

## *Remote sensing of exceptional winter aerosol pollution events and representativeness of the surface – column relationship over Paris metropolitan area*

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In the following, *reviewers' comments are in italic blue*. Responses are in normal black font. **Changes in the text are in black bold**. The numbers of the most **major comments** are highlighted in **red**.

### Response to Anonymous Referee #3 – RC1 & RC3

We are very grateful to Referee #3 to have reviewed the manuscript and submitted helpful comments and suggestions to improve both the study and the text. Here we respond to the reviewer point by point.

#### RC1 - Received and published: 21 August 2019

*However, this paper is mainly focused on reporting and describing some very interesting (but necessarily limited) observations. Incidentally, it touches on the issue of the relationship between surface and remote sensing observations, but this is far from being its main topic. If it were, it would use a multitude of datasets to address just this point, and it would not spend much time describing these events themselves in detail. The conclusions drawn are valid for the two events considered, and perhaps for similar meteorological conditions (anticyclonic with a shallow boundary layer). A reader may feel disappointed by how little of the paper is about the topic indicated in the title and abstract.*

*I suggest that prior to considering this paper for publication, therefore, the words quoted above should be removed from the title and the abstract, which would then start with "This work is carried out following a dedicated field campaign in the Paris area (France) during winter 2016-2017", clarifying from the onset what is the main content of the paper. The incidental study on the representativeness is mentioned towards the end of the abstract and this is ok ("During these two events [...] allows us to investigate the representativeness of optical parameters found in the planetary boundary layer to assess surface aerosol concentration."). What is in my opinion should be avoided is to start with a very broad promise, and then not be able to satisfy the reader's curiosity.*

*In summary, the dataset is too limited to be suitable for a general study on the abovementioned representativeness (few observations and specific meteorological conditions). This is was already addressed in the "quick review" report, but it seems that I have not persuaded the authors. I suggest that in the present form of the paper, the authors have not presented sufficient evidence to be able to state that the observations are generally representative of pollution events over Paris in the winter and therefore that the results can hint a relationship between surface and columnar properties, but cannot be considered to be general. I will also try and produce a detailed review, but please consider the above to be the major point to be addressed (in my opinion).*

*Best regards.*

During this first stage of the review process, our manuscript has been quite widely revised following your comments, without losing our scientific objective, mainly dedicated to the interest of lidar measurements to assess the impact of winter particulate pollution.

Still, you consider that the relationship between surface and remote sensing observations is not sufficiently addressed to be the topic of our study. Your comments were overwhelmingly constructive and helpful, yet we partly disagree with you in this last comment. Regarding the finite nature of the

dataset used in the paper, we show in Section 3 that even though the measurements are sampled in a given period of time the dataset are representative of pollution events occurring over the Paris area during winter.

It is difficult to do without the detailed description of the case studies used to establish the relationship between the surface and the PBL. It is indeed important to define them well in order to correctly set the boundary conditions of the study, and this last point answers your objection. Moreover, as we highlight, the two observed events of particulate pollution find equivalents in previous years, which shows that the statistical approach presented in our article is robust and may be generalized to other similar situations that are the majority in winter in the Paris region.

To follow your advice, be more concise and show that our study is mainly based on two major winter pollution events, we have revised the title and the abstract.

RC3 - Received and published: 25 September 2019

*Please refer also to my previous comment on the fact that "surface - column relationship is touched in the paper but not the main topic". I believe that the data collected are very valuable, but my major objection is with the scope of the paper, as explained therein. The paper as it stands raises the reader's expectations a bit too much.*

*Detailed suggestions follow below:*

➤ **MAJOR COMMENTS:**

- 1) *I suggest to modify the title and abstract as explained in my previous referee comment (21 August). This is the major objection I have to the paper as it stands now.*

The title of the paper has been shortened and modified to ensure that there is no ambiguity as to the scope of its results:

**"Remote sensing of exceptional winter aerosol pollution events and representativeness of ground-based measurements"**

The two first sentence of the abstract have also been removed.

