### Answers to the first referee:

We would like to thank the first referee for its good appreciation and explain with more details some points he raised.

- 1. Vertical gradients in the first layer: The referee wonders the profit of applying a vertical gradient in the first layer in order to take into account the potential influence of dry deposition at ground. At rural sites, measurements are performed at around 3 m above ground level (a.g.l.) and indeed, vertical gradients may exists in the first 40 m a.g.l. due to deposition that decreases concentrations at ground. However, the absence of buildings is assumed to increase the vertical homogeneity, and the measurement can probably be considered as representative. Conversely, urban areas are complex environments and strong vertical gradients may exist in the first 40 m a.g.l., mainly due to the presence of buildings that disrupt the horizontal and vertical mixing. However, as the urban site (PAR) is located on the rooftop of a building at around 20 m a.g.l., the measurements are assumed to be quite representative for the 40 m thick first model layer.
- 2. Uncertainties at the daily scale (page 6405, line 14-20): The referee asks the reasons of the decrease of uncertainties at large time scale (month, year). The decrease of uncertainties when considering monthly or annual average with respect to daily ones comes from a simple mathematical consideration, that the uncertainty decreases with the root of the number of days, when considering errors on individual days as independent. I will add that sentence at 6405-18: "[...] time scale. Such a decrease comes from a simple mathematical consideration, that the uncertainty decreases with the root of the number of days, when considering errors on individual days as independent.".
- 3. Minimum boundary layer height (page 6407): The referee wonders the origin of the minimum boundary layer height (BLH) value (150 m) applied in our simulations. In the CHIMERE model, a minimum BLH may be set over urban areas in order to indirectly correct the absence of urban heat island (UHI) effect (known to increase the BLH) in the meteorological input data. It mainly affects the BLH during nighttime, when the UHI effect is the highest (and the BLH the lowest). Of course, it is a very simplistic correction since this UHI effect is influenced by various parameters (e.g. building density and morphology, anthropogenic heat fluxes) and thus varies depending on the location or the season. Note that this minimum value applies to a 100% urban cell, and decreases proportionally to the amount of non-urban landuse within the cell (the minimum BLH at the SIRTA, a suburban site, where its measurements have been performed) is thus around 120 m). That value of 120 m roughly corresponds to the lowest values (actually, the 2nd percentile) observed at SIRTA during the 1 year round campaign. Additionally, as mentioned in the paper (and reminded by the referee comment of Sandry Pal), one should keep in mind that BLH estimations (from LIDAR observations in our case) during nighttime remain uncertain, and to our sense it thus appears difficult to discuss more deeply that point. I will change at 6409-22-23: "In our case, the value of 150 m is chosen for a 100% urban cell (and decreases proportionally to the amount of nonurban landuse within the cell), based on the 2nd percentile (120 m) of BLH measured at the SIRTA suburban site.".
- 4. Technical comments (page 6394, line 9): ", etc." will be removed.

5. Technical comments (page 6394, line 29): I only mean that the pollution produced locally (associated to local emissions) adds to the regional background. I will simply change the end of sentence into "[...] adds to the local urban pollution increment".

### Answers to the second referee:

We would like to thank the second referee for the detailed and relevant questions he raised, notably concerning our methodology, which give us the opportunity to probably improve the clearness of our approach.

- 1. Back trajectories and determination of the upwind site (page 6402-4-11): The referee asks several questions about the validity of the simplistic method used to estimate advected contributions. Each day, the whole set of 40 back trajectories calculated for one day (10 trajectoires each 6 hours) is considered for the determination of the upwind site and there is no separate determination each 6 hours. As the referee supposed, for a specific day, all back trajectories rarely indicate only one sector (this occurs only 8% of time), but most of time two (43%) or three (47%) sectors. Actually, even if it is not the only one, the dominant sector usually gathers the majority of back trajectories: in average, 73% of back trajectories belong to this dominant sector, against 22 and 5% for the second and the third sector, respectively. The occurrence of stagnant conditions remains very low, with only 4% of hourly wind speed values below 1 m s<sup>-1</sup> over the whole year. However, the most important aspect of our procedure is the calculation of uncertainties, which keep us from developing a more detailed and maybe more realistic (but still uncertain) procedure for the quantification of advected contributions.
- Indeed, whatever the selected upwind site (or even if we would have considered a mean concentration between several sites), by assuming an uncertainty defined as the maximal concentration range among the whole set of rural sites, the value of the advected contribution will always be included in the confidence interval. I will add the following sentences in the revised manuscript at 6402-11: "Due to the complexity of wind fields, this procedure is certainly too simplistic to account for all meteorological situations that may occur over the Paris region (e.g. back-trajectories originating from more than one sector, recirculation). However, all these problems relative to the choice of the appropriate upwind rural site are tackled by the quantification of advected contributions uncertainties in which all the three rural concentration values are included, as described in the next section."
- 2. Advected regional background (page 6402-13-14): The referee then asks several question about the representativeness of the quantified advected regional background. Both RUS and RNW stations take part of the AIRPARIF network and are officially classified as rural according to the criteria defined in the Annex III of the 2008/50/EC Directive. Specifically set for the PARTICULES campaign, the RNE station also follows these criteria and is thus classified as rural. All three sites are located in small municipalities with population density below 100 inhab km<sup>-2</sup>. Both RUS and RNW sites are clearly representative of the rural background. Concerning the RNE site, an influence of local woodburning emissions have been noticed from levoglucosan measurements, and OM concentrations measured at this site cannot thus be considered as representative (and have been removed). As the other chemical constituants are far less (EC) or not (secondary inorganic compounds) influenced by such emission sources,

