Response to Referee #2: We would like to thank the referee for the careful review and insightful suggestion throughout the manuscript, which helps us to improve the quality of the manuscript.

Our response follows *(the reviewer's comments are in italics and blue)*

General Comments:

This study attempts to investigate the effects of sequential assimilation of satellite-based aerosol size information (i.e., AÌ□ngstroÌ□m exponents) and aerosol optical depths (AOD) on the analyse of the aerosol concentrations. The assimilation experiments are conducted over the European region with the MODIS Deep Blue products. The results demonstrate that the assimilation of the MODIS observed aerosol size information could improve the surface fine particles analyses by correcting the model assumed aerosol geometric radius and subsequent the AOD observation operator. The paper is generally well written and scientific sound.

Main comments:

Q1. It looks the simulated AÌ□ngstroÌ□m exponents without any data assimilation are too low (below zero). The authours also claim that there are no dust events during the studied period, so does this mean that the default parameters of the aerosol radii for the fine aerosol particles such as the sulfate or carbonaces aerosol are too large in the LOTOS-EUROS model. If this is ture, why the model uses those values.

Reply: The description of aerosol radii in the model is indeed very important. A considerable amount of literatures on ground observations indicated that there is remarkable spatial and temporal variation in the aerosol size distribution, with the geometric mean radius ranging from ten to several hundreds of nanometers (Costabile et al., 2009). It is difficult to describe the spatiotemporal varying features using fixed values like most models do in practice. In addition, also the Ångström exponent measurements are uncertain, and might change with the assumption of aerosol size distribution in different satellite retrieval product. Our hybrid assimilation is therefore designed to solve this issue. Different from a standard AOD assimilation that directly assimilates AOD observations and ignores the potential mismatch of the particle radius distribution, the hybrid approach first estimates suitable aerosol size parameters by assimilating Ångström exponent observations, before performing the AOD assimilation.

To describe this better in the manuscript, remarks are now added in page 15, line 16-20: "***A considerable amount of literatures on ground observations indicated that there is remarkable spatial and temporal variation in the aerosol size distribution, e.g., the geometric mean radius ranging from tens to***

hundreds of nanometers (Costabile et al., 2009). It is insufficient to describe these spatiotemporal varying characteristics using a fixed value as is used in practice. Using a fixed value, the model AOD and Ångström exponents are likely to be strongly biased as will be discussed in Section 4.2, and AOD assimilation result will be misled as is illustrated in Section 5.3."

2. The results in Figure 2 demonstrate that there are some too low AÌ□ngstroÌ□m exponents. Probably, the quality of the satellite retrievals of the AÌ□ngstroÌ□m exponent over such region is not good. How does this unusual observation affect your assimilation result?

Reply: There are indeed some inconsistent MODIS Ångström measurements assimilated as shown in green colored box in Fig. 2b. However, the aerosol radii are assumed to be spatially and temporally constant during the whole assimilation window. The radii would be nudged to fit the dominant MODIS Ångström exponents while less influenced by these few inconsistent data as present. Of course, data quality control should be introduced if the spatial variability of aerosol radii is explored in future work.

To make this clear, remarks are added in page 19, line 27-30: "***The aerosol radii are assumed to be spatially and temporally constant during the short period used for the experiments. Spatially varying radii would of course allow the assimilated Ångström exponent to better fit the MODIS Ångström exponent. However, the locally inconsistent MODIS Ångström observations found during comparison with AERONET observations in Section 3.2 would introduce strong local mis-adjustments in case a (large) spatial degree of freedom is allowed. Data quality control for excluding these polluted data is required. Introducing spatial variations also requires information on spatial correlations and would increase computational costs; hence, this aspect has not been explored in this study.***"

3. As the assumption of diagonal matrix of the model background covariance B for AOD, do you mean only the aerosol mass concentrations over the model grid with MODIS observation could be optimized? How about the model grids without any available observations to be assimilated? Does this induce some unreason aerosol distributions?

Reply: Thanks for the in-depth comment. Yes, we only performed the ***AOD analysis*** over the pix where MODIS AOD is available.

