

Replies to Reviewers' Comments

Reviewer #1: (all replies in **bold**)

The paper by S. Deng and coworkers is a joint numerical/experimental work aiming to investigate the effect of hydrogen addition on soot formation in ethylene turbulent non premixed flames. The configuration of choice is a bluff-body stabilized jet, which was investigated prior by the authors with pure ethylene. The work reports and analyzes data from an LES of the flame and compares to experimental data. Although the soot volume fraction in the near field is significantly over predicted by the LES, the analysis is in depth and insightful, highlighting the role of chemistry effects (explored with flamelets) in the far field as well as mixing effects on the soot yield in the near field.

The paper is very well written and the conclusions are clearly stated and sound. The paper stands as one of the very few LES of sooting flames with comparison to experiments and much is to be learnt from it -- although the agreement between experiments and predictions is overall poor and much work is needed from this community.

I enjoyed the paper and found it very valuable. This paper is a clear acceptance. I have only two broad comment/observations.

Firstly, with all of the experimental and numerical framework available, it is unfortunate that cases at intermediate levels of H₂ have not been considered and reported. These intermediate H₂ cases would have helped tremendously in (a) characterizing the effects broadly and quantifying

non-linearities; (b) confirming that the LES and methods are sound in capturing trends (if not actual values, as it is the case here); (c) increasing the amount of soot well above the LII detection threshold.

The current study has demonstrated the ability of the model to capture the reduction in soot due to hydrogen addition and provided a mechanism for this reduction: chemistry and hydrodynamics. We agree with the reviewer that it is valuable to also consider other hydrogen blending ratios and note that another systematic study on this topic using a series of laminar flames has recently been submitted by the authors. However, this topic is well beyond the scope of the present investigation.

Secondly, the analysis of soot formation in the far field is lacking (but can be easily remedied). After all, that is where all of the soot is formed.

Both regions of the flame have been analyzed. However, only the recirculation zone is analyzed in greater detail with the LES results. The far-field converges to an axisymmetric turbulent jet, which has been well studied previously with both DNS and LES. This previous work is incorporated into our analysis as reviewed in the Introduction of the manuscript.

== Strengths

A) One of a kind study that leverages a detailed modeling approach to produce very useful information about the effect of H₂ on soot from ethylene flames in a pseudo-realistic

configuration. This is clearly useful from a practical perspective and stands as an example of the powerful contribution of LES to combustion technology.

B) Overall sound and insightful analysis supported by experimental data that show clearly how hydrodynamics and mixing effects, -- not only chemistry effects! -- contribute to controlling soot yields.

== Weaknesses

A) Lack of intermediate cases of H₂ doping

B) Somewhat incomplete analysis of the velocity/mixing field and their scaling for the two flames

C) Incomplete analysis of the soot yield in the far field -- where all of the soot is present.

** Note that (B) and (C) can be *easily* remedied in a revision as I suggest below (see comments 5-8).

== Detailed comments

1) p. 4 How are the "lumped PAH" defined and is there any sensitivity in the LES results to how those are defined? This is a key modeling point and should be discussed.

The lumped PAH is defined as the summation of the eight mass fraction of A2 to A4 stable species. The PAH model is taken from previously published work, which is explicitly given in the revised manuscript. The reviewer is referred to that work for further discussion.

2) Fig. 1, 5, 7. All of those are of low quality. In contrast to the EPS used in Fig. 2, 3, 4, and 6, those appear to be low resolution PNG or similar. The authors should improve the quality.

The figures will be enlarged in the revised manuscript.

3) The origins of the chemical effect of H₂ is somewhat brushed upon very quickly. Perhaps this effect is clear to those that have looked at it prior, but it would be desirable for the authors to spend a paragraph detailing why this is happening, i.e. expanding beyond a simple sentence. In this new paragraph, the authors should also comment on the technological implications of keeping Re constant, while changing some ethylene to hydrogen. What happens to the power from the flame? I would think this is an important consideration to discuss and/or comment upon.

