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How aerosol size matters in aerosol optical depth (AOD) assimilation and the optimization using the Ångström exponent

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1 Supplementary figures

Fig. S1 presents the radius-dependent variation of the extinction efficiency $\mathcal{Q}_{ext}$, total cross section per unite mass $\mathcal{S}$, extinction coefficients ϵ_{ext} as well as the interpolated Ångström exponent $\mathcal{A}$ for sulfate aerosol (so4) at various incident bands and relative humidity. Other inorganic aerosols, e.g., NO_3 and NH_4 have the very close density, hygroscopicity and refraction index. Therefore, they share the same curves to so4.

Fig. S2 presents the profiles of the extinction efficiency $\mathcal{Q}_{ext}$, total cross section per unite mass $\mathcal{S}$, extinction coefficients ϵ_{ext} as well as the Ångström exponent $\mathcal{A}$ for black carbon (bc) at various incident bands. Since the bc fails to absorb any water from the surround atmosphere (hygroscopicity index = 0), bc aerosols have the same optical properties in dry and wet atmosphere.

Fig. S3 presents the curves of $\mathcal{Q}_{ext}$, $\mathcal{S}$, ϵ_{ext} as well as $\mathcal{A}$ for organic carbon (oc). Though oc is capable of attracting moisture from the air, the hygroscopicity is very weak. Therefore, bc aerosols have very close optical properties in dry and wet atmosphere.

Fig. S4 and S5 presents the curves of $\mathcal{Q}_{ext}$, $\mathcal{S}$, ϵ_{ext} as well as $\mathcal{A}$ for sea salt (ss) and dust aerosols. In real situations, most of the ss and dust are in coarse-mode, therefore, they usually result in very small Ångström values.

