Interactive review of 'Factors that influence surface PM2.5 values inferred from satellite observations: perspective gained for the Baltimore-Washington Area during DISCOVER-AQ' by S. Crumeyrolle

We thank referee n°1 for the constructive comments on our manuscript, which are summarized with our italicized responses below.

__Science questions__

P. 23423 line 7 "Yet, the complexity and resolution of aerosol satellite retrievals..." P. 23423 line 25 "However, the uncertainty in relating a column integrated AOD to ground- level PM2.5 is compounded by timing mismatches bet ween the measurements."

→what sort of horizontal resolution would be required by satellite measurements to do more than have a broadbrush estimate of air quality in a region?

→it seems like spatial mismatches will also compound uncertainty, but your study seems ideally designed to address spatial mismatches due to the relatively close profiles within close proximity. Can you provide any sort of quantification of the uncertainty induced by spatial mismatches by comparing results from one profile site with those from another profile site?

The horizontal resolution of satellite required to get an accurate estimation of air quality (AQ) depends on the region you are interested in (i.e. urban or rural) and also on the local dynamics involved in the area. Indeed, the denser the city the larger the resolution should be. Over the Washington-Baltimore area, the presence of the bay leads to some peculiar local dynamics, i.e. the bay breeze described by Stauffer et al. (2012). This bay breeze modifies drastically the aerosol concentration profiles in the lower layer. For example, Edgewood and Essex are the DISCOVER-AQ sites the most affected by the bay breeze. Moreover, the error due to spatial mismatches is also dependent on the parameter you are interested in. Thus, a quantification of the spatial mismatches using these data would be … (I meant that giving one number would be a mistake. The spatial mismatches are important but even at the scale of one profile you can see some heterogeneities)

Black carbon (BC)/absorbing aerosol is an air pollutant of interest to both the air quality and climate communities. Are there any plans to use the large amount of data from this data set to evaluate AERONET retrievals of absorption optical depth? This would be HUGELY interesting to many people. See the Bond et al. 2013 so-called "bounding BC" paper published in JGR earlier this year.

The referee is right to say that the absorbing aerosols are of great interest. The AERONET retrievals of the absorbing aerosol optical depth (AOD) will definitely be studied and will be part of another manuscript. Indeed, Scarino et al. are already working on this comparison and will most likely publish it within the next year. In this study, we wanted to 1. prove the need of active remote sensing along with AOD measurements (grounded or from space) to estimate accurately particulate matter from column integrated measurements; 2. estimate the errors due to a lack of information on the relative humidity throughout the column.

The flights described here all took place in the summer time. Is more reflective surface albedo the only confounding effect to this type of analysis in the winter?

The snow cover in winter may have a strong impact on the AOD measurements from space as shown by Van Donkelaar et al. (2006) and is one of the major confounding effects to this type of study. Furthermore, during the winter, the environmental conditions characterised by low temperatures and weak solar radiation lead to a less-turbulent and thus less-developed boundary layer. The boundary layer height is usually closer to the surface (<1000m) during the winter while it's usually observed over 1000m during the summer. During this field campaign, the measurements show strong variability of the BL height from 300m and lower to 2200m depending on the site and of the time of the day. The lowest values of the boundary layer height were observed early in the morning and are surprisingly as low as the one expected during the winter (<400m). The corresponding profiles were not included in this analysis as the DC8 measurements within the BL were limited to few seconds. During the winter most of the profiles would be like those and thus might decrease the accuracy of the PM2.5 retrievals from the column-integrated measurements.

P23427 lines 15-25 Description of neph and PSAP on P3 – how was the low RH maintained for the PSAP? Was it also downstream of the perma pure dryer? Also – the PSAP is notoriously sensitive to both pressure changes and RH variability (even at low RH) – how was that dealt with in the dataset? Or was the absorption measurement a small enough fraction of the extinction that even with PSAP noise that wasn't a big deal?

The PSAP RH was not actively controlled, but filters were heated to 40C to reduce variability. Pressure fluctuations do induce noise, and extreme outliers are removed based on visual inspection. Overall, the absorption coefficient represents less than 5% of the total extinction. So the absorption coefficient fluctuations play a minimal major role in the extinction variability.

P23435, line 8-10 "According to the Mie theory, small particles (D_p less than 100 nm) are significantly less optically active than larger particles, but still impact the total aerosol mass" Perhaps I'm thinking about this wrong, but doesn't scattering vary as diameter-squared while mass varies as diameter-cubed? So wouldn't these smaller particles have more optical effect than mass effect? Also, typically Mie theory does not have 'the' in front of it so →"According to Mie theory..."

The extinction curves calculated from the theory of Mie for two different refractive indexes (m=1.5 and m=1.33) are presented in the figure 1 (Van de Hulst, 1957). These curves represent the scattering efficiency as a function of the dimensionless particle parameter (x). In our case the wavelengths used are in the visible range from 450 to 700nm and we are looking for the scattering efficiency of particles with a diameter smaller than 100nm. Given these variables, the 'x' parameter is thus varying between 0.45 and 0.70. In this 'x' range, the particle scattering efficiency is close zero. This is why we stated that those particles 'are significantly less optically active than larger particles'. Usually the concentration of the smaller particles is large and leads to an increase on the total aerosol mass.

The typo has been corrected in the manuscript.

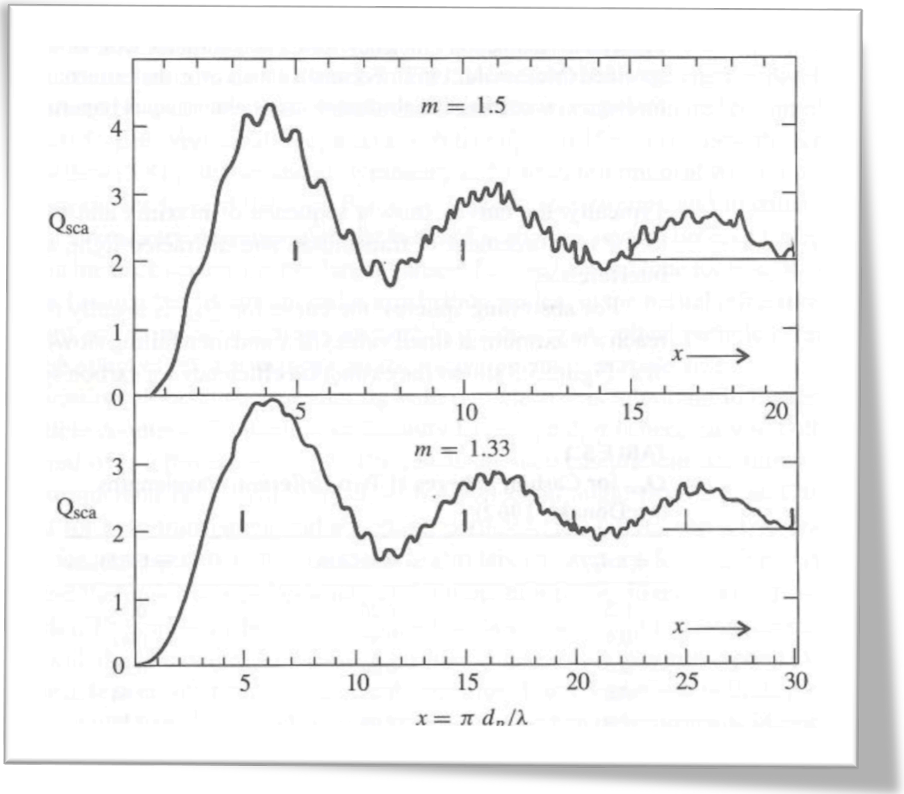


Figure 1 : Extinction curves calculated from the Mie theory for m=1.5 and m=1.33 (Van de Hulst, 1957).