We have added text to the Introduction to elaborate the chemical effect.

The heat release from the two flames is essentially the same, 42 kW, as have now been mentioned in the revised manuscript. The constant heat release helps reduce the number of varied parameters and results in similar thermal effects on flow dynamics.

Actually, for the hydrogen added flame with the same heat release, the jet Reynolds number is slightly lower than the neat ethylene flame by 8%, that is, 28,400. However, the flow structure in the recirculation zone, which is the focus of the current study, is dominated by the coflow, which is unchanged between the two cases (i.e., the same

turbulent Reynolds number). Furthermore, the reported jet Reynolds numbers are well above the threshold for turbulent transition, so this small difference is expected to be insignificant. This discussion has been added to the revised manuscript.

4) Since the soot field seems to be very sensitive to the fuel jet velocity, I would like to bring to the attention of the authors some confusion/inconsistency about the velocities/Reynolds nr/kinematic viscosities.

My calculations for the C₂H₄ (flame 1) and C₂H₄/H₂ (flame 2) cases with Re=30,900 (D=3.6 mm) are as below. All properties at 1 atm and 25 C with CHEMKIN and USC-Mech. The mechanism by Narayanaswamy, Blanquart, and Pitsch (CNF 2010) gave the same results to the reported digits.

	flame 1	flame 2

kin visc. (m ² /s)	8.73e-06	1.31e-05
Jet bulk (m/s)	74.9	112.8

What confuses me is the fact that my calculations for flame 1 give ~74 m/s, just like stated Mueller et al CNF 2013, while for the second flame (this paper), they give 112.8 m/s, while the authors state 102 m/s. A clarification is needed here.

The Reynolds number reported in the original manuscript for the ethylene/hydrogen mixture was based on the mixture viscosity estimated with a simple model, rather than the mixture-averaged viscosity from flamelet calculations, which is less accurate when the fuel

contains a high percentage of hydrogen. The experimental conditions were set based on the less accurate viscosity to match Reynolds number with the pure ethylene flame, but the flow rates are matched in the LES to ensure complete consistency between the experiment and simulation.

The more accurate Reynolds number of 28,400, corresponding to 102 m/s jet velocity, is provided in the revised manuscript. As stated above, this slight difference is expected to be insignificant in the far-field jet and plays no role in the near-field recirculation zone.

5) The discussion on the residence times in the recirculation region being constant is incomplete because, although the region is constant in size (as shown in Fig.5), the authors should bring more substantial proof that the velocities are the same.

The jet bulk velocity increases, suggesting that the velocities in the recirculation region should increase also, unless this increase in the jet bulk is inconsequential for the recirculation region due to the coflow air being constant. The authors should include information about the velocity in the recirculation region and its scaling with respect to flame 1 (pure ethylene). See also detailed suggestions/comments in 6) below.

As already mentioned in the manuscript, the recirculation zone is governed by the vortex in the outer mixing layer, and, therefore, the strength of the vortex as well as the residence time is set by the coflow, which is constant in these two cases.

6) On the velocity and scalar dissipation rate fields

When the authors replace ethylene with hydrogen, the fuel jet velocity is increased to match the Reynolds number, because, as shown above, the kinematic viscosity increases. The jet velocity increases by a factor equal to 1.36 from 75 m/s (flame 1) to 102 m/s (flame 2, this flame) (or 112 m/s -- see comment 4 above). If the scalar dissipation rate scales with U/D (as it should in the "high Re" limit), flame 2 has scalar dissipation rates that are 1.36 times higher.

a) Is this scaling for the scalar dissipation rate applicable? Despite the coflow velocity not having been scaled? The authors should present data on the conditional scalar dissipation rate (mean, resolved should be enough to say something, I reckon) scaled by U/D and compare across the two flames.