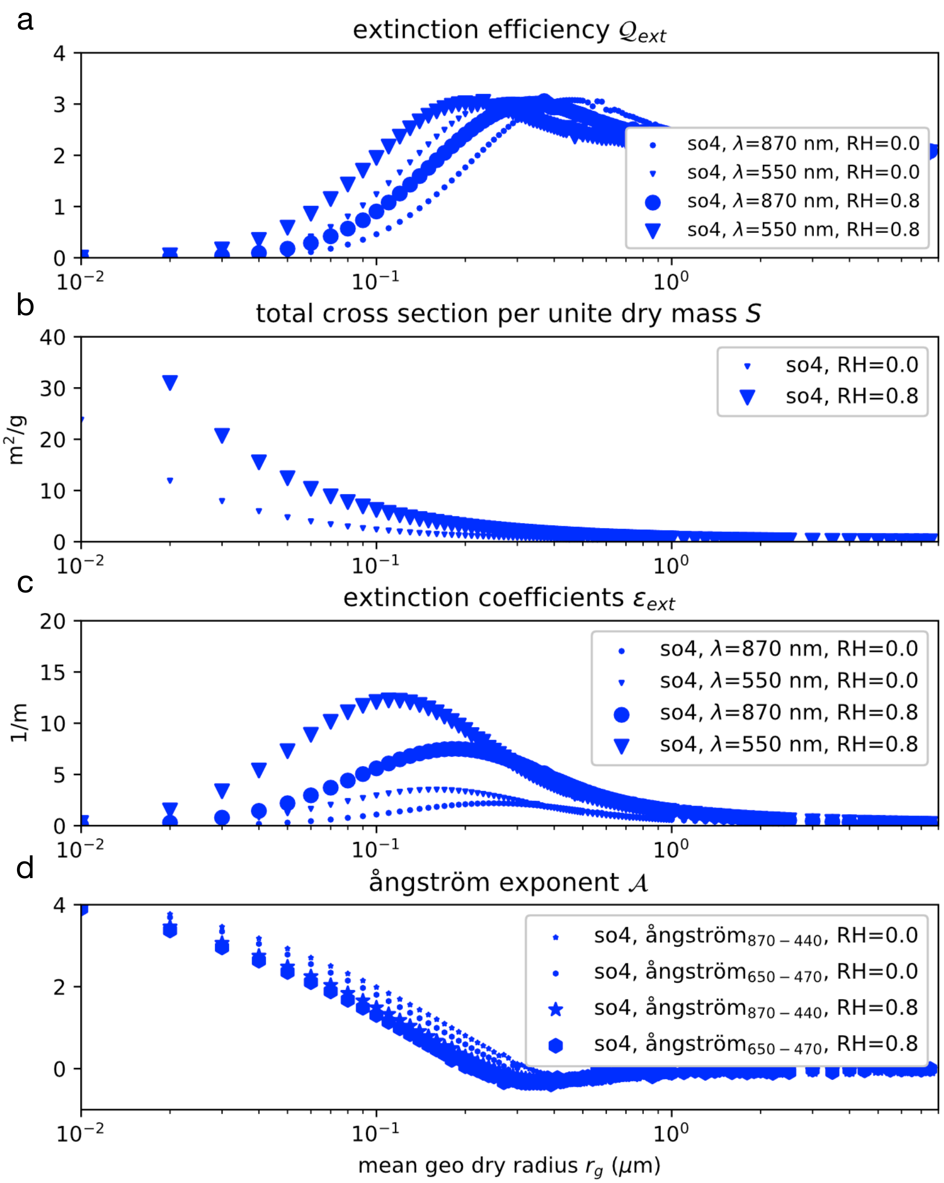

Figure S1. Inorganic aerosol extinction vs. mean geometric dry radius r_g. (a): extinction efficiency $\mathcal{Q}_{ext}$; (b): total cross section per unite dry mass $\mathcal{S}$; (c): extinction coefficients ϵ_{ext}; (d): Ångström exponent $\mathcal{A}$.