- 2) *Another statement that I think could be reviewed on the basis of the above is on page 3, lines 6-7.*

Changes have been made in the text:

"Hence, the main purpose of this paper is to **describe the meteorological conditions that underlie the establishment of significant winter APEs, characterize observed APEs using in situ and remote sensing data and finally** investigate the link between ground-based aerosol measurements and particles trapped within the winter PBL."

- 3) *Angstrom exponent, page 4, lines 25-28. A few points should be clarified in my opinion: (a) when you say "constant" do you mean constant with height or with time? (b) is the Angstrom exponent an instantaneous value or a daily average? (c) the last sentence is unclear (what assumption and what has the horizontal advection to do with it?).*

(a) Here, "constant" means constant with height, the sentence has been modified in this way:

(b) The value of Ångström exponent taken is a mean value over the studied period, e.g. a mean value of Å from the 20<sup>th</sup> to the 23<sup>rd</sup> January. Indeed, during such event this value does not present significant

variations. Moreover, E. Dieudonné et al (2017, referenced in the paper) show that the constant angstrom hypothesis induces a maximum relative uncertainty of 4% in the determination of the LR.

(c) The assumption of a constant Ångström exponent is obsolete in presence of different types of aerosols in the atmospheric column. Indeed, assuming a constant Å in that case would result in an enhanced error on the N<sub>2</sub>-Raman derived AOT. Thus, we have to argue that we met these conditions under the meteorological context that occurred at the time. See modification

The text has been modified following the three points discussed: (p.4 l.24 to 29)

**“In the inversion process, the extrapolation of AOT measurements from the N<sub>2</sub>-Raman wavelength to the elastic wavelength assumes a constant Ångström exponent with height for the particles in the atmospheric column. The Ångström exponent is derived from the AEROSOL ROBOTIC NETWORK database (AERONET, <http://aeronet.gsfc.nasa.gov/>) for the Paris site. Here a mean value of the Ångström exponent is taken for each APE. This assumption of constant exponent is consistent as all aerosols are concentrated in a well-mixed shallow PBL.”**

[...] (p.5 l.10 to 11)

**“Note that Dieudonné et al. (2017) show that the maximum relative uncertainty associated to LR induced by the constant Ångström hypothesis remains below 4%.”**

- 4) *Spatial averages, page 8, lines 15-16: I suppose that the way the spatial average is done is limits it to the available ground stations, which means a coarse spatial sampling and a limited overall area. Please mention these caveats in the text.*

A sentence has been added following the paragraph discussed to mention this caveat:

**“Even though the network of ground-based stations is designed to be the most representative of the regional air quality, the spatial resolution remains coarse and the average could not be representative of all areas of the Paris region.”**

➤ MINOR COMMENTS:

- 5) *Abstract, line 12: replace "continuously" with "during two 5-day periods" as the lidar was not operated continuously from 1 November to 31 January (see text).*

The sentence has been removed because it was not necessary in the abstract.

- 6) *Abstract, line 13: delete "submicron" (at this wavelength the lidar is also sensitive to supermicron particles) and add "thought to be" before "mainly" (you have no direct measurement of aerosol type/origin).*

The sentence has been removed because it was not necessary in the abstract.

- 7) *Abstract, line 15: explain the method used to determine the circular area and measure its diameter.*

We used maps of ensemble reanalysis of chemical transport model to assess the dispersion of the APE. In the first case it shows a concentrated zone around the Paris region and its surroundings. In the second event of January the APE is spread in England, Northern France, Benelux and Germany. Such maps are available at <https://atmosphere.copernicus.eu/>.

Changes have been made in the text:

“it concerned a circular area of ~250 km in diameter around Paris **as shown by ensemble reanalyses of chemical transport models**”

8) *Abstract, line 17: explain what other information you have to say that the event covered all of Western Europe.*

Same response as for the point 7).

