2nd Rebuttal ACPD

BOLD = reviewer comment
Italic = answer to reviewer comment
Red = highlighted changes in manuscript

The paper presents a machine learning approach to assess the impact of several meteorological features on air quality in Paris metropolitan area. A tree-based machine learning algorithm is used for modelling and a Shapley Additive Explanation is applied to interpret the resulting models. This is a quite interesting study that requires, however, major revisions before a possible publication.
In the next version, the authors must address all the points as follows:

1) The abstract should be improved. On one hand, the it is too long. On the other hand, important information is missing, such as the accuracy/performance of the models.
Thank you for your evaluation. We carefully considered your concerns and took great care to adequately address these.
In the abstract, the following changes were made:
L4: removed "as the effects of meteorological variables are not easy to separate and quantify."
L7-11: added "The model is able to capture the majority of occurring variance of mean afternoon total PM$_1$ concentrations (coefficient of determination (R^2) of 0.58), with model performance depending on the individual PM$_1$ species predicted"
L11/12: shortened the sentence to make it more concise
L24/25: shortened the sentence
L18/19: deleted sentence
L24/28: replaced sentences with "High-resolution case studies are conducted showing a large variability of processes that can lead to high pollution episodes."
L33/34: changed to "...to adapt policy measures, issue warnings to the public, or to assess the effectiveness of air pollution measures."

2) Even if the approach is interesting, it is a quite local study (Paris area). I would like that the authors provide a further discussion about the general impact of their work. In other words, you should discuss to which extend the study has implications in other urban areas worldwide, inclusively cities with more complex terrains than Paris.
We agree that further discussion about the implications of this local study for other urban and suburban regions increases the value of our findings. We have added the following thorough discussion on this (L490-509) and feel that this has helped improve the manuscript. "*The presented findings refer to the SIRTA supersite but the results are nevertheless transferable to other regions as well. For example, the importance of temperature-induced particle formation processes have been shown for the U.S.A. (Dawson et al., 2007), Europe (Megaritis et al.,*

2014), and China (Wang et al., 2016). Hence, it is likely that the detailed, species-dependent disclosure of the nonlinear relationship between temperature and PM1 of this study holds for other urban and suburban areas. This has implications for the PM concentrations in the context of climate change. The empirical perspective of the current study complement to the findings of various modelling studies (Dawson et al., 2007; Megaritis et al., 2013, 2014; Sá et al., 2016; Doherty et al., 2017).

Furthermore, the impact of shallow MLHs on PM1 concentrations investigated here is comparable to results found in a previous, regional-scale study over central Europe that highlighted the dominant role of MLH on PM10 concentrations (Stirnberg et al. 2020).

The importance of wind direction highlights the role of advected pollution by remote, highly polluted urban or industrial hotspots. In general, the interpretation of pollution advection patterns requires knowledge on source regions and terrain. Here, the Paris agglomeration is a major source of pollutants while the relatively flat terrain allows unimpeded advection of air masses.

Urban areas in a more complex terrain would likely be affected by slightly different and possibly more complex mechanisms., such as terrain- and meteorology-dependent air stagnation events (Wang et al. 2018) as well as orography driven wind and precipitation patterns (Rosenfeld et al., 2007).

Still, given the task of disentangling the impact of the various meteorological drivers on air quality is already a complex scientific subject, a continental, flat terrain city such as Paris was chosen as the subject area precisely to exclude other factors (such as orographic flow, or sea breeze) that would add further complexity. Certainly, the methods developed here could be transferred to more urban areas in more complex settings in the framework of future studies.

3) Lines 163-164. The sentence "Note that PM1 data is not normally distributed, i.e. there is more data available for mid-range concentrations" is awkward. Is it not a characteristic of a normal distribution to have more data in mid-range? Please, clarify this sentence.

Yes, this sentence was indeed awkwardly formulated. It was changed to "Note that PM1 data is not uniformly distributed, i.e. there is more data available for mid-range PM1 concentrations."

4) Section 4.1. The ten models you are talking about are not clear. More details must be provided regarding what is modelled by each model and the acronyms BCwb, BCff, etc… have to be defined.

The ten models are mentioned in L177. To make this clearer in the manuscript, we added in L208: "The performance of the species and total PM1 models, each with ten model iterations (of which each has different hyperparameters),..."

Added in caption of Fig. 3:

- *(Org: organics, NH4: ammonium, SO4: sulfate, NO3: nitrate, Cl: chloride, BCff: black carbon from fossil fuel combustion and BCwb: black carbon from wood burning)"*
- *...between the ten model iterations."*

5) Section 4.2. Why did you focus on temperature, MLH and wind direction, only? Considered that NO3 fraction and Wind Speed are also strong drivers, why did you skip a deep interpretation of the effect of these variables, as well?