Data should be shown in the near field region across the mixing layer and in the far field, where a more "canonical" soot formation mechanism (due to decreasing mixing rates) appears to control soot yield. This comparison is important to characterize the mixing and soot fields comprehensively.

b) The fact that the size of the recirculation region stays unchanged is not surprising, since this size scales as a geometrical feature (i.e. the diameter D) multiplied by a $f_{cn}(Re)$, all other factors being irrelevant (e.g. Damkoehler). Thus, since the geometry is the same and $Re = \text{const}$, the gradients of mixture fraction should be constant. Yet, the velocity of the jet increases, suggesting that the velocity in the recirculation region, and the residence times, may scale somewhere between 1 (no increase) and 1.36 (full increase). Data should be shown.

c) As explained clearly by the authors, it seems that in this configuration, scalar dissipation rate in the near field is important to soot yield only to the extent by which it induces leaning in the recirculation region (see Fig. 6), and not to the extent by which it controls PAH formation. Higher scalar dissipation rate may quantify the enhancement of leaning observed in flame 2 and should be shown.

d) I expect the scalar dissipation rate in the far field to increase in flame 2 compared to flame 1 (by a factor close to 1.36?). This may have implications for soot formation decreasing (see comment 8 below) beyond and in addition to the chemical effect. This aspect should be explored with statistics of scalar dissipation rate and supporting flamelets.

The reason why we do not focus our discussion on the downstream-like region has already been addressed above. However, we have computed laminar steady flamelets for the hydrogen added flame at $\text{CHI}=1$ /s and 1.36 /s. The PAH mass fraction decreases by only 20%. Therefore, the soot reduction with hydrogen addition in the downstream part is still primarily due to chemical effects.

The scaling provided by the reviewer, indicating that the scalar dissipation rate goes with U/D , is not applicable to the recirculation zone, for the recirculating flow pattern is different from a free stream boundary condition. According to Fig. 6, in the recirculation zone near the fuel jet, where PAH based soot formation is dominant, the slopes of the resolved mixture fraction profile are almost identical. The mixture diffusivities estimated

at these mixture fractions are quite similar as well. Inside the recirculation zone, as discussed in the manuscript and previous work, the dominant soot formation mechanism is surface growth, which is less sensitive to dissipation rate. Nonetheless, according to Fig. 6, again, at mixture fractions that support soot surface growth, the scalar dissipation rates for both flames are essentially identical and small due to negligible gradients. Therefore, the scalar dissipation rate difference in these two flames, if any, is very unlikely to contribute to the significant soot reduction with hydrogen addition in the recirculation zone.

7) p. 7. The predictions of the authors for soot volume fraction in the near field are much higher than what it is obtained experimentally. They suggest that the issue may lie in the experiments because of the detection threshold being close to the value measured. This interpretation forgets that soot is highly intermittent, so that, although the mean soot volume fraction is low, the instantaneous values may not be and be much higher than the detection threshold, so that the experimental data is fine.

Could the authors comment (and include in the manuscript) about the PDF of the soot volume fraction at $r/D_B = 0.4$ ($x/D_B=0.432, 0.576$), especially with regard to possible issues with the instantaneous soot volume fraction being too close to the detection threshold?

The PDF of the soot volume fraction in the recirculation zone would suffer from the same uncertainty as the mean soot volume fraction. As a further note, the intermittency in the recirculation zone and downstream jet-like region are being investigated in a separate work.

8) Most of the soot is present in the far field, with little to no soot being formed in the near field. The authors attribute the change to chemistry and bring proof of this from flamelets where $Y(\text{PAH})$ decreases by 2 (suggesting a decrease by 4 in soot yield -- as observed in experiments). Yet, the authors should include a more thorough analysis of soot formation (and mixing) in the far field. In particular:

a) The authors should show soot predictions in the far field, where the LII threshold is much smaller than the average soot volume fraction: ~15 ppb vs. 3 ppb detection limit. If the predictions differ from the experiments there also, this tells us that experimental shortcomings cannot explain the differences and the explanation may lie in the (chemistry?) models. For example, the LES results are absent from Fig. 2, while they need to be included.

