Anonymous Referee #1

General comments:

This paper aims to estimate PM10 using empirical models with remote-sensed aerosol data in Seoul. I think that this paper is generally well written and the conclusion is acceptable and consistent with the previous studies. However, it seems to miss out some of important papers that has already dealt with the issue in question through different approaches, but supported their conclusions. Also, equations are somewhat needed to clarify.

We reflected all the comments by the reviewer. The comments and suggestions by the reviewer were appropriate and improved the scientific quality of our manuscript. We sincerely appreciate such efforts.

Specific comments:

1. P21713, L8: Add Choi et al. (2009), which is a study relevant to the issue in question for East Asia including Seoul, Korea. The study estimated PM10 using MODIS AOD via M1 type model with coefficients obtained from GEOS-Chem, and documented that poor PM10 estimation in spring can be attributed to dust aerosols. Table 1 may include Choi et al. (2009).

Ans.) As suggested by the reviewer, we added a reference "Choi et al., 2009" in P21713, L8 and table 1. Following reference was also added in the reference list:

Choi, Y. S., Park, R. J., and Ho, C. H.: Estimates of ground-level aerosol mass concentrations using a chemical transport model with Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol observations over East Asia, J. Geophys. Res.-Atmos., 114, D04204, doi:10.1029/2008JD011041, 2009.

2. P21713, L24: Add Song et al. (2009), which reported PM10-MODIS AOD relationship over China.

Ans.) As suggested by the reviewer, we added a reference "Song et al., 2009" in P21713, L24. Following reference was also added in the reference list:

Song, C.-K., Ho, C.-H., Park, R. J., Choi, Y.-S., Kim, J., Gong, D.-Y., and Lee, Y.-B.: Spatial and seasonal variations of surface PM10 concentration and MODIS aerosol optical depth

over China, Asia. Pac. J. Atmos. Sci., 45, 33-43, 2009.

3. P21717: Methodology section needs to add further discussion about PM2.5. Perhaps the upper limit of integration in equation (2) should be replaced by 1.25, and equation (3) has to be replaced by FOD?

Ans.) We agree with reviewer's suggestions for need of further discussions about PM2.5. At first, we had a research plan for both PM2.5 and PM10 estimation using various empirical models with AERONET and MODIS dataset. However, we couldn't perform and evaluate PM2.5 estimation due to a limited number of PM2.5 measurements; PM2.5 data could be obtained at only one site in study area. Thus, we focus on the PM10 estimation and leave discussions about PM2.5 out to clarify the purpose of this study and avoid confusions.

The reviewer is right that the upper limit of integration in Eq. (2) should be changed to 1.25 for the expressions of PM2.5. Also, fine mode AOD (FOD) is better suited for PM2.5 estimation theoretically. However, fine mode fraction retrievals over land from MODIS have low accuracy due to the uncertainties in the spectral variation of land surface reflectance (Levy et al., 2007; Lee et al., 2013), which might lead to poor results for application of FMF in PM2.5 estimation. Finding relationship between AOD (and/or FOD) in PM2.5 estimation needs to be further investigated by using the sufficient number of measurements. We added a following sentence to reflect this point in P21718, L19 as:

"In order to extend this analysis to the PM2.5, upper size limit in integral in Eq.(2) need to be corrected and fine mode fraction (FMF) to be additionally considered in Eq.(3). However, since available PM2.5 measurements were quite limited in this area and time, we focused only PM10 in this present study."

Levy, R. C., Remer, L. A., Mattoo, S., Vermote, E. F., & Kaufman, Y. J. (2007). Second-generation operational algorithm: Retrieval of aerosol properties over land from inversion of Moderate Resolution Imaging Spectroradiometer spectral reflectance. Journal of Geophysical Research: Atmospheres (1984–2012), 112(D13).

Lee, K. and Chung, C. E.: Observationally-constrained estimates of global fine-mode AOD, Atmos. Chem. Phys., 13, 2907–2921, doi:10.5194/acp-13-2907-2013, 2013.

4. P21718, L2: In equation (1), both the extinction efficiency Q_ext and the size distribution n is also function of RH (Seinfeld and Pandis, 2006). This leads to confuse the concept of f(RH) independent of <Q_ext> in equation (3). In equations (1) and (2), you may replace r (radius) by D (diameter) for better presentation. Is H is the same as BLH?