- *As pointed out in chapter 4.1, the NO3 fraction is found to vary with PM1 total mass concentrations. Lower wind speed generally leads to higher particle concentrations. Temperature, MLH and wind direction, on the other hand, require an in-depth analysis of the different PM1 species, as changes of these variables causes nonlinear responses in PM1 predictions, which vary also between species.*
- *To make this clearer in the manuscript, L221 was changed: "Lower wind speeds generally lead to higher particle concentrations (see Fig. B2) due to a lack of dispersion (Sujatha2016). Temperature, MLH and wind direction require an in-depth analysis, as changes of these variables cause nonlinear responses in PM1 predictions, which vary also between species. "*
- *Added "(see Fig. B)" in L 221*
- *Plots showing the influence of wind speed and NO3 fraction, respectively, were added in the appendix*

6) Line 247. Change "Fig. 6" to "Fig. 5-7".
Done

7) Lines 282-285. You noticed that north/north-eastern winds increase air pollution and you conclude that this pollution should come from Paris, which is located north-eastern from SIRTA. Did you confirm this assumption by analysing wind data from the Airport Charles de Gaulle? If the hypothesis is true, bad air should come from south/south-western in this case. Right?
No PM1 data is available to us from Charles de Gaulle (we used only MLH data for a limited period). This is why we point out that advected particles come from the Paris region and/or continental Europe, as suggested by previous studies. For example, results by Petit et al. 2017, who compared PM1 concentrations at different locations in France during a high-pollution episode suggest that long-range transport of polluted air from continental Europe can be a dominating driver. Our approach is not well suited to distinguish between long-range advection from continental Europe or the Paris region as we focus only on one station.

8) Section 4.2.4. It is not clear which species you are interested in for the interaction analysis. Is it PM1, only? Please, be more specific.
L310: added "Pairwise interaction effects, where the effect of a specific predictor on the total PM$_1$ prediction is dependent on the state of a second predictor, are analysed in the model."

9) Figure 8. How do you explain the red cluster on the top-right corner of the right panel? In other words, how do you explain that high wind speed and high MLH tend to increase the Shap values?
A physical explanation could be the more effective transport of SO4 and its precursor SO2 and ammonium nitrate under high-MLH-conditions and stronger winds (Pay et al., 2012).
L325: added "High MLHs in combination with high wind speeds, however, increase SHAP values. A physical explanation of this pattern could be the more effective transport of SO4 and

its precursor SO_2 as well as ammonium nitrate under high-MLH conditions and stronger winds, and increased formation rates of secondarily formed particles as mentioned in chapter 4.2.2"

10) Section 4.4, 1st paragraph. This paragraph should be reorganized. You give several details about Figs 11-16, which are irrelevant here (lines 361-363). On the other hand, this information misses in the caption of these respective figures.
Paragraph was moved to the caption of Fig. 11.