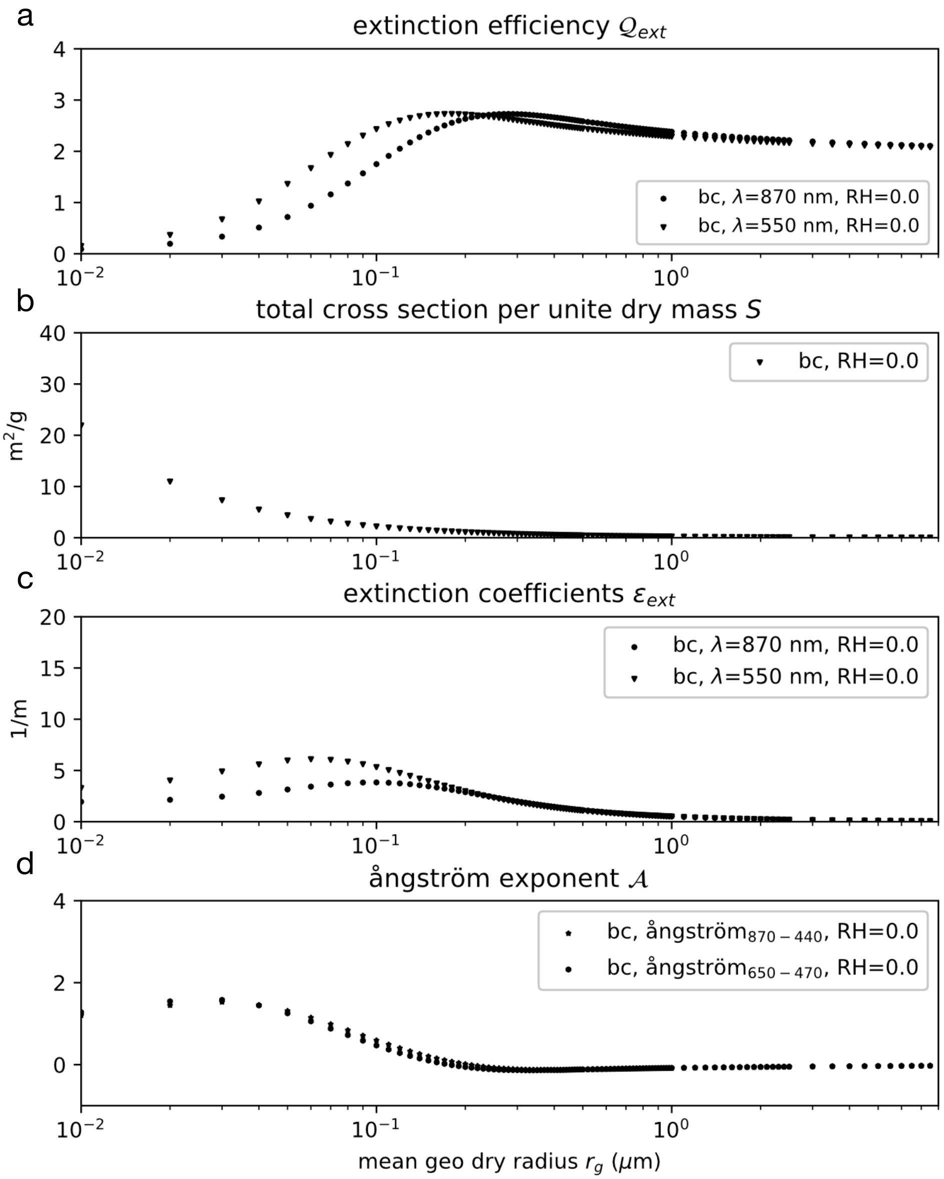

Figure S2. Black carbon extinction vs. mean geometric dry radius r_g. (a): extinction efficiency $\mathcal{Q}_{ext}$; (b): total cross section per unite dry mass $\mathcal{S}$; (c): extinction coefficients ϵ_{ext}; (d): Ångström exponent $\mathcal{A}$.