Ans.) The reviewer's comments are correct. The confusion of f(RH) concept might be caused by a simplified equation (1), and unclear expressions about the extinction efficiency and the size distribution which is also a function of RH. Thus, the Eq.(1) and (2) have been replaced by the following as cited in Koelemeijer et al. (2006) in the revised manuscript as:

“
$$
\begin{aligned}
AOD &= \pi \int_0^H \int_0^\infty Q_{ext,amb}(m,r,\lambda) n_{amb}(r,z) r^2 dr dz \\
&= \pi f(RH) \int_0^H \int_0^\infty Q_{ext}(m,r,\lambda) n(r,z) r^2 dr dz
\end{aligned}
\quad (1)
$$

where $Q_{ext,amb}(m,r,\lambda)$ is the unitless extinction efficiency influenced by the refractive index (m), particle radius (r), and wavelength (λ) under ambient conditions, $Q_{ext}(m,r,\lambda)$ is the extinction efficiency under dry conditions, $n_{amb}(r, z)$ the size distribution under ambient conditions representing the number of aerosols at corresponding height (z) with a radius (r), $n(r, z)$ the size distribution under dry conditions and H the top height for the integration.

The PM10 concentration, which is the mass concentration of surface-level aerosols with diameters less than10 μm in dry conditions, is given by:

$$
PM_{10} = \frac{4}{3} \pi \rho \int_0^5 r^3 n(r) dr \quad (2)
$$

where ρ is the particle mass density and r is the dry aerosol radius.”

Although Eq. (1) and (2) can be expressed by diameter (D), we still use radius (r) to clarify the derivation of R_{eff}. As mentioned in P21718, L10-12 (“With the assumption of a homogeneous aerosol distribution within the BLH, the integration from the surface up to the TOA (H) can be simplified by multiplying by the BLH.”), H can be substituted by BLH with the assumption of a homogeneous aerosol distribution within the BLH.

5. P21719, L24: Can f(RH) from Beijing during the spring be applied to Seoul where chemical compositions are very different?

Ans.) Although aerosol chemical composition is somewhat different between Seoul and Beijing, air quality in Seoul and Beijing are predominantly affected by anthropogenic pollutants. Furthermore, Seoul is usually under the influence of prevailing westerlies due to its location in the downwind region of Northeast China. Among aerosol hygroscopic growth functions that can be used, f(RH) obtained from the site near Beijing is appropriate to apply to this study with respect to both time and location. Also, according to the Table 3 of Pan et al. (2009), the values of f(RH=80%) for the pollution aerosols from Korean sectors during the spring season which is derived from Kim et al. (2006) shows the range of 1.55 – 1.77. This range of f(RH) value is similar to that of Pan et al. (2009) for urban pollution aerosol types (1.55 – 1.59).

The sentence in P21729, L24-26 has been revised as:

"In this study, f(RH) based on experimental data obtained near the Beijing mega-city during the spring was employed (Pan et al., 2009), which is appropriate to this study with respect to both temporal and spatial conditions."

6. P21720, L3: The accuracy of AERONET measured Re can hardly be guaranteed. What is the known accuracy?

Ans.) As suggested by the reviewer, we inserted the following sentences in P21720, L5 in the revised manuscript as:

"This R_{eff} is one of the main features derived by the particle volume size distribution retrieved by AERONET inversion algorithm, which was demonstrated to be adequate in practically all situations, especially, for the intermediate particle size range (0.1 $\leq$r $\leq$7 μm) with 10-35% of retrieval errors, as reported by Dubovik et al. (2002)."

Following reference was added in the revised manuscript:

Dubovik, O., Holben, B. N., Eck, T. F., Smirnov, A., Kaufman, Y. J., King, M. D., Tanr´e, D., and Slutsker, I.: Variability of absorption and optical properties of key aerosol types observed in worldwide locations, J. Atmos. Sci., 59, 590–608, 2002.

7. P21720, L19: What does AE stand for? L22-25: Hard to understand.

Ans.) AE (Angstrom exponent) which is above mentioned in P21725, L7-10 is used as a parameter to indicate the aerosol size in the MLR model, M6. Several previous studies used the AE to classify aerosols into fine and coarse mode aerosols as a qualitative indicator of aerosol particle size. To clarify the role of AE as a parameter in the M6, sentences in P21720, L22-25 have been revised as:

"The R_{eff} inversion product is from diffuse sky radiance measurement which has strict stability criteria. Thus, the number of data (N=713) is quite lower than products from direct sun measurements including AE (N=2112) which also implies the aerosol size information. For that reason, AE was used as a variable in the MLR model instead of R_{eff} to secure enough number of data sample (Dubovik et al., 2000; Schuster et al., 2006)."

Following references were added in the revised manuscript:

Dubovik, O., Smirnov, A., Holben, B., King, M. D., Kaufman, Y. J., Eck, T. F., and Slutsker, I. Accuracy assessments of aerosol optical properties retrieved from Aerosol Robotic Network (AERONET) Sun and sky radiance measurements, J. Geophys. Res.-Atmos., 105, 9791–9806, 2000.