9) *Abstract, lines 17 and 18: explain where exactly the values of 121 and 156 have been observed. Explain what is the value after the +/- sign (experimental error? Variability in time? standard deviation of measurements at different stations?). You give the AEC of the second episode: why not give also the AEC of the first episode?*

The +/- sign is related to standard deviation derived from the spatial averaged.

Changes have been made in the text:

“**The maximum** PM<sub>10</sub> (PM<sub>x</sub> is the mass concentration of particles with an aerodynamic diameter smaller than x μm) **was** 121±63 μg m<sup>-3</sup> (**spatial average ± standard deviation**) **and the aerosol extinction coefficient (AEC) ranged from 0.2 to 1 km<sup>-1</sup>**. The second event took place from **20<sup>th</sup> to 23<sup>rd</sup>** January which covered all of **North-western Europe**, with maxima of PM<sub>10</sub> **around** 156±33 μg m<sup>-3</sup> and AEC between 0.6 and 1 km<sup>-1</sup>, within the winter atmospheric boundary layer.”

10) *Abstract, line 20: the sentence about weather conditions is vague, I suggest to be more specific and describe which type of weather conditions you are referring to.*

Changes have been made in the text:

“However, they did not take place **under identical anticyclonic** weather conditions”

11) *Page 3, lines 8-9: "the most severe winter APEs above the Paris area": specify over which period of time they are the most severe (e.g. "from year Y to nowadays").*

Changes have been made in the text:

“This study is based on a specific field campaign performed during the most severe winter APEs **that occurred** in the Paris area **since 2009**.”

12) *Page 4, line 20: "downgraded" → "integrated"*

Agreed. Changes have been made in the text:

“To obtain a sufficient signal to noise ratio (SNR, **SNR > 10 (Royer et al., 2011a)**) from the N<sub>2</sub>-Raman channel during daytime, the vertical resolution is **set** to 15 m”

13) *Page 5, line 6: "sources of uncertainties" → "uncertainties for our lidar system" (it helps to know that Royer et al is not a generic paper but one that details the uncertainties for this specific lidar).*

Changes have been made in the text:

“The different sources of uncertainties **for our lidar system** are discussed in Royer et al. (2011a)”

14) *Page 8, line 19: specify over which time period the 136 (27) values are valid (is it the 11 years in Figure 2?).*

Changes have been made in the text:

**“Among these eleven winters (figure 2), we count 136 (27) days with at least one station exceeding the information (alert) threshold.”**

15) *Page 8, lines 21-24: the judgment on pollution could be worded differently, relating to the actual data that are shown. "winter 2016/2017 stands out with a large number of threshold exceedances", "2015/16 and 2017/18 had few threshold exceedances".*

Changes have been made in the text:

**“Yet, winter 2016/2017 stands out with a large number of threshold exceedances. In opposition, the previous and following winters, i.e. 2015/2016 and 2017/2018, present few threshold exceedances.”**

16) *Page 8, line 23: I suggest to omit "despite the increasingly coercing political measures to improve air quality".*

Agreed. The sentence has been removed.

17) *Page 8, line 24: expand better on the link between pollution levels and anticyclonic conditions.*

Three sentences have been added in the text:

**“Indeed, despite a general trend in emissions to decline in the Paris region, there are still noteworthy episodes of pollution. When a strong high pressure system sets in over a long period of time, it prevents air mass advection, blocking the weather situation. Thus, the pollution still emitted, even if it is less than in the past, remains blocked by the high pressure system and ends up exceeding the health thresholds.”**

18) *Page 8, lines 29-31: make dates consistent with dates in the abstract, please.*

In the abstract we were referring to the most polluted days of each event. We agree that was not clear, dates in the abstract have been modified.