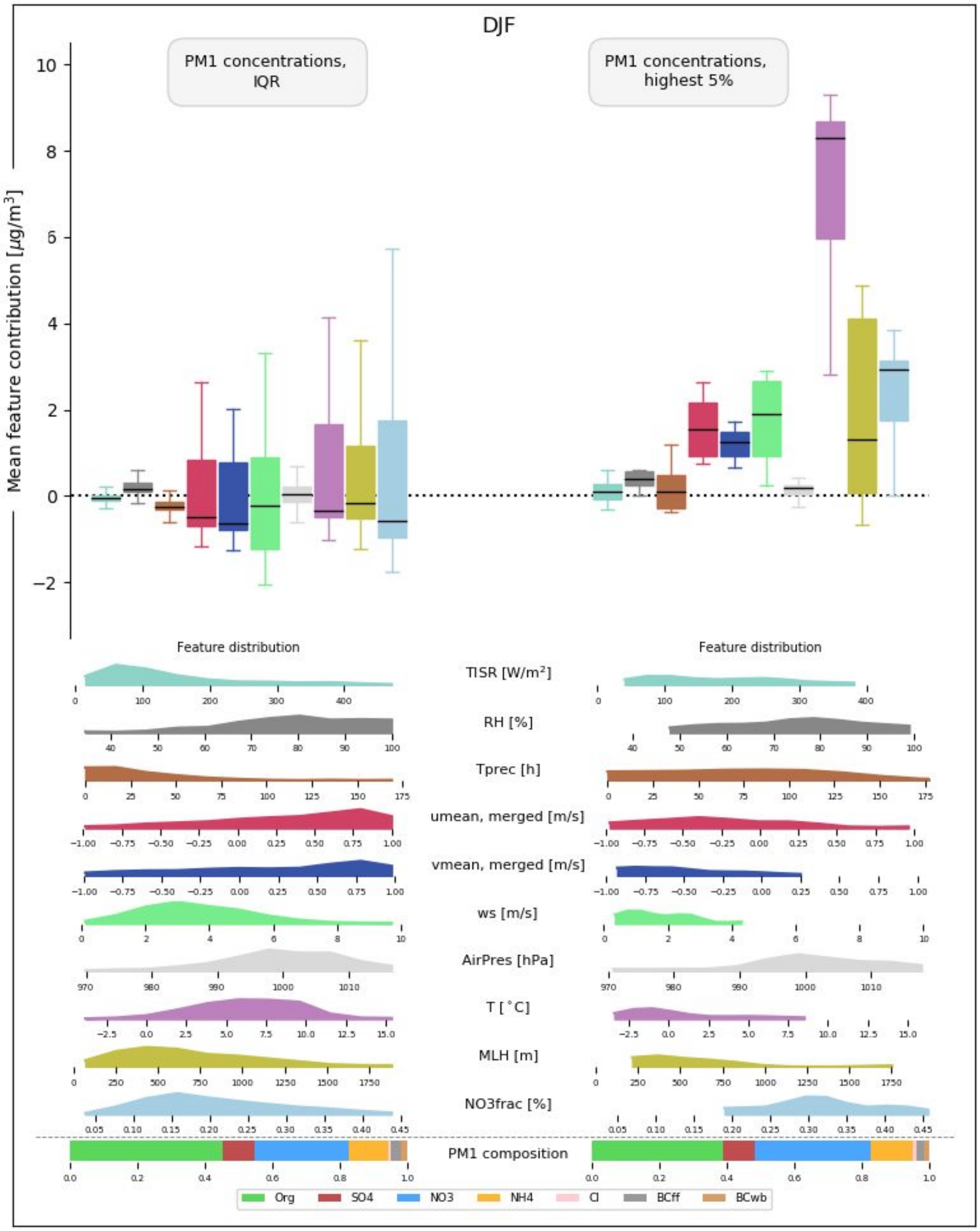
11) Line 393-395. You explain the high pollution in terms of weak "north-north-easterly winds, i.e. a regime of low ventilation". However, it can also be a weak wind that brings pollution from Paris. Please, comment on this point.
Yes, it is true that advection also plays a role here (can be deduced from u-wind SHAP values). Since wind speeds are low and MLHs are also low, thus impeding effective transport of air masses, advection is expected to play a minor role.
L401: added … "and possibly some advection of polluted air from the Paris region"

12) Figures 11-14. The quality of these figures must be improved. First, the legend is too small. Second, indexes a)-h) are missing in Fig. 11. Third, it is not straight-forward to understand the matching between the bar/scatter plots and the right/left side of the Y-axes. Finally, you do not describe in the caption how the predicted vs the observed PM1 are represented. So, the caption needs to be improved, based on my comment 10, as well.

- *Legend sizes in Figs 11-14 were increased*
- *Indexes a)-h) were added to Fig. 11*
- *The caption was extended*
- *An explanation for left and right y-axes was added*

13) Conclusion. We understand that your models do a better job in Winter and Summer than in Spring. So, what about Fall? Why do you not present data for this period? Is it also more difficult to do a good prediction at this season? If, yes, can we conclude that the approach is less suitable for the midseason, maybe because the meteorological conditions are less "extreme" (e.g., average temperature)?

- Fall is very similar to winter in terms of prediction accuracy and drivers of high-pollution situations, except for the occurrence of temperatures below zero, which have a distinct positive influence on PM1 concentrations in winter.


DJF
PM1 concentrations, IQR
PM1 concentrations, highest 5%
Mean feature contribution [μg/m3]
Feature distribution
Feature distribution
TISR [W/m2]
RH [%]
Tprec [h]
umean, merged [m/s]
vmean, merged [m/s]
ws [m/s]
AirPres [hPa]
T [°C]
MLH [m]
NO3frac [%]
PM1 composition
Org
SO4
NO3
NH4
Cl
BCff
BCwb