You mention Ångström exponent, but how does the in-situ and AERONET Ångström exponent change for times when the smaller particles are present? Is it sensitive enough to be a useful measurement if these particles aren't very optically active? This would be a useful additional plot to show as it's a more readily available parameter than sub0.1um particle size.

The scattering Angström exponent is related to the size distribution of the aerosol particles sampled in the atmosphere, but is somewhat qualitative as other factors confound a direct size-AE parameterization (e.g., chemical composition, aerosol mixing state). Most importantly, interpretation of the scattering Ångström exponent when the particle size distribution is dominated by sub-100nm particles is not valid as illustrated in Figure 2.

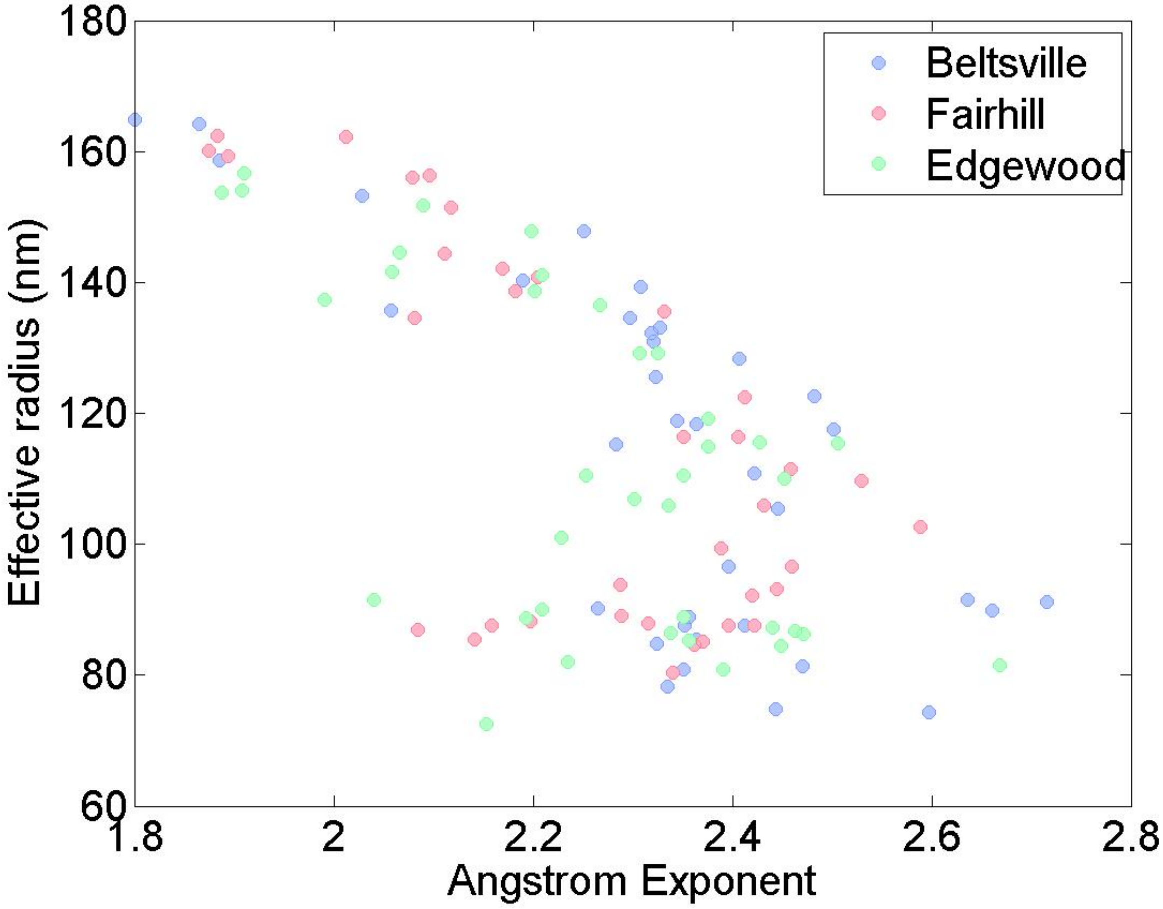


Figure 2 : Effective radius (nm) as a function of the Ångström exponent (450-700nm) measured aboard the P3-B over Beltsville, Fairhill and Edgewood during the entire campaign.

P23436, lines 11-12 "while the presence of an elevated layer is leading to a spread of the data set (0.71) and a slope 1.6 times lower…" →while the presence of an elevated layer leads to a wider spread of the data set (R2~0.71) and a lower slope (46.3).

This has been corrected in the manuscript.

→ does this increased slope for the BL dominated profiles suggest that the Hoff and Christopher and Engel-Cox citations were affected by the pitfall of elevated layers? 46.3 is right in the middle of the slope range reported by Engel-Cox.

Hoff and Christopher (2009) and Engel-Cox et al. (2006) do not describe the aerosol vertical distribution thus it is not possible to directly attribute differences to the presence of elevated layers. Nonetheless, the differences with the results from Hoff and Christopher (2009) could be due to observations performed all over the United States and not only over one specific region. Indeed, it might be due to the presence of different predominant type of aerosol (sulphate in Texas, nitrate in California…), the surface type (urban area, desert…), and different dynamics at a regional scale, which may lead to the presence or the absence of an elevated layer. Indeed, the slopes over the United States as reported in Hoff and Christopher (2009) highlight a large variability and vary from 22.6 to 77.

The comparison with Engel-Cox results is more straightforward because the measurements were performed in the same area (Baltimore) during the same season (summer). Nonetheless, the PM2.5 and the AOD were not measured with the same instruments, respectively measured with a TEOM (Tapered Element Oscillating Microbalance) and MODIS (Moderate Resolution Imaging Spectrometer). Thus, some small divergences are expected due to instrumental biases. Overall, the slopes are similar from 38.7 to 48.5 in this study compare to 31 and 49 over Baltimore during the summer 2004. In our study, Beltsville show a larger slope (66.1) but the profiles over Beltsville were performed from 400m to 1.5km (instead of 3 km for the other sites) and thus the presence of an elevated layer cannot be excluded.

Discussion related to figure 8: it seems like other measurements such as effective size and chemistry (sulfate/OC) which you also have for all these profiles would be better indicators of the similarities between BL and BuL.