In data assimilation, all states ($\in R^n$) could be optimized by the observations correlated. The spatial correlation (anisotropic) matrix is described using a background covariance matrix **B** ($\in R^{n*n}$) in variational method, or using an ensemble approximated **B** ($\in R^{n*n}$, but much lower rank) in EnKF. In practice, how observations would help improve the state estimation not only depends on the observations, but also depends on the spatial correlation in **B**. For this study, we only aimed to explore how AOD measurements would help/harm the state estimation. To prevent the influence from spatial correlation, we carried out the assimilation in the subspace where MODIS measurements are available and assumed the **B** is independent.

To make this clear, remarks are added in page 19, line 8-12: "***In the background covariance B_τ, the main diagonal defines the assumed variance of the model AOD, while the off-line elements represent the correlations between two AOD values in different grid cells. In this study the focus is on using the available AOD observations to obtain insight in the validity of the assumptions on the aerosol size distribution. Correlations between grid cells that would also influence the assimilation in practice are therefore simply ignored, and all optimizations are done per grid cell.***"

4. P19 Line 29, How to obtain the optimal aerosol radius using a 4DvEnvar? Please clarify it more detail?

Reply: Details of the 4DEnVar are now added in the ***Supplementary material***.

Remarks are added in page 20, line 18-19: "***The aerosol radius ra that minimizes the cost function Eq. (12) is obtained using a 4DvEnvar method (Liu et al., 2008) and the detailed procedures can be found in the Ångström analysis cost function minimization.***"

2 Ångström analysis cost function minimization

The minimization of the cost function follows the 4DEnVar processes. An ensemble of aerosol radius vector are generated randomly using the prior $\boldsymbol{r}_b$ and the assumed error covariance $\mathbf{B}_r$:

$$[\,\boldsymbol{r}_1,...,\boldsymbol{r}_N\,] \tag{1}$$

An ensemble of Ångström model simulations then forward with the ensemble aerosol radius vectors in parallel:

$$[\,\mathcal{M}(\boldsymbol{r}_1),...,\mathcal{M}(\boldsymbol{r}_N)\,] \tag{2}$$

Denote the ensemble perturbation matrix by:

$$\mathbf{L}' = \frac{1}{\sqrt{N-1}}[\,\boldsymbol{r}_1 - \bar{\boldsymbol{r}},\ ...,\ \boldsymbol{r}_N - \bar{\boldsymbol{r}}\,] \tag{3}$$

and mean of ensemble simulation by:

$$\overline{\mathcal{M}(\boldsymbol{r})} = \frac{1}{N}\sum_{i=1}^{N}\mathcal{M}(\boldsymbol{r}_i) \tag{4}$$

where $\bar{\boldsymbol{r}}$ is the mean of the ensemble aerosol radii. In the 4DEnVar assimilation algorithm, the optimal radii $\boldsymbol{r}_a$ is defined as weighted sum of the columns of the perturbation matrix $\mathbf{L}'$ using weights from a control variable vector $\boldsymbol{w}$:

$$\boldsymbol{r}_a \;=\; \bar{\boldsymbol{r}} + \mathbf{L}'\boldsymbol{w} \tag{5}$$

The cost function could then be reformulated as:

$$\mathcal{J}(\boldsymbol{w}) = \frac{1}{2}\boldsymbol{w}^{\mathrm{T}}\,\boldsymbol{w} \;+\; \frac{1}{2}\{\,\mathbf{M}\mathbf{L}'\boldsymbol{w} + \overline{\mathcal{M}(\boldsymbol{r})} - \boldsymbol{\mathcal{A}}\,\}^{\mathrm{T}}\,\mathbf{R}_{\mathcal{A}}^{-1}\{\,\mathbf{M}\mathbf{L}'\boldsymbol{w} + \overline{\mathcal{M}(\boldsymbol{r})} - \boldsymbol{\mathcal{A}}\,\} \tag{6}$$

here $\mathbf{M}$ is the linearization of the LOTOS-EUROS Ångström simulation model required for cost function minimization, and is approximated by:

$$\mathbf{M}\mathbf{L}' \;\approx\; \frac{1}{\sqrt{N-1}}[\,\mathcal{M}(\boldsymbol{r}_1) - \overline{\mathcal{M}(\boldsymbol{r})},\ \dots,\ \mathcal{M}(\boldsymbol{r}_N) - \overline{\mathcal{M}(\boldsymbol{r})}\,] \tag{7}$$

with the uncertainty in radii transferred into the observations space, the minimum of the cost function in Eq. 6 could then be directly calculated, and the posterior emission $\boldsymbol{r}_a$ subsequently be updated.