As mentioned above, both regions of the flame have been analyzed. However, only the recirculation zone is analyzed in greater detail with the LES results. The far-field is simply a turbulent jet, which has been studied in detail by both DNS and LES (with conclusions leveraged in our analysis here), as reviewed in the Introduction of the manuscript.

b) The authors should include flamelet comparisons at more than just one scalar dissipation rate (1 $1/s$ in the data shown in Fig. 3 presently). Also, why did the authors choose this value? Is it representative of conditions in the LES?

Ideally, the authors should show that their conclusions on chemistry effects are robust with respect to changing scalar dissipation rate. If the relative reduction of PAH species w/ H_2

changes with scalar dissipation rate, this aspect should be addressed thoroughly, especially if the effect appears to saturate at low scalar dissipation rates.

c) Related to b) above. The authors should explore the sensitivity of the PAH yield to changes in scalar dissipation rate from flame 1 to flame 2 (the 1.36 factor...) and compare to the effect of chemistry. This analysis would support statements on soot yield in the far field.

In response to comments 8b and 8c, we note that we have investigated other scalar dissipation rates as well. The representative scalar dissipation is $O(1)$, and, therefore, $1/s$ was chosen. As for higher scalar dissipation rates, such as $10/s$, PAH reduction with hydrogen addition is about a factor of 2.5. At lower scalar dissipation rates, PAH shows substantial departure from the steady flamelet [5], so such scaling would not be expected to apply. This is discussed in the revised manuscript.

d) p. 6. The statement ", ...indicating that chemical effects alone are responsible..." is too definitive given the limited scope of the present work. It should be replaced with a more circumstantial sentence with "suggests based on the present scope of work".

This sentence is modified accordingly in the revised manuscript.

e) The authors should show a figure similar to Fig. 7 (sources) for the far field (see also comment 9) below)

9) In Fig. 7 (and the newly proposed figure -- see 8(e)), the authors should include also data from the C₂H₄ flame, so that direct comparisons can be made.

In response to comments 8e and 9, as mentioned above, the currently study is focused on the recirculation zone since the far field is quite similar to that of a nonpremixed jet and has been investigated through other DNS and LES studies. Therefore, considering the scope of the current work and the word limit, the far field information is only briefly discussed in the manuscript.

Reviewer #2: (all replies in **bold**)

The paper presents a combined numerical and experimental investigation of the effects of H₂ addition in an ethylene turbulent flame on the formation of soot. The paper is well written and easy to follow. As you will see below, I have some two major comments: there is no discussion at all of differential diffusion; and some of the conclusions are in fact no more than either coincidence or correlation. I wish the paper had more space to discuss completely all these issues.

* Major comments

1. The paper is organized around the idea there are only two possible effects: hydrodynamics and chemical. But how differential diffusion. After-all, there are 3 terms in each species transport equation (convection, diffusion, reaction) and H₂ is a very light species and will diffuse very fast.

- What is the Reynolds number in the recirculation region? On page 5, it is said "turbulence intensity is low".

- What is the size of the Kolmogorov length scale compared to that of the flame thickness?

- Can we really assume that the effective Lewis numbers of hydrogen (H₂ and H) is unity?

- The authors should clearly state throughout the paper that the Lewis numbers are all set to unity. Especially on page 6 with Fig. 3.

A detailed analysis of differential diffusion is provided in Ref. 46 for the same configuration with a similar fuel mixture. In that work, unity Lewis number flamelets were found to reproduce the temperature and stable species more accurately than detailed

transport. Therefore, differential diffusion is not expected to be important. This issue is clarified in the revised manuscript.

2. Page 4 (right before section 4). While adjusting the boundary conditions might be acceptable, it does need to be justified and explained. Simply stating "Following from the results of a boundary condition sensitivity study" is not a justification. Was that done as part of a previous study (already published)? The authors should explain 1) why the changes were made and 2) in what way they are physical and not purely arbitrary to get better comparison with measurements.