The authors plot as requested the effective radius and the Sulfate to Water Soluble Organic compound ratio averaged within the BL as a function of those averaged within the BuL (Figure 3 herein). The results show a strong correlation of the aerosol chemical composition and the effective radius averaged within the BL and the BuL, similar to the one observed with the f(RH) and the Ångström exponent. Unfortunately, the temporal resolution of the chemical measurement performed by the PILS varied from 3 to 5 minutes. Thus, in many cases the PILS samples overlapped between BL and BuL. Following the referee's suggestion, the Ångström exponent has been replaced by the effective radius in the Figure 8.

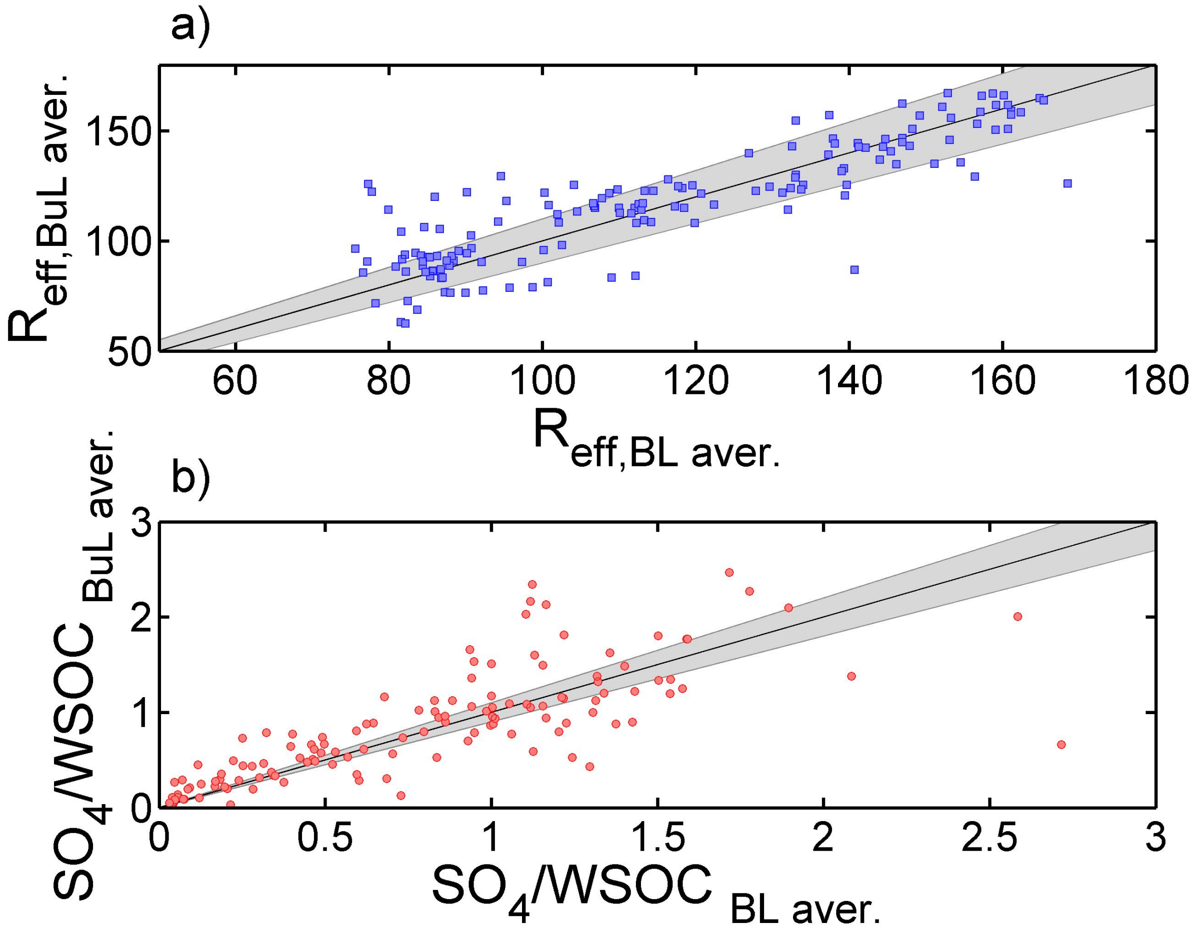


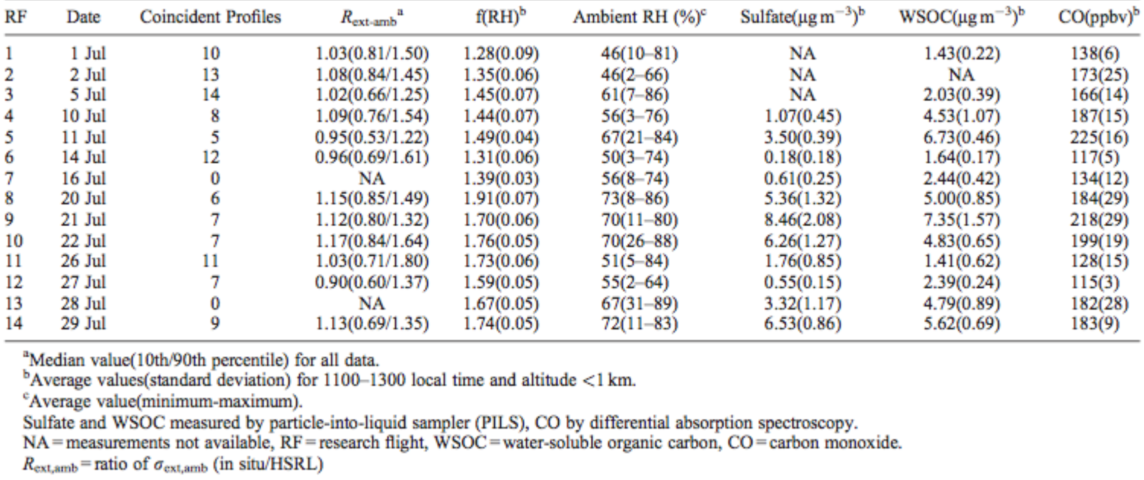
Figure 3 : Comparison of the effective radius (a), and the sulphate to water soluble organic carbon (b)

averaged within the BL (Boundary Layer) and the BuL (Buffer Layer) at the different DISCOVER-AQ sites (Beltsville, Padonia, Fairhill, Aldino, Edgewood and Essex). The black line corresponds to the 1:1 line and the gray area represents the 10% variability.

P23439, line 3-4 "During this campaign, the f (RH)amb values were observed to vary significantly from 1.03 to 2.3 on a day-to-day basis, but the profiles were fairly constant within the BL." How did the f(RH=80%) and/or gamma values vary over the course of the campaign? This would give a good indicator of changes in aerosol chemistry with time.