19) *Page 9, line 1: next to meteorological patterns add "see section 3.2"*

Changes have been made in the text:

**“According to ERA5 reanalyses, the meteorological patterns (see section 3.2) are similar over the 8 days”**

20) *Table 2, caption: add "in winter" after "decade"; explain if the max/min value is instantaneous, hourly, daily, etc. In the table, I would suggest to group the event by winter and not year (e.g. 2007/2008 instead of 2007): this would be coherent with Fig. 2.*

The table header has been changed following your suggestion, and the caption too:

**“The 8 most severely polluted days of the past decade in winter. For each day we give both PM<sub>2.5</sub> and PM<sub>10</sub> measured at ground level (Airparif network) in the format Max/Mean/Min where: Max and Min are the hourly maximum and minimum value measured at a given background station during the day and Mean is the daily average over all background stations.”**

21) *Figure 3, caption: wind velocity and direction at which altitude level? surface?*

Changes have been made in the caption:

“The geopotential altitude (**white lines**) and the wind direction and velocity (**black arrow**) are given at a 975-hPa level.”

22) *Page 12, line 7: add "single" before "grid point" and give lat/lon of the grid point centre.*

Changes have been made in the text:

“We consider the **single** grid point of 0.25° x 0.25° which includes central Paris (**48.875°N, 2.375°E**).”

23) *Fig. 4, x-axis: Month-Day-Hours is confusing; I suggest Day/Month HH:MM. In the caption you should also mention the wind rose.*

The x-axis format has been changed following your suggestion and for more coherence with other figures. Changes have been made in the caption:

“Figure 1: **Wind rose and** temporal evolution of wind intensity and direction **at 10 m** from ERA5 during four days of the 2016 **APE**.”

24) *Page 13, line 8: "clear" → "aerosol-free".*

Changes have been made in the text:

“the sky is rather **aerosol-free**”

25) *Page 13, line 19: state in the paper that you have chosen to keep the data associated with the middle and high altitude clouds and why. Even better, they could be displayed in a different colour for easy identification.*

Changes have been made in the text:

“These discrepancies are mainly due to the presence of middle and high-altitude clouds identified on lidar vertical profiles, which may bias the AERONET operational products (Chew et al., 2011). **As far as lidar data are not disturbed by high clouds these profiles are kept in the figure.**”

26) *Page 15, line 7: are you referring to particle size? I suggest to specify "particle size" after "smaller aerosol".*

Changes have been made in the text:

“These temporal variations trace a diurnal evolution with smaller **particle size** during night time”

27) *Page 15, line 18: if I have understood your reasoning, then the following sentence could be added for more clarity at the end of this line: "We therefore do not believe that the influence of RH on LR is significant".*

Changes have been made in the text:

“As a result, the RH measured by radiosondes and modelled by ERA5 does not exceed 60% while we observe the diurnal variations of the LR (29<sup>th</sup> November to 1<sup>st</sup> December). **We therefore do not believe that the influence of RH on LR is significant, as also demonstrated during the LISAIR field campaign (Raut and Chazette, 2007) for RH < 80%.**”

28) *Page 15, line 28: "shows that the CALIOP and CATS spaceborne observations may be complementary" → "shows the CALIOP and CATS tracks".*

Changes have been made in the text:

“**Erreur ! Source du renvoi introuvable.** shows the CALIOP and CATS **ground-tracks for the January APE**. Within a 24-hour time interval, their tracks are crossing in the middle of France, along a south-north axis for CALIOP and a west-east axis for CATS.”

29) *Page 19, line 3: add "surface" before "PM".*

Changes have been made in the text:

“The temporal evolutions of **surface** PM during the two particulate pollution events”

30) *Page 23, line 10: add "over Paris" after "decade".*

Changes have been made in the text:

“In this paper we investigate the lidar-derived optical properties of two major APEs of the winter of 2016/2017, found to be part of the most severe pollution events of the 2007-2017 decade **over Paris.**”