as an online supplement).

Table 1: Annual emission rates of gaseous and particulate species at the two industrial sources (tons year^{-1})

	SO_2	NO_x	CO	PM_{10}	$PM_{2.5}$	VOC
Vitry power plant	4324	3088	270	107	49	29
Grandpuits refinery	3778	1078	409	441	287	540

Our response: The altitudes are described with constant values in meters in the Polyphemus PinG model. The list of the altitudes has been added in the revised manuscript.

"The altitudes of the vertical upper boundary of the grid cells are 40 m, 120 m, 300 m, 800 m, 1500 m, 2400 m, 3500 m, 6000 m, and 12000 m."

The emission rates of the point sources are given in Table 1 and the corresponding text has been added in the revised manuscript.

"The annual emission rates for the Vitry power plant and the Grandpuits refinery are presented in Table 5. The NO_x emission rate of the Vitry power plant is about three times greater than that of the Grandpuits refinery, while the latter has a greater VOC emission rate than the former."

4. Based on the description in the Korsakissok and Mallet (2010b) study, the column injection approach seems to be less physically realistic than the integrated injection approach. Conceptually, the column injection approach seems to over-estimate vertical dispersion and under-estimate horizontal dispersion relative to information predicted by the puff model. Why not use the integrated injection method?

   Our response: As mentioned above in our response to main comment #1, the integrated injection approach was used instead of the column injection approach in our simulations. However, it is not evident which approach is better. According to Korsakissok and Mallet (2010b), the integrated injection approach can lead to an overestimation of the puff horizontal dilution.

5. The ideas in Section 4.2 could be conveyed more clearly with vertical cross-section plots showing how PinG impacts the vertical structure of pollutants near the source and downwind. This would also illustrate how diluted the puffs are in the vertical direction and provide insight on the value of using sub-grid treatments for emissions sources.

   Our response: Vertical cross-section plots for the mean NO_x concentrations at Vitry and Grandpuits and the corresponding text have been added in the revised manuscript.

   " Figures 5a and 5b present the vertical cross-section of the differences in the NO_x concentrations at Grandpuits and Vitry, respectively. As discussed above, the NO_x plumes are emitted at higher altitudes at Vitry than at Grandpuits, which leads to a plume touchdown farther downwind at Vitry. Therefore, the impact of PinG modeling on the surface concentrations of NO_x is greater at Grandpuits than at Vitry. "

6. In the conclusion section, it is not clear how the authors determined that the time criterion is significant for the formation of secondary aerosols. It might be better just to state the percentage impact associated with the sensitivity run.

   Our response: Following the referee's comment, the corresponding text in the conclusion has been corrected as follows.

   " Nevertheless, the use of the size criterion is recommended for $PM_{2.5}$ although an upper limit of 3 h could be imposed for puff travel times to minimize the effect of uncertainties in puff trajectories. However, it must be noted that the injection criterion has less effect on the air quality simulation than that of the plume-in-grid treatment since the effect of the former on pollutant surface concentrations is typically 20 % or less of the effect of the latter. "

2 Reply to the anonymous referee #2's comment

1. The paper discusses only surface concentrations, but the changes in pollutants at all levels. The surface levels are clearly important for atmospheric exposure, but higher altitudes will affect wet-deposition. Have you characterized the affects on total column and/or total domain mass?

   Our response: A new paragraph which concerns the effects at higher levels has been added as below in the revised manuscript.

   " Figures 5a and 5b present the vertical cross-section of the differences in the NO_x concentrations at Grandpuits and Vitry, respectively. As discussed above, the NO_x plumes are emitted at higher altitudes at Vitry than at Grandpuits, which leads to a plume touchdown farther downwind at Vitry. Therefore, the impact of PinG modeling on the surface concentrations of NO_x is greater at Grandpuits than at Vitry. "

2. The article compares pure Eulerian results to the updated PinG model, but heavily references the previous PinG model. The updates developed in this paper are the PM treatment and the volume source. Given that only the VOC emissions use the volume source update, many of the changes in gas concentrations (NO_x and O_3) likely have small incremental changes. Why would the base (no PinG) be a better reference case than the pre-existing PinG model? Given that this is a model development journal, it would be nice to characterize the contribution of the original development in this paper.

   Our response: The previous PinG model that was used in this development is that of Korsakissok and Mallet (2010a). It was developed to study the impact of PinG on ozone formation and did not include PM treatment. Therefore, it cannot be used here to study the impact of PinG on PM formation.

   Following the referee's comment on the volume source, we have performed an additional simulation in which the volume source is not taken into account using the PinG treatment. Results of the additional simulation are now discussed in Section 4.3 in the revised manuscript.