The change of boundary condition was made to be consistent with the previously published neat ethylene flame. In the previous study, this change was guided by matching specific features of the experimental profiles near the nozzle since detailed measurements of the velocity are not available. More details regarding this issue have been provided in the previous publication.

3. Throughout the documents, intermediate "conclusions" are drawn which are not really possible.

- Page 5, the authors state "The difference in the soot reduction with hydrogen addition indicates different soot reduction mechanisms and different roles that hydrogen addition plays in these two regions." I am not sure we can draw such conclusion. I would agree that the relative importance of the various processes/mechanisms might be different; but not that these processes/mechanisms are different in the two regions. Or at least, Fig. 1 and 2 alone do not support this conclusion.

It seems that the reviewer does not disagree with these intermediate summaries that help structure the discussion; rather, the reviewer offers some suggestions on the wording. The above mentioned sentence is changed to "...indicates a different dominant soot reduction mechanism".

- Page 7, I am not sure what is the cause-consequence relation leading to the sentence "Therefore, the LII measurements may underresolve the soot volume fraction by as much as 3 ppb". Please explain/revise.

As mentioned in the manuscript, because the LII detection threshold on the soot volume fraction is 3ppb, the measured value can be low by as much as 3 ppb. This is clarified in the revised manuscript.

- In the conclusion, I am not sure we can use the word "quantified" to state we used laminar steady state flamelets to "quantify" effects in turbulent flames.

- Conclusion: "Therefore, the chemical effect is dominant in this region". The paragraph discusses at best a correlation (at worst only a coincidence). The cause-consequence relation is not proven.

- Conclusion: "This hydrodynamic effect together with the chemical effect accounts for the soot reduction in the recirculation zone." In the LES, maybe. In the experiments, not proven.

Unfortunately, due to the limited information that can be obtained by direct measurements, LES was performed to understand the role of these effects. The LES model

has been validated through previous studies in the cited literature, and, in the current study, comparisons based on the soot volume fraction qualitatively validate the computational results.

* Minor comments

1. Typos: page 2 "The objective of the this"

The typo is corrected in the revised manuscript.

2. Missing ref for the "lumped PAH species" approach.

The definition and reference of lumped PAH species is clarified in the revised manuscript.

3. It would good to relate the select value for χ (1/s) on page 6 and Fig. 1 to certain locations in the flame. Stated differently, can the authors state that this value corresponds to $x/D=...$ from the LES results? Also, isn't that value a little small for the "downstream jet-like region"?

The scalar dissipation rate in the flow field is of order 1. We have also performed flamelet calculations for $\text{CHI} = 10/\text{s}$, which gives a factor of 2.5 reduction of PAH with hydrogen addition, which is quite similar to what we reported in the manuscript with $\text{CHI} = 1/\text{s}$. Such discussion is added to the revised manuscript.

4. Fig. 6b, make the vertical dashed line thicker.

We revised the figure correspondingly.

5. I assume that the steady state flamelet calculations shown in 3 were done with removal of PAH from the gas phase. Please confirm. Also, what are the species contained in "PAH" on that plot?

The reviewer is correct in their assumption: in the steady state flamelet calculation the PAH was removed from the gas phase. Eight aromatic A2 to A4 stable aromatics are considered as PAH. The lumped PAH approach has been reported in the previous study, which can be found in the Introduction.

Reviewer #3: (all replies in **bold**)

The paper presents an experimental and numerical study of the addition of H₂ on the production of soot in non-premixed bluff body ethylene flames. Not surprisingly the addition of H₂ reduces soot by reducing the production of PAH. Although the simulation strongly underestimates the decrease of soot, the authors propose an analysis of the mechanisms responsible for the reduction. The paper is well written and contains interesting data. The numerical approach is one of the most advanced in the field to compute soot in turbulent flames. The main weakness of the paper is to not propose an explanation for the underestimation of soot decrease by the addition of H₂. The effect is very strong in the experiment and one would expect that it is reproduced by the modeling approach used in the paper. It is also suggested to specify in the figure caption if it is experiment or numerical data.