measurements remain representative of the rural background. representativeness scale of a rural station is difficult to assess precisely since it depends on the heterogeneity of the regional background which in turn is determined by various factors (e.g. location and intensity of emission sources, wind field structure). Except when they are impacted by the Paris plume, these rural sites are representative of the regional background within a large extent. The Paris agglomeration is located in a region mostly occupied by agricultural activities with thus limited PM<sub>2.5</sub> emissions, as illustrated by the PM<sub>2.5</sub> emissions map (Fig. 1 in this Author Comment). The region is rather flat and windly (mean wind speed of 3.1 m s<sup>-1</sup> measured at the ground MONTSOURIS site in the Paris center), and as previously mentioned stagnant conditions are very rare. All these features thus favor a good dispersion of air pollution in the Paris region. Therefore, most of time, the Paris agglomeration cannot influence PM<sub>2.5</sub> levels at all three rural sites, which allows us to assume that the regional background advected toward the city is comprised between the lowest and the highest rural sites concentration. This hypothesis (on which is based the estimation of uncertainties in Sect. 4.2) thus appears quite reasonable. I will add at 6402-20: "Except when they are impacted by the Paris plume, rural sites can be considered as representative of the regional background. As the Paris region is rather flat and windly, stagnant conditions remains very rare (over the year, only 4% of hourly wind speed values measured at the MONTSOURIS site are below 1 m s<sup>-1</sup>), which most of time prevents all rural sites to be simultaneously influenced by the Paris pollution". And at 6402-24: "[...] at this site. The RNE rural site can thus not be considered as representative of the OM rural background, but this local wood burning pollution is not assumed to impact significantly the other species. In order to [...].".

- 3. Back trajectories 4-hours before Paris (page 6410-1-3): The referee asks how an continental origin can be deduced from back trajectories only 4 hours before Paris. The 4-hour backward time period only allows to assess which rural sites have been potentially overflown by air masses before reaching Paris, but is of course insufficient to assess the geographic origin at the continental scale. The link established in the paper between some intense episodes (in particular during wintertime) and a continental origin of air masses from the north-east derived from an analysis of back trajectories a few days before reaching Paris. To avoid any ambiguity, I will change the sentence at 6410-1-3 to: "The variability (standard deviation of 8.6  $\mu$ g m<sup>-3</sup>) strongly depends on the wind regime, with large episodes mostly linked to advection of continental air masses from the north-east wind sector as indicated by back trajectories over a few days."
- 4. Modeled contributions: The referee wonders how modeled contributions are derived. Both the observed and modeled contributions are calculated the same way, after the interpolation of simulated concentrations at the four sites. I will add at 6406-16: "After interpolation of concentrations at all four sites, simulated contributions are derived in the same way as observed ones.".
- 5. Vertical distribution of emissions (page 6401): The referee asks how emissions are vertically allocated in the model. Area and line sources are emitted in the first layer, while emissions from point sources are vertically allocated depending on several parameters (e.g. ambient temperature, ejection speed and temperature, smokestack height). I will add at 6401-8: "Area and line sources are emitted in the first layer, while

emissions from point sources are vertically allocated depending on several parameters (e.g. ambient temperature, ejection speed and temperature, smokestack height)."

- 6. MFB and MFE (page 6406-15): The referee asks about the absence of MFE and MFB definitions in Sect. 4.3 (and maybe in the general discussion). To our sense, MFB and MFE statistical indicators are easier to interprete on a graphic way with the Boylan and Russel performance goals, as in Fig. 13, while the traditional indicators (bias, RMSE) make the discussion easier to understand. This explains why these two metrics are not mentioned before Sect. 5.8.
- 7. OC versus OM: The referee finally asks why comparisons are not performed on OC rather than OM. Actually, while primary organic aerosol emission inventories correspond to OC (due to emission factors mostly measured by thermo-optical methods), the secondary organic aerosol produced in the model cannot be considered as OC but is closer to OM. That is why we apply a conversion to emission input data (1.6 in our case) in order to represent OM concentrations rather OC ones. Additionally, the OM to OC conversion remains mandatory in the comparisons of PM<sub>2.5</sub> total mass.

## **Answers to Sandip Pal:**

We thank Sandip Pal for his interest in the discussion, and the interesting references he proposed.

1. Boundary layer height uncertainties associated to aerosol stratification (page 6409): I agree that some references are missing on that point, and the references you mention are relevant and worth being added. I will modify the text at 6409-9: "[: ::] or in the presence of clouds (Pal et al., 2013; Cimini et al., 2013)" and at 6409-13: "For the Paris megacity, urban-rural temperature contrasts up to  $7^{\circ}$ C have been noticed at night (Lac et al., 2013).", and add these references to the references list.

### Additional modifications:

6398-2 : « emissions factors » → « emission factors »

6400-22-23 : « Ntziachristos et al., 2012 »  $\rightarrow$  « EEA, 2013 »

6410-25: « represents 75 »  $\rightarrow$  « represents 74 »

6411-27-28 : « R below 75% » → « R below 0.48 »

6420-4-5 : « in Supplement »  $\rightarrow$  « in the Supplement »

6420-9: « in Supplement »  $\rightarrow$  « in the Supplement »

6425-20-21 : « +40 and 76% respectively »  $\rightarrow$  « +40 and 76%, respectively »

# References:

« Ntziachristos, L. and Samaras, Z.: COPERT IV documentation – Methodology for the calculation of non-exhaust PM emissions, Tech. rep., EEA, 2012. » replaced by « EEA : EMEP/EEA air pollutant emission inventory guidebook 2013, Tech. Rep. 12, EEA, 2013. »