   " In the previous section, the fugitive VOC emissions at the Grandpuits refinery are treated with a volume source. For further analysis about impacts of the volume source treatment on the VOC emissions, an additional simulation was conducted in which the VOC emissions at the refinery were injected into grid cells as in the Reference simulation, but other pollutants (NO_x and SO_2) were emitted through the stack using the point source subgrid-scale treatment. The results of this simulation (hereafter PinG-NonVolumeSource) are compared to those of the PinG simulation.

   Figure 6a presents the differences in the concentrations of a VOC, namely toluene. As expected, toluene concentrations are greater. Differences in oxidant concentrations are shown in Figures 6b and 6c. The volume source PinG treatment leads to greater oxidant concentrations downwind of the source due to greater VOC concentrations from the refinery fugitive emissions. However, the differences are small (< 1 %) because VOC refinery emissions are a small contribution to total VOC ambient concentrations. Differences in secondary organic aerosol (SOA) concentrations between the PinG and PinG-NonVolumeSource simulations are shown in Figure 6d. SOA is formed in the atmosphere from semi-volatile organic compounds (SVOC), which are formed by oxidation of VOC emitted in the atmosphere from both anthropogenic and biogenic sources. The impact of the volume source in PinG modeling on the concentrations of VOC can lead to the differences obtained between the two simulations near the Grandpuits refinery (3 %). Although the differences in the concentrations of VOC are significant only near the Grandpuits refinery (Figure 6a), the differences in the concentrations of SOA extend farther downwind of the source.

   The differences near the Grandpuits refinery are partly due to the differences in the concentrations of anthropogenic SOA formed by oxidation of VOC emitted in the source (toluene, xylene and glyoxal oligomer). However, the impact of the PinG modeling on the anthropogenic SOA is negligible downwind of the source (not shown). In addition, the contribution of the anthropogenic SOA to the total differences in the concentrations of SOA is rather low

(about 10 % of the total differences) even near the source. In fact, the differences in the SOA concentrations are due to differences in the concentrations of biogenic SOA, due mostly to the oxidation of monoterpenes. Since monoterpenes are not emitted from the Grandpuits refinery, the differences in the concentrations of biogenic SOA are due to oxidation of monoterpenes emitted in the surrounding region by oxidants (OH, O_3 and NO_3), which differ in concentrations between the two simulations (see Figures 6b and 6c). However, SOA concentration differences are small (about 0.05 μg m^{-3}) "

3. Page 5868-5869: The description of chemistry in the bullets was not clear without reading the citation. Either add more details here or simply cite the other document.

   Our response: The text has been modified in the revised manuscript as follows.

   " Following the modeling approach used in the reactive puff model SCICHEM (Karamchandani et al., 2000), the concentrations of the chemical species in each puff are treated as perturbations (Δc) of the background concentrations,

$$\Delta c = c^p - c^b \tag{2}$$

   where c^p is the concentration in the puff and c^b is the concentrations modeled by the Eulerian host model, i.e., the background concentration. To calculate the concentrations of the species in the puff, we use the procedure described by Korsakissok and Mallet (2010a):

   (a) Let a chemical reaction in a puff be A + B → P, where A and B are gas-phase or particulate species. The concentration of the species A and B in the puff at time t are $c_A^p(t)$ and $c_B^p(t)$. These puff concentrations correspond to the background concentration c^b added to the perturbation of the puff Δc.

   (b) The new concentrations of the species A in the puff due to the chemical reaction is computed for the time step Δt. The reaction rate of the species A in the puff is

$$\frac{dc_A^p}{dt} = \frac{d(c_A^b + \Delta c_A)}{dt} = -k(\underbrace{c_A^b c_B^b}_{(1)} + \underbrace{c_A^b \Delta c_B + c_B^b \Delta c_A}_{(2)} + \underbrace{\Delta c_A \Delta c_B}_{(3)}) \tag{3}$$

   where k is the rate constant, (1) represents the background chemistry, (2) is the interaction between the puff and the background species, and (3) is the chemistry specific to the puff.

   (c) The new background concentration due to the chemical reaction is computed separately from the previous equation for the time step Δt. The reaction rate for species A in the background is

$$\frac{dc_A^b}{dt} = -k(c_A^b c_B^b) \tag{4}$$

   (d) The new background concentration is subtracted from the new puff concentration to obtain the new perturbation of the puff. The reaction rate representing the perturbation is then,

$$\frac{d\Delta c_A}{dt} = -k(\underbrace{c_A^b \Delta c_B + c_B^b \Delta c_A}_{(2)} + \underbrace{\Delta c_A \Delta c_B}_{(3)}) \tag{5}$$

   The new perturbation of the puff corresponds to the terms (2) and (3) in Equation 3."