One reason for the potential overprediction of soot volume fraction in the LES was provided in the manuscript: potential underestimation of the soot volume fraction in the experiments due to the fact that the mean volume fraction is very close to the detection limit. This could account for a majority of the observed discrepancy. An additional explanation could be that the recirculation zone is slightly too rich in the LES compared to the experiment, for which experimental diagnostics are only recently available [Buxton et al., 51st AIAA Aerospace Sciences Meeting, 2013-0479] and have not been applied to this flame. This latter point is added to the revised manuscript. Additional characterization of this flame is certainly an interesting area of future work. Clarifications in the figure captions are implemented according to the suggestions.

Reviewer #4: (all replies in **bold**)

The authors investigate the sooting behaviour of a turbulent bluff body stabilized ethylene/hydrogen flame both experimentally and numerically: Laser-induced incandescence (LII) was utilized to measure the soot volume fraction in the flame whereas an integrated Large Eddy Simulation (LES) model was adopted to determine the interactions between soot, turbulence, and chemistry numerically. The results obtained in the ethylene/hydrogen flame are compared with those of a neat ethylene counterpart flame which was analysed previously by the authors.

Similar to the neat ethylene bluff body flame, the authors observed experimentally three distinct flow regions: a sooting recirculation zone, a non-sooting, high-strain neck region, and a sooting jet-like zone. Although the ethylene/hydrogen flame is overall significantly less sooting than the neat ethylene flame, soot reduction in the recirculation zone near the bluff body is more pronounced than in the downstream jet-like region.

From a steady flamelet calculation the authors quantify the chemical effect. Due to the hydrogen addition, PAH mass fraction was found to decrease by a factor of two, indicating a factor of four decrease in the PAH-based soot growth rate by nucleation and PAH condensation. This is consistent with the soot reduction in the downstream jet-like region of the ethylene/hydrogen flame, where PAH-based growth is dominant. The authors conclude from this finding, that the chemical effect is dominant in this region. The hydrodynamic effect was then analyzed, since both the experimental measurements and LES calculations demonstrated additional soot reduction in the recirculation zone, which could not be explained with the chemical effect alone.

The authors identify that both, a hydrodynamic effect together with a chemical effect accounts contribute to the soot reduction in the recirculation zone of the ethylene/hydrogen flame. In summary, the paper reveals interesting new results and the conclusions drawn from the findings of the authors sound reasonable (with the exception of point 6 - see below). Therefore, to my opinion the manuscript should be published after some revisions (recommendation: good/very good).

It is a major weakness of the manuscript that no details about the bluff body flame is given (see also point 1 below). Furthermore (as a consequence), the reader can hardly judge whether the findings are a specialty of the flame investigated by the authors or whether the results would generally be found in bluff body flames.

As mentioned by the reviewer, the reference to the sketch of the bluff body configuration is already provided. However, the reviewer failed to notice that details about the dimensions have already been specified in the first paragraph of Experimental Methodology.

Additionally, there are some minor points which should be addressed in the final version of the manuscript:

1.) The reviewer really miss a detailed figure of the burner configuration and its dimensions. Instead, the authors mentioned two older articles from 1996 (reference [22]) and 1998. This does not really help, because (1) the design is of importance with respect to the discussion and understanding of the hydrodynamic effects of hydrogen addition on soot evolution. Additionally,

(2) figure 1 in reference [22] is - see figure caption in this latter reference - is only a 'sketch of a typical bluff-body-stabilized flame....'. Therefore, also in this reference the reader do not find any further details of the flame.

This point has been addressed above that the detailed geometry is given in the first paragraph of Experimental Methodology.

2.) Figure 1 is tiny - especially the right part of the figure.

The figure has been enlarged in the revised manuscript.