Schuster, G. L., Dubovik, O., and Holben, B. N.: Angstrom exponent and bimodal aerosol size distributions, J. Geophys. Res.,111, D07207, doi:10.1029/2005JD006328, 2006.

8. P21720, L9: There is no explanation how equation (4) is related with equation (3).

Ans.) As previously mentioned in P21719, L12-14 ("Models M1 to M5 are empirical models based on the relationship between AOD and PM concentration, as described in Sect. 3.1, whereas M6 represents a multiple linear regression model."), equation (3) is a simplified equation based on the physical relationship between AOD and PM10 concentration with some assumptions (homogeneous aerosol vertical distribution within the BLH, effects of aerosol hygroscopic factor for RH variations), on the other hand, equation (4) is a statistical model of multiple linear regression. Thus, there is no direct relationship between the two equations. To clarify this, following sentence in P21720, L6-8 has been revised as:

"In addition to the simple empirical models (M1-M5) derived from the relationship between AOD and PM (Eq. (3)), a multiple linear regression (MLR) model was used as a statistical approach to determine PM10 concentrations as a function of eight different parameters associated with PM estimation:"

9. Table 1 mentioned the methods, M1 to M4, which will be introduced later in Table 3. What makes confusion is that, however, M2 in Table 1 uses RH, while in Table 3 M2 does not use RH. Which one is correct?

Ans.) Table 1 is to summarize different methods from previous studies, but Table 3 lists the methods for this study. Methods from M1 to M4 in Table 1 indicate the classification of various methods for estimating PM concentrations using AOD applied in previous studies. On the other hand, the models from M1 to M6 in Table 3 describe the developed empirical linear models used in this study. To avoid confusions, we changed the notation in Table 1 from M1, M2, M3 and M4 to MT1, MT2, MT3 and MT4, respectively.

10. There is a recent study by Escribano et al. (2014), which pointed out BLH is critical in the AOD-PM relation. Also they found that the misfit in surface reflectance in MODIS algorithm especially in semi-arid region may lead to a spurious MODIS AOD, leading uncoupled relation between PM and AOD. Please at least discuss their findings too in *relation with your study.*

Ans.) Following sentences were inserted in P21727, L24 in the revised manuscript:

"In order to understand smaller scale features of the air quality, higher spatial resolution AOD products such as a MODIS 3km product are under development. Although this high resolution product has been expected to explain aerosol gradients in detail at a small scale, the 3km product showed poor performances compared to the 10 km product due to improper characterization of the urban surfaces (Levy et al., 2013; Munchak et al., 2013). This bias in surface reflectance of MODIS algorithm indeed resulted in misfit between column AOD and surface PM concentration, as discussed in Escribano et al. (2014). Thus, estimated spatial characteristics of surface PM concentrations are reliable when aerosol products are satisfied with both higher quality and finer resolution."

Following reference was added in the revised manuscript:

Escribano, J., Gallardo, L., Rondanelli, R., and Choi, Y.-S.: Satellite retrievals of aerosol optical depth over a subtropical urban area: the role of stratification and surface reflectance, Aerosol. Air. Qual. Res., 14, 596-U568, 2014.

Anonymous Referee #2

General comments:

This paper evaluates the performance of empirical models for studying spatio-temporal variability of PM10 concentration over Seoul using AERONET and MODIS AOD, and other ancillary data including boundary layer height (BLH), relative humidity (RH), and effective radius (Reff) of the aerosol size distribution during the DRAGON-Asia campaign in 2012. The methods/results are well documented and summarized. The topical area is also suitable for the special issue ("Meso-scale aerosol processes, comparison and validation studies from DRAGON networks") in ACP journal. I favor publication of this paper in ACP with some clarifications and minor changes.

We reflected all the comments by the reviewer. The comments and suggestions by the reviewer were appropriate and improved the scientific quality of our manuscript. We sincerely appreciate such efforts.

Specific comments:

1. The title appears to be a bit descriptive and awkward. Reword it to be concise.

Ans.) As suggested by the reviewer, we changed the title as "Estimation of PM10 concentrations over Seoul using multiple empirical models with AERONET and MODIS data collected during the DRAGON-Asia campaign".

2. Making scatter plots of measured PM10 against major parameters (e.g., AOD, BLH, RH, Reff, AE, and their combinations) may give some insights of the relationship between them. And linear fits can be added as well.