4. Page 5871, lines 11-13: There is a suggestion that size criteria gives better results for grids < 25 km and time criterion gives better results for > 50 km grid cells. This is written as a universal truth, but your paper investigates the effect. Your results section suggests that there is negligible effect on performance of PinG at all. Thus, it seems that the injection criteria would not substantially affect performance evaluation. Can you clarify whether this is your result or previous literature findings?

   Our response: A relevant reference was missing and has been added (Korsakissok and Mallet, 2010b). For the corrected text, see the response to the next comment.

5. PinG-injection was very confusing. Both plumes were "injected" into the grid. How about PinG-sizeonly? Further, I recommend that you give add more introduction of the criterion-sensitivity and resolution-sensitivity to the "Simulation setup" section. Adding light outlines to the figures would help with the zones, which are only clear after much discussion.

   Our response: Following the referee's comment, the simulation name has been replaced by PinG-SizeOnly.

   A discussion of the sensitivities to the injection criteria has been added in the revised manuscript.

   " The sensitivities of modeling results to the puff injection criteria have been reported by Korsakissok and Mallet (2010b). They are summarized here. The use of the time criterion (time elapsed since the emissions from the source), with a 1 h injection time, improved the performance of the PinG model compared to that of the Eulerian model for a domain with a grid cell size less than about 60 km. The size criterion (puff size commensurate with the grid size) gave better results than the time criterion for a domain with a fine grid cell size ($<$ 25 km); however, the results for a domain with coarser grid cell size were poor because of a greater injection time (more than 6 h), which may lead to large errors in puff trajectories. Therefore, the time criterion gave better results than the size criterion for a domain with a coarse grid cell size ($>$ 50 km). "

   Following the referee's comment, the figures have been redrawn with the zones being depicted.

6. The horizontal grid-sensitivity analysis needs clarity and potentially more analysis. First, it is not clear if the PinG and Reference at 0.1 degrees are being compared to a 0.02 degree model with the same criterion (size-only) or with a different criterion (sizetime). Second, it would be more useful if it more fully characterized the competing factors: small-cell-size induced injection and large-grid insensitivity. The analysis as presented shows a "high" value at 0.02 degrees (∼1.5km) and "lower" response at 0.1 (∼7km) degrees. The native WRF resolution of 0.0555 degrees might have shown some added value without diluting the total response.

   Our response: The same criterion (size only) was used for both 0.1° and 0.02° simulations. It has been clarified in the revised manuscript.

   " For consistency in the injection criteria, the results of the simulations with the puff size injection criterion presented in Section 4.4 are used when the sensitivity simulations for the horizontal grid size are compared to the simulations with the fine grid resolution. "

   Additional paragraphs have been added to provide further analysis in the revised manuscript.

   " Two test simulations were conducted to compare characteristics of puff traveling with the fine and coarse grid size during 24 hours. Puffs in the simulation with the coarse grid size can travel longer than in the simulation with the fine grid size. This more traveling time of puffs can reinforce impacts of PinG modeling in the simulation with the coarse grid size. The traveling times in the simulations are compared in Table 2. The mean puff traveling time with the coarse grid size is twice greater than that with the fine grid size. About 80 % of puffs in the simulation with the coarse grid size could reach the boundaries of the simulation domain without being injected into the Eulerian grid. However, 70 % of puffs in the simulation with the fine grid size were injected into the host model due to the smaller grid size."

Table 2: Characteristics of puffs in the simulations with different horizontal grid sizes.

	0.02° resolution	0.10° resolution
Emitted puffs	432	432
Injected puffs	301	36
Puffs reaching domain boundaries	50	334
Puffs not injected at the end of the simulation	81	62
Mean traveling time	2.2 hours	4.7 hours

7. In the conclusions, you state "The PinG modeling results presented here demonstrate that fugitive emissions need to be taken into account in addition to stack emissions for industrial sites treated at the subgrid scale." However, this has not been demonstrated. In this work, the fugitive PinG emissions are being compared to a no-PinG model. To truly demonstrate the need for fugitive emissions to be treated as PinG, the comparison would have been between the pre-existing PinG model and the updated PinG model. Was that comparison made? Comparing to the Karamchandani paper (in the conclusion) is less than ideal because it used a different host model with potentially different conditions. Either the comparison between Polyphemus PinG and updated PinG would be ideal, or it need to be clearer how different the present model is from the previous Polyphemus PinG.

   Our response: We agree with the referee's comment. Accordingly, we have performed an additional simulation in which the fugitive emissions are injected in grid cells as in the Reference simulation. For our detailed response, we refer to the response to the referee's comment No. 2 above.

References

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