The Figure 4 in the paper shows the mass extinction efficiency (MEE) and the sulfate-to-WSOC (water soluble organic carbon) as a function of the Julian day. As sulphate and WSOC were the two major compounds of the aerosol chemical composition, the ratio of both compounds give an indicator of the modification of the aerosol chemical composition on a daily basis. In the manuscript, the authors add a quote to Ziemba et al. (2013) that published the f(RH) values for each flight listed in the Table 1. F(RH) was found to vary with the ratio of organics-to-sulfate with the highest f(RH) measured on 20 July when then Sulfate-to-WSOC ratio was greater than 1:1.

Table 1: Summary of the DISCOVER-AQ flight data as shown in *Ziemba et al. (2013)*



RF	Date	Coincident Profiles	$R_{ext\text{-}amb}$[a]	f(RH)[b]	Ambient RH (%)[c]	Sulfate($\mu g\,m^{-3}$)[b]	WSOC($\mu g\,m^{-3}$)[b]	CO(ppbv)[b]
1	1 Jul	10	1.03(0.81/1.50)	1.28(0.09)	46(10–81)	NA	1.43(0.22)	138(6)
2	2 Jul	13	1.08(0.84/1.45)	1.35(0.06)	46(2–66)	NA	NA	173(25)
3	5 Jul	14	1.02(0.66/1.25)	1.45(0.07)	61(7–86)	NA	2.03(0.39)	166(14)
4	10 Jul	8	1.09(0.76/1.54)	1.44(0.07)	56(3–76)	1.07(0.45)	4.53(1.07)	187(15)
5	11 Jul	5	0.95(0.53/1.22)	1.49(0.04)	67(21–84)	3.50(0.39)	6.73(0.46)	225(16)
6	14 Jul	12	0.96(0.69/1.61)	1.31(0.06)	50(3–74)	0.18(0.18)	1.64(0.17)	117(5)
7	16 Jul	0	NA	1.39(0.03)	56(8–74)	0.61(0.25)	2.44(0.42)	134(12)
8	20 Jul	6	1.15(0.85/1.49)	1.91(0.07)	73(8–86)	5.36(1.32)	5.00(0.85)	184(29)
9	21 Jul	7	1.12(0.80/1.32)	1.70(0.06)	70(11–80)	8.46(2.08)	7.35(1.57)	218(29)
10	22 Jul	7	1.17(0.84/1.64)	1.76(0.05)	70(26–88)	6.26(1.27)	4.83(0.65)	199(19)
11	26 Jul	11	1.03(0.71/1.80)	1.73(0.06)	51(5–84)	1.76(0.85)	1.41(0.62)	128(15)
12	27 Jul	7	0.90(0.60/1.37)	1.59(0.05)	55(2–64)	0.55(0.15)	2.39(0.24)	115(3)
13	28 Jul	0	NA	1.67(0.05)	67(31–89)	3.32(1.17)	4.79(0.89)	182(28)
14	29 Jul	9	1.13(0.69/1.35)	1.74(0.05)	72(11–83)	6.53(0.86)	5.62(0.69)	183(9)

[a] Median value(10th/90th percentile) for all data.
[b] Average values(standard deviation) for 1100–1300 local time and altitude <1 km.
[c] Average value(minimum-maximum).
Sulfate and WSOC measured by particle-into-liquid sampler (PILS), CO by differential absorption spectroscopy.
NA = measurements not available, RF = research flight, WSOC = water-soluble organic carbon, CO = carbon monoxide.
$R_{ext,amb}$ = ratio of $\sigma_{ext,amb}$ (in situ/HSRL)

In general I found the discussion of relative humidity/f(RH)/water uptake confusing. I think more work needs to be done on this section including some discussion of changes in column RH. water uptake is not going to happen unless there is water vapor available to be taken up, but the focus was on f(RH)amb which is a less readily available parameter than atmospheric RH. I suggest the authors add some information about how know ambient RH or column RH or BL RH (some parameterization of atmospheric RH) would help with PM2.5 retrieval even if f(RH) is not known.

Good point. We have revised the discussion to improve clarity and to emphasize the difference between ambient meteorology (i.e., RH) and composition-dependent particle properties (i.e., f(RH)). Indeed, these two parameters are not theoretically *linked and any correlation between these two parameters is purely coincidental. The figure 10 (in the manuscript) suggests that both parameters are needed to accurately retrieve PM2.5.*

Typos, missing information and semantic quibbles

General comment – I feel that the authors, in general, went for the easy citations – the ones they had at hand – rather than citing the original piece of work. That feels ethically wrong to me. At the very least they could say 'and references therein' to show that the work they've cited is a branch instead of a root. Unfortunately, due to my limited access to bibliographic resources at the moment I'm not able to provide the exact citations that I think should be included.

Done. We have added additional references that highlight first findings to the already sizable list of recent citations, which were originally chosen because they represent the current state of the field.

In terms of reporting values – both the AERONET retrievals and the in-situ measurements are spectral – you need to make clear what wavelength you are using in the text and in the figures. I've made notes for all the figures, but you should also do so in the text, particularly when a wavelength dependent property is used in a calculation (e.g., MEE). Abstract: "...(AOD) calculated with the extinction (532 nm) measured during the in-situ profiles..." 532 or 550 nm? The TSI neph measures at 550 nm and while the PSAP green wavelength is 532 nm the text immediately after (p23427, line 22) says that the PSAP measurements are interpolated to 550 nm. The only other mention of 532 nm in the paper is in reference to a paper by Ziemba et al comparing in-situ measurements to the HSRL retrievals, which are made at 532 nm.

Excellent point! We have added wavelengths to all of the measurement descriptions in the manuscript. In this study most of the calculations have been performed at 550nm except for the comparison with AERONET data that have been made using measurements performed at three wavelengths (440, 500 and 675 nm).

"This motivates the use of active remote sensing techniques to dramatically improve air quality retrievals." I'm not sure this sentence belongs in the abstract as it suggests/implies that active remote sensing techniques were used in the study described in the manuscript. Perhaps it could be rephrased: "This suggests that the use of active remote sensing techniques would dramatically improve AQ retrievals." This point should more strongly be made in the text and conclusions if it is worthy of a place in the abstract.

Good point. We have revised the abstract per the suggestions of Reviewers 1 and 2. The presence of an aerosol layer above the boundary layer has been shown to decrease the accuracy of the $PM_{2.5}$ *retrievals by a factor of 1.6. Moreover, knowing the height of the BuL is also shown to improve the* $PM_{2.5}$ *retrievals. The aerosol vertical distribution as well as the BuL height can be measured with active remote sensing techniques. This point has been reinforced throughout the paper and especially in the conclusion.*

"...the $f(RH)_{amb}$ (obtained from two nephelometers at different relative humidities – RHs)..." fRHamb needs to be defined for the uninitiated. Alternatively, you could say: "...the $f(RH)_{amb}$ (ratio of scattering at ambient relative humidity (RH) to scattering at low RH) and leave the description of how you arrived at it for section 3.

Done.

P23424 line 24. ***"This changes the ambient aerosol mass…"*** **Change to: "Water uptake change the ambient aerosol mass…"**

Done.

P23427, line 25 "*The standard corrections…*" it's not the corrections per se but the issues with filter-based measurements of absorption and the PSAP instrument in general.