Ans.) The following sentences and figures have been inserted on pages 21719, lines 4 in the revised manuscript as:

"To gain insights of the relationship between PM10 and major predictors, all PM10 concentration was plotted against AOD, BLH, RH, and R_{eff}, which were used in this study for development and validation of PM10 estimation as shown in Fig 4. The correlation coefficient (R) between PM10 and AOD was 0.5 and that of R_{eff} was 0.32. As expected, BLH showed negative correlation with PM10 (-0.36). However, RH did not show any significant relationship with PM10."

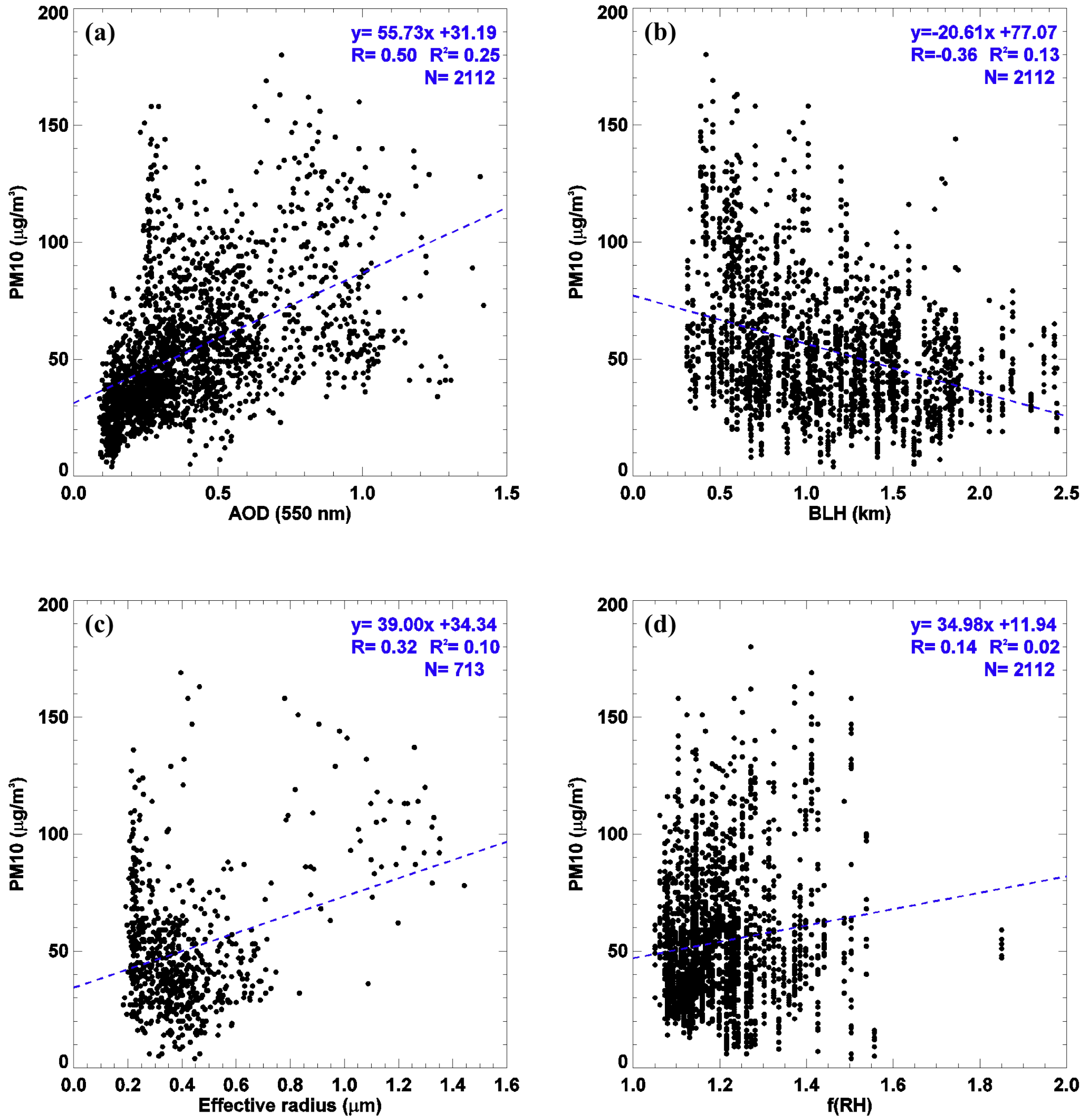

Figure 4. Scatter plots of the various parameters including (a) AOD, (b) BLH, (c) effective radius, and (d) RH against the dependent variable of PM10 concentration. The regression line is shown as a blue dashed line.