3.) Page 3, first sentence: Please, give also the velocity for the pure ethylene flame - don't impose the calculation of this value on the reader of the manuscript.

The ethylene inner jet has a velocity of 74.2 m/s. This is added to the revised manuscript.

4.) Page 5, second section: 'As demonstrated in Fig. 2, the axial profiles of the radially integrated soot volume fraction (ppm-mm²) are compared for the two flames.' As mentioned in the figure captions, the extension 'per unit height' is missing. Additionally, the minus sign between ppm and mm² is misleading.

The y-axis label has been modified in the revised manuscript.

5.) Page 5, last section: Please let the reader know the fuel jet to air co-flow momentum flux ratio for both flames. Alternatively, the percentage change should be mentioned. Again: please don't let the reader calculate these values.

The fuel jet to coflow momentum flux ratios in both flames and the relative momentum flux ratio has been added to the revised manuscript.

6.) Page 5, last section: 'As Dally et al. [46] demonstrated, the increase in the fuel jet momentum flux decreases the strength of the mixture in the outer vortex in the recirculation zone, further inhibiting soot formation.' This argument can hardly be understood without reading the details of reference 46: If the strength of the mixture in the outer vortex in the recirculation zone is decreased, the fuel is less diluted with ambient air. Therefore, the reviewer would expect an increase in soot formation rather than a decrease. Maybe I understood the expression 'strength of the mixture' (better mixing?) incorrectly. - Page 7 last section: "As discussed above, the increase of the fuel/coflow momentum flux ratio inhibits entrainment from the jet, resulting in a leaner recirculation zone than the neat ethylene case (recall that the stoichiometric mixture fraction is essentially the same for the two mixtures)." If the recirculation zone is leaner mixing seems to be promoted rather than being inhibited? To my opinion, the discussion in this context is not consistent and confusing. The authors should make clear what they mean.

The mixing effect is clarified in the revised manuscript: the recirculating motion of the outer vortex, whose momentum is set by the unchanged coflow, transports rich mixture fraction from the fuel jet to the recirculation zone. In other words, the mixture fraction in

the recirculation zone is set by the rate at which the outer vortex entrains fuel from the central jet. When the jet velocity is increased, among other effects, the momentum of this vortex decreases relative to the central jet, so this vortex is less effective at disturbing this high-momentum jet, reducing the fuel entrainment rate and the recirculation zone mixture fraction.

7.) Figure 4: Please, indicate the difference of both figures in the figure caption.

The figure caption is revised accordingly.

8.) Page 7, first line: 'Quantitatively, the soot volume fraction of the ethylene case is slightly underpredicted but within experimental uncertainty [15]. For the hydrogen added case, the soot volume fraction appears to be overpredicted.' Please, add "calculated" (soot volume fraction) in both cases.

Revised.

9.) Page 7, last section: 'The mixture fraction radial profiles from LES as well as the characteristic time scales of the soot formation and oxidation processes from a lightly strained ($-\chi_{st} = 1 \text{ s}^{-1}$) steady flamelet solution are shown in Fig. 6.' Why is this value chosen for χ in the simulations? Please, explain because the authors observe a high-strain neck region.

From the resolved mixture fraction profile in Fig. 6, it can be estimated that the scalar dissipation rate is of order 1/s. As mentioned above in response to the other reviewers, higher scalar dissipation rates such as 10/s have been investigated, and the PAH reduction with hydrogen addition increases from a factor of 2.0 to a factor of 2.5.

10.) Figure caption 6: 'Horizontal lines' Please, add black to horizontal lines.

The manuscript is revised as suggested by the reviewer.

11.) Figure 7: The orientation of the figure should be changed (r/D_b should be the horizontal direction, x/D_b the vertical). Additionally, explanations given in the last section of page 8 should indicated by e.g. arrows in this figure. Furthermore, for comparison reasons it would be helpful, if also the results of the neat ethylene flame would be given in the figure.