Good point. This sentence has been reworded.
"Basic principles of filter-based measurements, like the PSAP, limit the accuracy of the observed absorption coefficient to 20 to 30% (Ryder et al., 2013)."

P23424, line 26-28 "Currently, aerosol liquid water content is not measured at the ground sites of the global atmosphere watch(GAW) network and nor at a global scale…" You need to be careful not to paint with such a broad brush – aerosol hygroscopicity either in terms of scattering as a function of RH (i.e., f(RH)) or particle size as a function of RH (sometime called g(RH) and also sometimes called f(RH)) is measured at some GAW surface sites, it's just not a standard measurement at most sites.

We apologize for this incorrect statement, which has now been corrected as :' Currently, aerosol liquid water content is not systematically measured at the ground sites of the global atmosphere watch (GAW) network and nor at a global scale'

P23425 lines 1-2 ***"..or use an empirically-derived dependence of extinction coefficient on relative humidity f (RH).."*** **Interesting – I would say that the majority of papers I've read comparing in-situ vertical measurements with AOD retrievals (more in the climate sphere than the AQ sphere) utilize measured hygroscopicity rather than 'empirically derived' functions.**

We have clarified this sentence to indicate that the "empirically derived" function is the same thing as the "measurement derived" gamma.

P23425 line 14 "..over geographically complex source regions.." Seems like the above statement contradicts this earlier statement: "The eastern United States has been shown to be a good location for ascertaining $PM_{2.5}$ information from aerosol optical depth (AOD) due to (1) more uniform vertical distribution of aerosols, (2) chemical composition that is dominated by sulfates, (3) a uniform topography and (4) widely distributed anthropogenic emission sources (Engel- Cox et al., 2006)." (from p23423, lines 19-23).

Good point. The text has been modified to be more clear. "The DISCOVER-AQ strategy is to make systematic, co-located observations of aerosol properties by in-situ and remote-sensing techniques over a large diversity of source regions"

P23425 line 15 "*San Juaquin*" → "San Joaquin" P23425 line 15 "*the Houston, TX*"→ "Houston, TX" unless you meant "the Houston, TX region"

Done.

P23425, line 22 "...*P-3B instrumentation and the observations...*"→"...P-3B instrumentation and the ground-based observations..."

Done.

P23427 line 1-3 *"Beltsville, Fairhill and Edgewood were also equipped with in-situ aerosol and trace gas monitors that were operated within EPA's AQS network (http://www.epa.gov/ttn/airs/airsaqs/)."* Add after the above sentence – 'Relevant to this study are the EPA's PM2.5 and ozone measurements.'

Done.

P23427, line 19 *"...corrected from angular truncation..."* "corrected for angular truncation and other neph non-idealities" (AO1998 also corrects for non-idealities in the neph light source and I think something else as well)

The sentence has been reworded per the reviewer's suggestion. 'The scattering coefficient has been corrected from angular truncation errors and illumination intensity non-idealities based on Anderson and Ogren (1998).'

P23428, line 14-15 "..and was inversely correlated with the organic mass fraction of the aerosol (Beyersdorf et al., 2013)." I'm assuming Beyersdorf will cite the seminal work by Quinn relating hygroscopicity and organic mass fraction. Since Beyersdorf isn't published – it might be useful to cite the Quinn paper.

The citation has been added.

P23428, lines 16-18 "Recently, Ziemba et al. (2013) presented a statistical comparison of in-situ extinction coefficient measurements coincident with remote-sensing observations performed by the HSRL (both measurements were performed at 532 nm)." Just out of curiousity – were these different in-situ measurements (e.g., using an extinction instrument measuring at 532 nm? If these were the same measurements as described here then this should be rephrased because the scattering portion of extinction would need to be adjusted to 532 nm.

The extinction coefficients were not measured directly and are calculated from the sum of the scattering coefficient (measured at 550nm and adjusted at 532nm) and the absorption coefficient (measured at 532 nm). Thus it was not correct to state that the extinction 'measurements' were performed at 532nm. Good point. The sentence has been rephrased per the reviewer's suggestion.

"Recently, Ziemba et al. (2013) presented a statistical comparison of in-situ extinction coefficient measurements (adjusted at 532 nm) coincident with remote-sensing observations performed by the HSRL (measured at 532 nm)."

P23428 – should state where the measurement of ambient RH came from, i.e., standard meteorological measurements on the P3 or whatever.

The ambient RH measurements were performed by a hygrometer located outside the aircraft.

P23429 – eq 1 –the ‘wet’ neph measures at 80% is not necessarily measuring at ambient RH, correct? I would recommend changing the subscript in equation 1 to ‘wet’ and clarifying the procedure to determine fRHamb: first gamma is calculated using the wet neph RH (80%) and the dry neph RH. Then that calculated gamma value is used in conjunction with the measured ambient RH and the dry neph RH to determine the actual fRHamb value. Should also note that equation 1 does not model deliquescent aerosol.

The subscripts have been modified in the equation 1 as follows :

$$fRH_{amb} = \frac{\sigma_{ext,amb}}{\sigma_{ext,dry}} = \left[\frac{1-\frac{RH_{amb}}{100}}{1-\frac{RH_{dry}}{100}}\right]^{(-\gamma)} \qquad \textit{Equation 1}$$

with :

$$\gamma = \frac{ln\left[\frac{\sigma_{scat,80\%}}{\sigma_{scat,dry}}\right]}{ln\left[\frac{100-RH_{dry}}{100-80\%}\right]} \qquad \textit{Equation 2}$$

Moreover the referee is right to note that this equation is valid only for non-deliquescent aerosols. This point is clarified in the revised version of the manuscript : ‘According to the equation 1, the particle extinction efficiency is monotonically modified as RH increase or decrease and thus do not account for the hysteresis behavior of deliquescent aerosol particles (Fierz-Schmidhauser et al., 2010).’

P23428, lines 24 *“...the Gasso parameterisation* (Eq. 1)” This is one of those places where I’m pretty sure that calling this ‘the Gasso parameterisation’ is ignores a bunch of tandem nephelometer hygroscopicity research that has gone before. See for example work by Mark Rood’s group at the University of Illinois and Covert/Hegg group at the University of Washington. Work from both these universities, particularly a paper with lead author Carrico (I can’t remember what study unfortunately), cites even earlier work (from the late 1960s?) using a similar (identical?) function for hygroscopic growth. Furthermore there is no citation of a paper by Gasso to support calling equation 1 ‘the Gasso parameterization’.

Excellent point. As the referee suggested, the authors add several quotes to studies from the early 80’s to late 90’s that describe the aerosol optical properties as a function of the relative humidity. Moreover, this parameterisation will not be called Gasso parameterisation anymore. The list of publications that has been added is :

Carrico, C. M., Rood, M. J. and Ogren, J. A. 2000. Aerosol light scattering properties at Sagres, Portugal, during ACE-2. Tellus 52B, 694–715.

Day, D. E., and W. C. Malm (2001), Aerosol light scattering measurements as a function of relative humidity: A comparison between measurements made at three different sites, Atmos. Environ., 35, 5169 – 5176, doi:10.1016/S1352-2310(01)00320-X.