3. In Table 5, results were well summarized by various statistical measures, however, I wonder if they are statistically significant among models. Are their performances significantly different among models (M1~M6)?

Ans.) As the reviewer pointed out, there might be no statistically significant differences between the performances of models in some cases. However, the main purpose of this study is to evaluate the performances of various empirical models by incorporating different parameters. For this reason, we tested various models and provided correlation coefficient, RMSE etc., which makes possible to determine important factors in PM10 estimation.

4. R2 (coefficient of determination) is a more appropriate quantity than R (correlation coefficient) in explaining the performance of empirical linear models throughout this paper. Refer to a statistics textbook.

Ans.) As suggested by the reviewer, we added the R^2 (coefficient of determination) values in Table 5, 6 and 7.

5. What empirical models are best for estimating PM10? And Why? These things need to be clearly discussed and stated in abstract and conclusion. I expect the best performances from M3 and M5 because they look close to the form of equation 3. If not, explain why. Even the best performance of M5 during the winter season (R=0.81, R2 = 0.66) in Table 7 shows remaining 34% variance is not explained by the model. What other factors should be taken into account for future improvement?

Ans.) As suggested by the reviewer, we inserted some sentences in the revised manuscript to emphasize the empirical model which derives the best result in PM10 estimation.

The following sentence has been revised in abstract [P21711, L11-13] in the revised manuscript as:

"Among various empirical models, the model which incorporates both BLH and R_{eff} showed the highest correlation, which indicates the strong influence of BLH and R_{eff} on the PM10 estimations. Meanwhile, the effect of RH on the relationship between AOD and PM10 was appeared to be negligible during the campaign period (spring) when RH is generally low in Northeast Asia."

The following sentences have been inserted in conclusion [P21729, L21] in the revised manuscript as:

"The improved performances were found when the vertical correction on AOD using the BLH was applied in both AERONET and MODIS datasets (M2) compared to the simplest model (M1). These empirical model performances were further enhanced by additionally including the effective radius for size correction (M3, M5). However, not meaningful improvements were found when RH was considered additionally (M4). Among different empirical models based on the physical relationship between AOD and PM concentration (M1-M5), model M5 which follows the nearest form of that relationship with the largest number of parameters showed the best performance."

We discussed about other factors which should be considered for future improvement in conclusion in the revised manuscript. Please refer to answers to comments 6 of Referee #2.

6. Page 21730, lines 20-24: it does not necessarily support that AOD at a finer spatial resolution from such as GOCI or MODIS would help to improve the predictability of PM10. In general, the accuracy of MODIS AOD from a higher spatial resolution of 3km is not better than that from a standard product of a 10 km resolution, especially in urban areas due primarily to inadequate characterization of surface properties (Refer to the paper, "MODIS 3 km aerosol product: applications over land in an urban/suburban region", L. A. Munchak, R. C. Levy, S. Mattoo, L. A. Remer, B. N. Holben, J. S. Schafer, C. A. Hostetler, and R. A. Ferrare, Atmos. Meas. Tech., 6, 1747-1759, 2013). Moreover, very accurate point measured AOD data from AERONET after additional cloud screenings were already tested in this study. Further in-depth discussion about possible factors and mechanism other than AOD for improving the predictability of PM10 is expected to enhance the quality of this paper.

Ans.) As the reviewer pointed out, AOD products at a finer spatial resolution didn't show better performances over urban areas than those at a lower spatial resolution (Munchak et al., 2013). To mention a limitation in application of the finer resolution AOD data for improving the PM10 estimation and difficulties of surface reflectance characterization, we inserted following sentences in P21727, L24 in the revised manuscript as:

"In order to understand smaller scale features of the air quality, higher spatial resolution AOD products such as a MODIS 3km product are under development. Although this high resolution product has been expected to explain aerosol gradients in detail at a small scale, the 3km product showed poor performances compared to the 10 km product due to improper characterization of the urban surfaces (Levy et al., 2013; Munchak et al., 2013). This bias in surface reflectance of MODIS algorithm indeed resulted in misfit between column AOD and surface PM concentration, as discussed in Escribano et al. (2014). Thus, estimated spatial characteristics of surface PM concentrations are reliable when aerosol products are satisfied with both higher quality and finer resolution."

Also, to suggest other factors needed to be considered for improving the predictability of PM10, sentences in P21730, L20-24 have been revised as:

"For better estimating surface PM concentrations by satellite remote sensing, especially in urban areas where diverse aerosol sources are distributed, aerosol products with a higher quality and a finer resolution are required. Additionally, accurate and detailed information about aerosol vertical distribution, size distribution, and composition will contribute to improve empirical models."