Since the source terms in the ethylene flame are already published in the literature, they are not repeated here due to the word limit. The orientation is kept the same as in the previous published work to facilitate comparison between the different studies.

Reviewer #5: (all replies in **bold**)

The paper presents experiments on the evolution of soot in a turbulent non premixed ethylene/hydrogen (2:1 by volume) diffusion flame. The experimental setup used was kept the same as for a previous neat ethylene case of the same research group. The operating conditions are identical to this case allowing a comparison of the results from both cases. Soot measurements are performed using calibrated LII, the experimental method and data evaluation for the LII measurements being well elaborated also in previous work of the authors.

Soot modeling was performed employing an approach where the soot-chemistry-turbulence interaction is aided by a statistical soot model, a modified Radiation Flamelet/Progress Variable (RFPV) combustion model for sooting flames, and a presumed subfilter PDF for closure. All details of the integrated LES model for sooting turbulent non premixed flames have been developed are state of the art tools for the research group and have been applied for a variety of sooting flames in previous work.

The experimental finding is that the maximum soot volume fractions in the jet-like region of the ethylene/hydrogen case are significantly lower than that of the neat ethylene case. Also, in the recirculation zone above the "bluff body" soot volume fractions are substantially lower.

Flamelet calculations demonstrated that hydrogen addition suppressed soot formation due to the reduction of the C/H ratio, resulting in an estimated fourfold reduction in soot volume fraction due to chemical effects. The observed soot reduction in the downstream jet-like region of the flame in the experiments is qualitatively consistent with this chemical effect. The authors found,

however, soot reduction in the recirculation zone above the "bluff body" substantially larger than the flamelet analysis suggests. Therefore, the authors performed Large Eddy Simulation to investigate soot evolution in the recirculation zone above the "bluff body" and to elucidate the role of hydrogen addition there. They found that for the same jet Reynolds number as the neat ethylene case ($\sim 30,900$), the addition of hydrogen requires a higher jet velocity, and this leads to a leaner recirculation zone that inhibits soot formation and promotes soot oxidation.

The paper is well written and structured. All the experimental techniques and modeling approaches are of high quality and highly estimated in the combustion community. My concern with this paper is, that the progress with regard to experimental techniques and modeling of sooting turbulent flames is marginal. The effects of hydrogen addition to hydrocarbon flames in suppressing soot are known since decades from experiments and they are obvious from the reaction mechanism (HACA mechanism) underlying numerical simulation of soot formation and oxidation. Numerical simulation of sooting flames doped with hydrogen can be found also in the literature. All details of the soot modeling and of the LES model have been presented by the authors (in changing combinations) in previous work. The hydrodynamic effect which has been identified by means of the LES is not surprising and the leaner recirculation zone for the jet with higher jet velocity may be estimated with simpler tools.

(The reviewer recommends avoiding the notation "bluff body flames" for this arrangement. In hydrodynamics a bluff body consists of a surface which is fully immersed into the main flow. This is not the case for this arrangement, where the "bluff body" can be regarded as a considerably thickened wall of the burner nozzle.)

The reviewer has misunderstood the purpose of the paper, which is NOT to develop new experimental techniques or computational approaches. As the above four reviewers commented, “the paper stands as one of the very few LES of sooting flames with comparison to experiments and much is to be learnt from it.” With this, we strongly disagree with Reviewer 5 that the hydrogen addition effects on ethylene bluff body sooting flames can be elucidated with simple laminar flame calculations considering only the HACA mechanism (surface growth) and simple estimation. First, we have demonstrated that the PAH-based growth mechanism dominates the surface growth mechanism in the downstream region, and the former explains the chemical effect in all regions of the flame. Second, we have demonstrated that, besides the chemical effects, hydrodynamic effects (i.e., turbulence) are also responsible for soot reduction in the recirculation zone, requiring a sophisticated computational tool. (As a minor note, “bluff body flame” is ubiquitous in the community for this configuration.)