Hänel, G. (1976). The properties of atmospheric aerosol particles as functions of the relative humidity at thermodynamic equilibrium with the surrounding moist air. Adv. Geophys., 19, 74–189.

Hänel, G. (1984). Parameterization of the influence of relative humidity on optical aerosol properties. In H. Gerber and A. Deepak, editors, Aerosols and their climatic effects, pages 117–122. A. Deepak, Hampton, VA.

Hegg D., Larson,T., and P.-F. Yuen, A theoretical study of the effect of relative humidity on light scattering by tropospherical aerosols, J,. Geophys. Res., 98, 18, 435-18, 439, 1993.

Tang, I. N. (1996), Chemical and size effects of hygroscopic aerosols on light scattering coefficients, J. Geophys. Res., 101(D14), 19,245 – 19,250, doi:10.1029/96JD03003.

Carrico, C. M., M. J. Rood, and J. Ogren A., 1998: Aerosol light scattering properties at Cape Grim, Tasmania, during the First Aerosol Characterization Experiment. J. Geophys. Res., 103, 16 565–16 574.
Gassó, S., Hegg, D. A., Covert, D. S., Collins, D. R., Noone, K. J., Öström, E., Schmid, B., Russell, P. B., Livingston J. M., Durkee, P. A., and Jonsson, H. H., 2000. Influence of humidity on the aerosol scattering coefficient and its effect on the upwelling radiance during ACE-2. Tellus, 52B (2), pp 546-567.

P23429, lines 7-10 "and assuming a particle refractive index of 1.53 – 0.00 i for ammonium sulfate (Ziemba et al., 2013). This closure exercise (slope of 0.991±0.004 and R_2 of 0.98) gives confidence in both the _$_{scat,dry}$ and dry size distribution measurements." Do you need the PSAP measurements at all? This closure result suggests that there is virtually no absorbing aerosol.

The reviewer is correct. During this study, aerosol absorption represents less than 5% of the total extinction However, the closure exercise is limited to the comparison of the measured scattering coefficient (with the integrating nephelometer) to a calculated scattering coefficient using Mie code, the measured size distributions (UHSAS), and the refractive index of ammonium sulphate.

P23429 SP2 measurements – if they aren't used don't take up the space with mentioning them here...

Done.

P23431 lines 4-5 *"...The comparison shows good correlation (R_2 = 0.96 for each wavelength),"* **Presumably you have used the Ångström exponent of scattering from the neph and absorption from the PSAP to adjust the P3 values to the AERONET wavelengths for figure 3. You should state that somewhere – in the P3 instrument description you only mention adjusting the PSAP to 550 nm.**

The statement has been added.
" To evaluate whether the measured AOD$_{P-3B}$ are representative of the entire atmospheric column, values were directly compared to the AOD measured by the AERONET sun photometers (AOD$_{TOA}$, see Figure 3), which is considered a reference for AOD measurements (Holben et al., 1998). (…) The AOD$_{P3B}$ was calculated using the measured scattering coefficient adjusted to 440, 500 and 675nm using the scattering Ångströmangstrom exponent and the measured absorption coefficient adjusted tot he same wavelength using the absorption Ångströmangstrom exponent. The comparison shows …"

P23431 lines 9-14 "The larger offset (23%), compared to the 11% offset calculated by comparing AOD$_{HSRL}$ and AOD$_{P-3B}$ mostly due to inlet and dryer particle losses, may be due the presence of an aerosol layer above the HSRL flight level (above 8.5 km), incorect AERONET AOD cloud screening, or underestimation of the contribution below the P-3B profile height (closest to the surface)." This sentence is confusing – is it saying that the main issue with the HSRL/P3 comparison is inlet/dryer losses, but the AERONET/P3 comparison may have additional causes resulting in a larger offset? I would rephrase the sentence. It seems that inlet/dryer losses and the estimation of aerosol between 0-300 m would also be a problem with the HSRL/P3 comparison. You may also want to look at Esteve et al (2012?) which looks at reasons for differences between AERONET and in-situ measurements. Also 'incorect' is spelled incorrectly.

Good point. The manuscript has been corrected according to the referee's suggestion :
"Esteve et al. (2012) listed the different hypotheses to explain the discrepancy between the AOD measured by AERONET and the one calculated from the in-situ measurements. The good agreement (discrepancy of 11%) between the in-situ and HSRL measurements allow us to estimate at 11% the errors due to measurements adjustments (angular truncations, wavelength changes, hygroscopic growth), particle losses, underestimation of the contribution below the P-3B profile height. Thus, the larger offset (23%) observed between the AOD from AERONET and from in-situ measurements may be due to underestimation of AOD contribution below the P-3B profile height, the presence of an aerosol layer above the HSRL flight level (above 8.5 km), incorrect AERONET AOD cloud screening, the temporal variability of the aerosol optical properties, and the bias in the AERONET measurements."

P23432 , lines2-3 "*Each integral of the Eq. (3)*..." → "Each integral in Eq. (3)..." P23432, line 10 caped→capped

Done. These typos have been corrected.

P23432, line 10-12, *"The aerosol present in the BuL accounts for 48% of AOD*$_{P\text{-}3B}$ *compared to 46% within the BL (Table 1)."* Perhaps make it more clear for the three types of profiles that the relative % contribution of AOD for the different sections of the vertical profie is the mean (median) contribution for profiles of that type rather than for the individual profiles represented in figure 4. Note: I think this whole paragraph is confusing – there are percentages of types of profiles and percentages of contributions from different parts of the profiles and it's poorly organized. Perhaps talk about the percentages of each type of profile first and then talk about characteristics of the profiles types after that. See my comments about Table 1 as well.

Thank you for pointing this out. The paragraph has been rephrased to improve clarity.
"The first study case on the 20 July 2011 (Figure4a) represents more than 60% of observations and highlights high values of $\sigma_{ext,amb}$ *within the BL. The second study case on 21 July 2011 (Figure4b), represents 17% of the observed profiles and shows the presence of the aerosol capped by the top of the BuL. Finally, an aerosol layer with significant* $\sigma_{ext,amb}$ *values (550 nm) can be detected above the buffer layer (case on 28 July 2011, Figure4c). The presence of an aerosol layer accounting for a large part of the* AOD_{550nm} *has been observed in 23 % of the profiles. The Table1 describes the averaged contribution of the aerosols present whithin each layer to the total* AOD_{550nm} *for the three different vertical aerosol distributions observed during DISCOVER-AQ. One can see that the BuL contribution to the total AOD is in average larger than 38% and is then non-negligeable. Moreover, when an aerosol layer is present aloft the BuL, the averaged contribution of the aerosol within the BL to the total* AOD_{550nm} *decrease to 16%."*

P23432, line 16 *"Alternatively…"* Alternative to what?

The word has been removed

P23432, line 19 *"Furthermore, the value of* _$_{ext,dry}$ *has an averaged small variability within the BL (< 9 %)."* Presumable you mean for a given profile? Rephrase "...has a variability of 9% within the BL based on BL averages for all profiles.