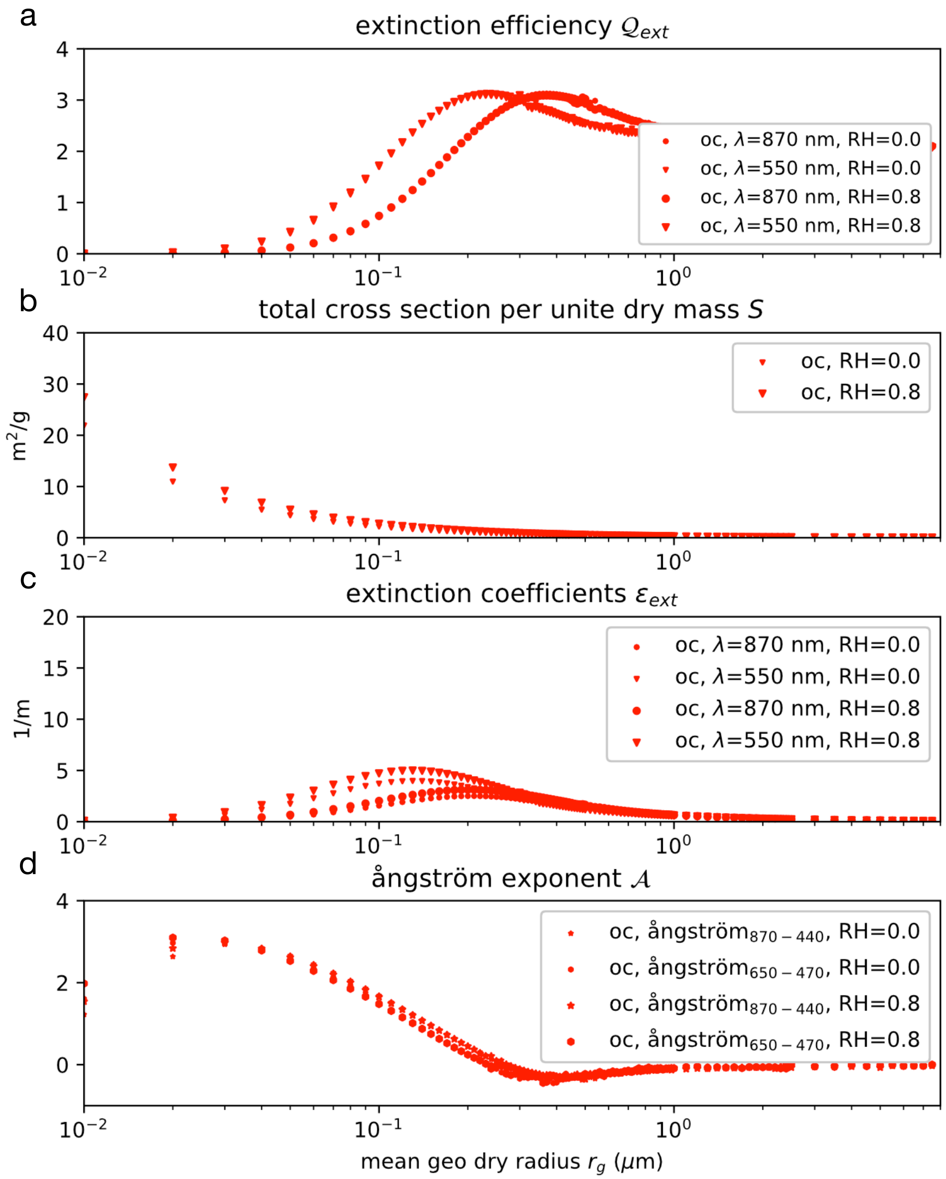

Figure S3. Organic carbon (oc) extinction vs. mean geometric dry radius r_g. (a): extinction efficiency $\mathcal{Q}_{ext}$; (b): total cross section per unite dry mass $\mathcal{S}$; (c): extinction coefficients ϵ_{ext}; (d): Ångström exponent $\mathcal{A}$.