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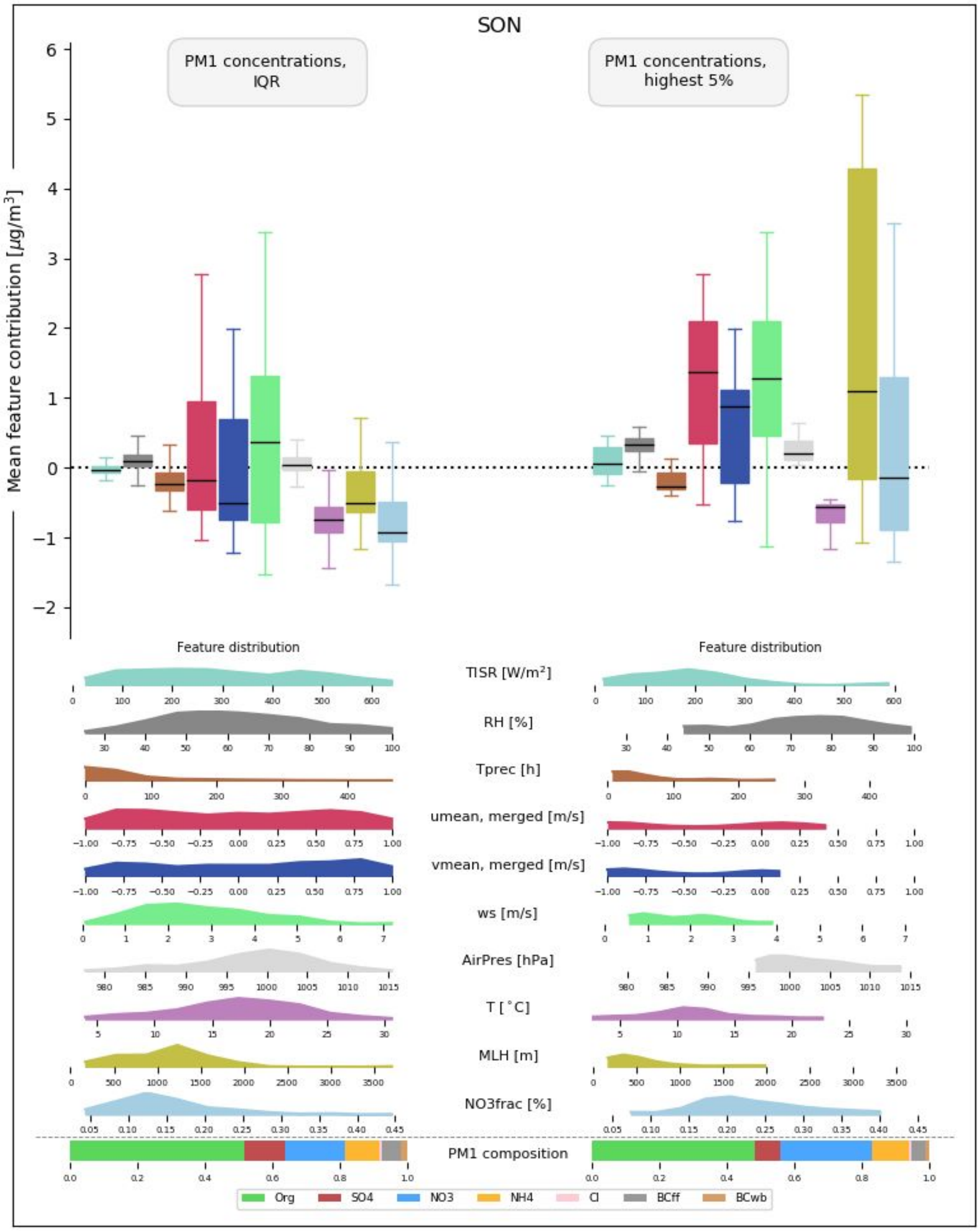

- The reduced prediction accuracy in spring shown in FIg. 14 is due to the exceptional character of this pollution episode. In general, we decided to focus on the most extreme seasons (summer vs. winter) and contrast these in chapter 4.3 and 4.4. T

14) Lines 474-475. Which evidences support this quite strong statement. More arguments are expected, especially to address my comment 2).

Please see changes referring to your 2nd comment.

15) Conclusion, last paragraph. This paragraph is very redundant. We understood at the first sentence that a meteorological prediction is important if we want to use your approach. However, it seems that you repeat the same idea again and again. The proof is the fact that the word "expected" appears three times in the next sentences. This last paragraph must be improved by reorganizing its structure.

The last paragraph was restructured. The idea of this paragraph was actually to convey three different ideas, i.e., three different possible approaches of the presented model approach.

- *Preventative warnings to the public based on knowledge of meteorological conditions exacerbating air pollution*
- *Quantitative analysis of the effects of air pollution measures*
- *Pollution forecast based on short-term weather forecasts*

These different ideas are hopefully conveyed more concisely now.