Done.

P23432, line 22 *"...commonly cited in literature (Koelemeijer et al., 2006)."* →"... commonly cited in literature (e.g., Koelemeijer et al., 2006).

Done.

P23432, eq 5 - to generalize equations 3 and 5 wouldn't it be better to replace AOD_P3B with plain AOD? They should be equally true of any vertical measurements of extinction (e.g., in-situ, lidar)

The referee is right. The equations have been generalised.

P23433, line 6 *"serie"*→series

Done.

P23433, lines 6-8 *"This time serie highlights the large variability of the hourly averaged PM$_{2.5}$ within a highly polluted period (Julian Day 201–205, PM$_{2.5}$ greater than 30 μgm^{-3}) and a clean period (Julian Day 195–199, PM$_{2.5}$ less than 10 μgm^{-3})."* This is, perhaps, a quibble about phrasing – are you trying to contrast the wide range in PM2.5 values between clean and polluted periods or say that there is lots of variability within clean periods and within polluted periods? It sounds like the latter, but I'm guessing the former was what was intended.

Our intent is to highlight the large range of PM2.5 values, and have rephrased this sentence to make this more clear.
"This time series highlights the large range of the hourly averaged PM$_{2.5}$ values within a highly polluted period (Julian Day 201-205, PM$_{2.5}$ greater than 30 $\mu g\ m^{-3}$) and a clean period (Julian Day 195-199, PM$_{2.5}$ less than 10 $\mu g\ m^{-3}$)."

P23433, lines 14-16 *"The back trajectories showed that the highly polluted periods were associated with air masses coming from the Ohio River valley, a region typically associated with power plant emissions."* Where were the back trajectories for clean periods from? Should also comment on that even if there was no consistent pattern.

Good point. We have added text indicating that no systematic pattern has been observed for origins of clean air masses.

P23433, lines 26-27 *"...and the average aerosol extinction..."*→"...and the average dry aerosol extinction (wavelength=XXX nm)"

The sentence has been rephrased.
"The aerosol MEE$_{550nm}$, sensitive to both aerosol physical and chemical properties, was calculated using the PM$_{2.5}$ and the average dry aerosol extinction (550 nm) measured at the lowest P-3B flight altitude."

P23434, line 16 "*...may have cause...*"→"...may have caused..." P23434, line 27-28 *"...which is in good agreement to the values..."*→"...which is in good agreement with the values..."

We corrected these two sentences.

P23435, lines 19-20 ***"Indeed, the non-linearity can be avoided using a threshold value for the Ångström exponent (less than 2.4)."*** **Where does this value of 2.4 come from – it should either be cited or discussed based on the presented data. Also, Ångström is a proper noun and should be capitalized (there are also diacritical marks circle over the 'A' and umlaut over the 'o' for it to be written completely correctly).**

This threshold value is based on the data shown in Figure 2 *(herein). The non-linearities between the AOD and the* $PM_{2.5}$ *were found for effective radius lower than 100nm. From Figure 2, the fine particles are clearly associated with the larger values of the Ångström exponent. The use of the 2.3-2.4 thresholds for the Ångström exponent would effectively remove the non-linearities due to the presence of fine and ultrafine particles from the data set. The Figure 2 will be added as supplemental material and part of this discussion has been included in the manuscript.*

P23436, line 19 "*The BL and BuL heights are used to represent the height of this mixed layer*" Do you mean BL and BL+BuL? As suggested further on in the paragraph? Note the figure caption for figure 7 only says BL or BuL. Should be clarified and caption/figure labels corrected if needed.

In this paragraph, the authors wanted to estimate the validity of using the concept of a mixed layer. To evaluate which layer (BL or BL + BuL) should be used as the mixed layer, the authors used both layers (BL and BuL) height corresponding to the BL and the BL + BuL. Indeed, the use of the BL is justified because the $PM_{2.5}$ *measured by air quality station is directly related to the aerosol present within the BL. Using the BuL height, bounding the height of the layer (including both layers, the BL and the BuL) in which most of the aerosols are observed, is also justify cause the column-integrated measurement (e.g., AOD) is related to the particle presence throughout the column. To clarify that in the manuscript we modified the sentence quoted by the referee into:*
The BL and then the BuL (bounding the height of the layer (including the BL and the BuL) in which most of the aerosols are observed) heights are used to represent the height of this mixed layer.

P23436, line 20-21 ***"represents the BL and the BL + BuL contribution to the total AOD (Fig. 7a and c, respectively)"*** **→"represents the BL (Fig 7ab) and the BL + BuL contribution to the total AOD (Fig. 7cd)"**

Done.

P23436, line 27 "The slopes of these tendencies..."→The slopes of these PM2.5 vs volume relationships..." this discussion of density is a little distracting. Perhaps remove?

Done.

P23436, line 29 ***"Strong relationships (with low variability)..."*** **I would move this sentence up to where you are specifically talking about figure 7bd.**

Done.

P23437, line 3 "*the haze layer concept*" What is the haze layer concept? Do you just mean the BL+BuL?

The haze layer concept has been clarified in the manuscript. The haze layer is commonly used by the Lidar community and is defined as the level where the aerosol extinction coefficient decreases to 1/e of that on the top of the BL. The haze layer include thus the BL and a large part of the BuL.

P23437, lines 6-8 "The same study has been done using the haze layer calculated from the HSRL measurements (Scarino et al., 2013) and showing similar improvements (Fig. 7c and d, R_2>0.95)." This sentence is confusing – does Fig 7c include HSRL measurements? Please clarify

The figure reference was incorrect, which is now fixed.

P23437, lines 8-10 "Nevertheless, the haze layer is a Lidar product and might not been available for most of the AOD and $PM_{2.5}$ relationship." Bad grammar.

Fixed.

P23437, lines 10-11 "Thus, this results show that using the BuL instead of the BL from radiosounding measurements will improve the $PM_{2.5}$ retrivals from the AOD." Do you mean BL+BuL? Or the height of the BuL which is BL(z)+BuL(z)?

Good point. This has been clarified in the text. 'By integrating Equation 2 from the surface to the top of the BuL (available from radio soundings), the relationship between the AOD_{550nm} and the $PM_{2.5}$ is strongly improved (Figure 7c and d, $R^2 > 0.95$ compare to $R^2 \sim 0.84$ using the BL).' Indeed, using the height of the BuL means to integrate from the surface to the height of the BuL and this layer thus includes the BL and the BuL.

P23437 lines 12-13 *"Few cases show the presence of an aerosol layer above the BuL similar to the case study shown in Fig. 2c"* Do you mean figure 4c? on P23432, line 15 it sounds like 23% of the cases have an aerosol layer like fig 4c.

This has been replaced by : 'Some cases (23% of the profiles) show the presence of an aerosol layer above the BuL similar to the case study shown in Figure4c.'