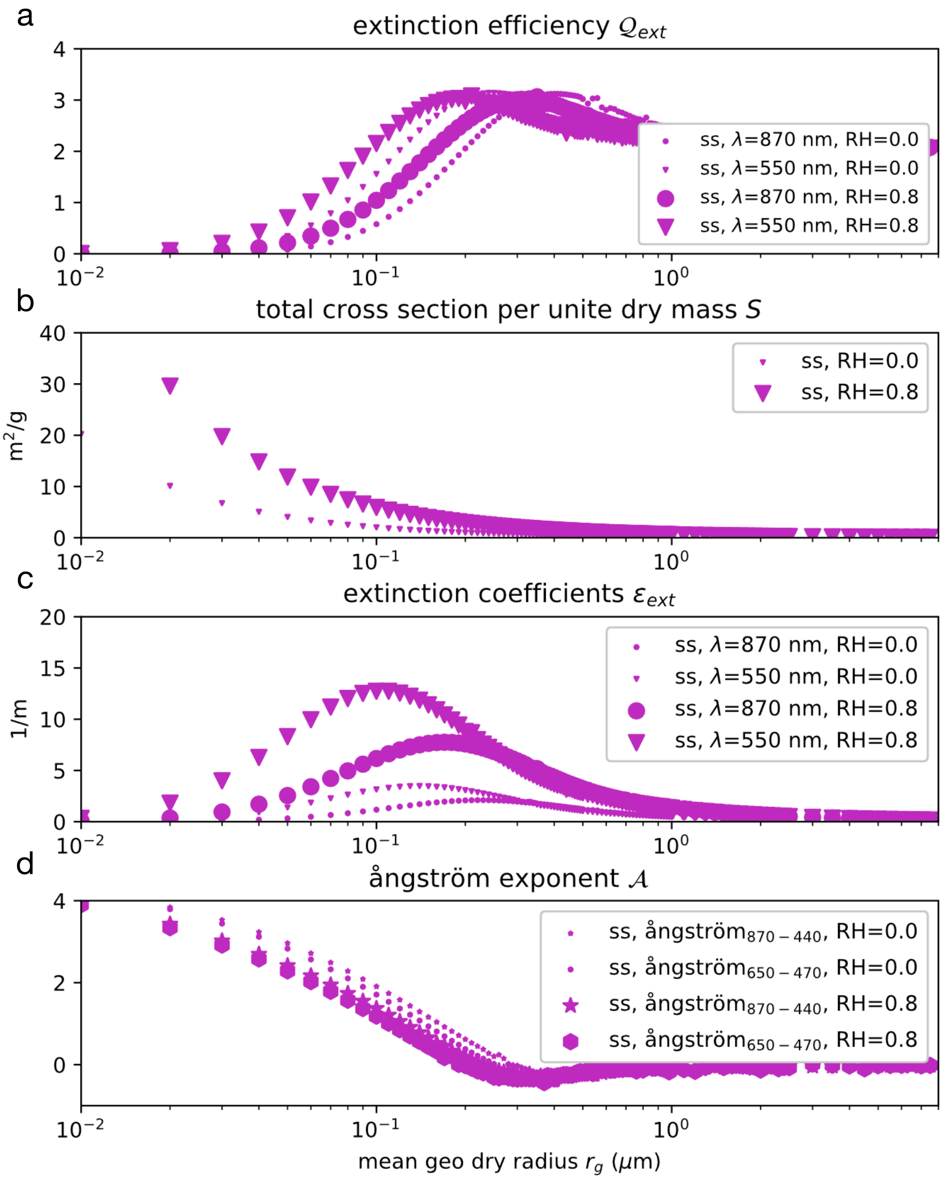

Figure S4. Sea salt extinction vs. mean geometric dry radius r_g. (a): extinction efficiency $\mathcal{Q}_{ext}$; (b): total cross section per unite dry mass $\mathcal{S}$; (c): extinction coefficients ϵ_{ext}; (d): Ångström exponent $\mathcal{A}$.

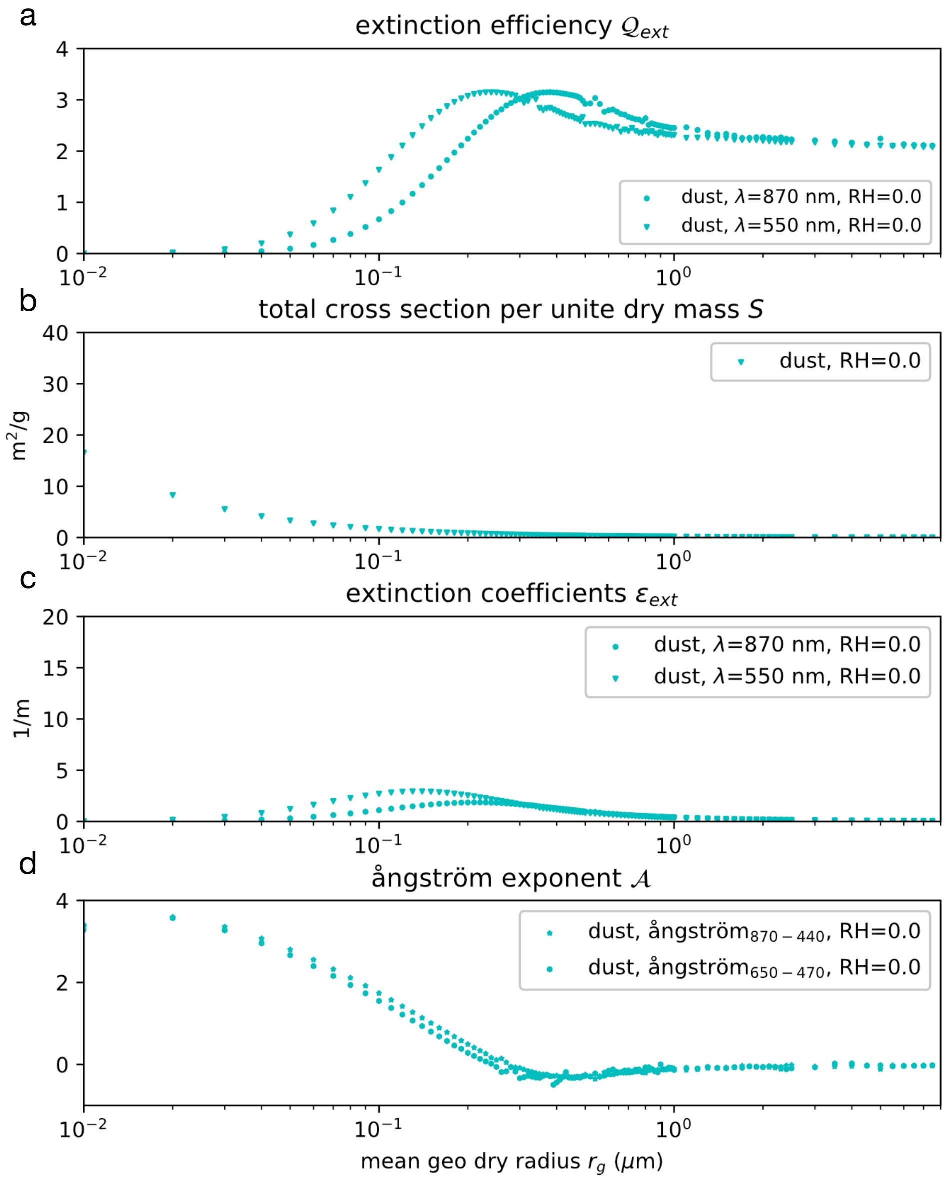

Figure S5. Dust extinction vs. mean geometric dry radius r_g. (a): extinction efficiency $\mathcal{Q}_{ext}$; (b): total cross section per unite dry mass $\mathcal{S}$; (c): extinction coefficients ϵ_{ext}; (d): Ångström exponent $\mathcal{A}$.

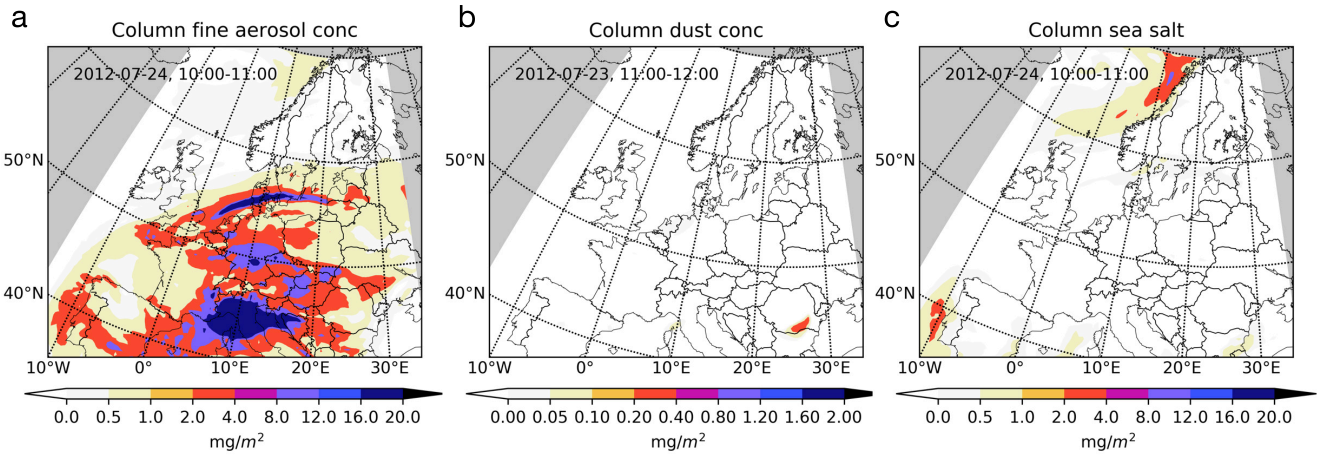

Figure S6. Snapshots of the LOTOS-EUROS simulated column concentration of the fine aerosol (a), dust (b) and sea salt (c) at 2012 July 24, 10:00 to 11:00.

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:

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

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

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

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.