P23437, lines 13-14 "*From the profiles of the AOD contribution, the layers above the BuL contribute to more than 10% of the total AOD, on average*" From which profiles? All profiles or just the ones that look like figure 4c? also – 'to more than 10%' do you mean 'no more than 10%'? otherwise you should clarify how much more than 10% because that could potentially be a significant contribution.

This paragraph has been corrected into : "While we show that using the BuL height as the aerosol layer top to be reasonable for the large observational data set obtained in the Baltimore-Washington D.C., there may be other locations where this assumption does not hold. Indeed, the systematic presence of an aerosol layer above the BuL (only 23% of the profiles in this study) would increase the variability of the correlation shown in Figure 7d."

P23437, line 23-24 "...and f (RH) constriction due to similar properties of the aerosol sampled within the BL and the BuL"→"...and f (RH) constraints. We assume this is due to the similar properties of the aerosol within the BL and BuL, e.g., f(RH) and Ångström exponent (Fig 8)."

The reviewer is correct.

P23437, lines 24-29 "The comparison of the scattering Ångström exponent (between 450 and 700 nm) and the f (RH) measured during each P-3B profile performed over the DISCOVER-AQ ground sites (Fig. 8) and the average within the BL and the BuL highlights strong similarities of the aerosol physical and chemical properties in each layer."

This has been done. "Figure 8 shows a comparison of BL and BuL values for scattering Ångström exponent (between 450 and 700 nm) and f(RH=80). The plots show the parameter averages for each vertical layer (BL or BuL) during each P-3B profile and highlight the strong similarities of the aerosol physical (represented by Ångström exponent) and chemical (represented by f(RH=80) in each layer."

23438, lines7-8 are Twohy and Schuster really the first references to note aerosol properties might change at high RH?

A reference to Hänel, 1984 has been added showing the historical study of the relationship.

23438, eq 6. This water fraction equation is also equivalent to 1-f(RH)amb, right? You could provide a second x-axis on figure 9 with the values of f(RH)amb.
By integrating the f(RH)amb over the column, the equation 1 would become

$$f(RH)_{amb,integrated} = \int \frac{\sigma_{ext,amb}}{\sigma_{ext,dry}} = \frac{AOD_{amb}}{AOD_{dry}}$$

Thus,

$$WF = 1 - \frac{AOD_{dry}}{AOD_{amb}} = 1 - \frac{1}{f(RH)_{amb,integrated}}$$

As the f(RH)amb is not constant throughout the entire column, the integration of f(RH)amb would introduce large uncertainties on this value. This is why the authors chose to use the water fraction concept.

23438, lines 14-21 There has got to be an AOD climatology paper that would be a more appropriate reference for AERONET AOD values at different site regimes. How about the paper by Augustine which summarizes AOD at several remote rural sites including the Oklahoma site mentioned in the Andrews 2011 paper. MLO is perhaps a bad site for comparison. It's in the free troposphere at night, but AERONET measurements occur during the day so the average AOD measurements at MLO are more likely to be representative of upslope air from the coastal communities below the volcano unless the data are screened for that.

The results of Augustine et al. (2008) have been added to the manuscript. Their results show that the AOD_{550nm} *over these areas are between 0.11 and 0.47.*

P23438, line 24 ***"…aerosol loadings versus the contribution of water uptake and the aerosol loading…"*** **I would rephrase: "…aerosol loadings versus the combined contribution of aerosol loading and water uptake…"**

Done

P23438, line 25 "The AOD is increasing with the water fraction (Eq. 6), on average from 0.15 for AOD around 0.1 to 0.35 for AOD around 0.35, showing that the larger AOD values (> 0.4) are mainly driven by aerosol water uptake…" This is a confusing sentence. I think what you mean to say would be clearer if you said: AODamb increases with water fraction by XX% for AODdry around 0.1 to XX% for AODdry around 0.35. This suggests, that at this location and for these profiles, the larger AOD values are mainly driven by water uptake."

The statement has been rephrased. 'The AOD_{550nm} *increases with the water fraction (Equation 6) by 15% on average for* $AOD_{dry,550nm}$ *around 0.1 to 35% for* $AOD_{dry,550nm}$ *around 0.35. This results suggest that, at this location and for these profiles, the larger AOD values (> 0.4) are mainly driven by water uptake.'*

Also, please clarify - are the higher AOD cases also associated with higher ambient RH values? Is this a meteorology effect or an aerosol chemistry effect or a combination of the two?

Excellent point and we clarify that point in the manuscript. The AOD values lower than 0.1 have been observed at significantly lower ambient RH. All the other AOD values were observed in a similar range of humidity. Thus the increase of the WF is not only due to an increase of the ambient RH but to a combination of the aerosol chemistry and the meteorological conditions.

P32439, lines 5-11. This section is very confusing to me. Are you trying to separate the contribution of aerosol water AOD and aerosol AOD? Are the AODP3-B values for dry or ambient AOD? In the figure, presumably the volume measurements are at low RH while the AOD are at ambient RH so you are observing that low aerosol volume corresponds with low hygroscopicity (low fRHamb). Please make this section more clear – I think there is something interesting to talk about here.

In this section we are trying to isolate the effect of the water uptake from the aerosol vertical distribution. To do so we used only the observations performed when the BL contribution is larger than 60%. So the difference observed are not anymore linked to the presence or the absence of an aerosol layer aloft the BL. In the figure 10, the differences between both tendencies are only due to a f(RH) effect (aerosol chemistry and ambient relative humidity). Thus, the results show that knowing the f(RH) of the aerosol is essential to retrieve the aerosol volume concentration from AOD at ambient RH.
The referee is right to note that the larger volume concentrations correspond to the highest value of fRH_{amb}*. Statistically, the number of measurements is not sufficient to conclude on this point. For example, over dust sources, the larger volume concentrations would be associated with dust plume that have relatively low f(RH) values. Indeed, Zieger et al. (2013) reported f(RH) values for transported dust (most likely chemically modified) of 1.28.*

Zieger, P., Fierz-Schmidhauser, R., Weingartner, E., and Baltensperger, U.: Effects of relative humidity on aerosol light scattering: results from different European sites, Atmos. Chem. Phys. Discuss., 13, 8939-8984, doi:10.5194/acpd-13-8939-2013, 2013.

P32439, lines 12-22 this section is also not clear. The paper by esteve et al in ACP also found better agreement between in-situ derived AOD and aeronet AOD when atmospheric RH was low. I think the point you are trying to make is that there can be vertical variations in RH which will affect aerosol optics so surface measurements of hygroscopicity or RH may not be representative of the column. I would rephrase this section. I would also put a second set of points on the plot showing the ratio of RH_ground/RH_P3-B. That's the more important ratio as vertical profiles of RH are more readily available from sondes.

Thank you for pointing this out. The requested data have been added to the figure. The variability of the f(RH) was really small throughout the column while the ambient relative humidity was extremely variable throughout the column. It is thus not as easy to compare the RH profile with the ground measurements. We added in the figure below the ratio of the maximum and the median RH observed throughout the column to the RH measured at the ground. One can see that large errors could be made assuming that the RH remains constant throughout the column.

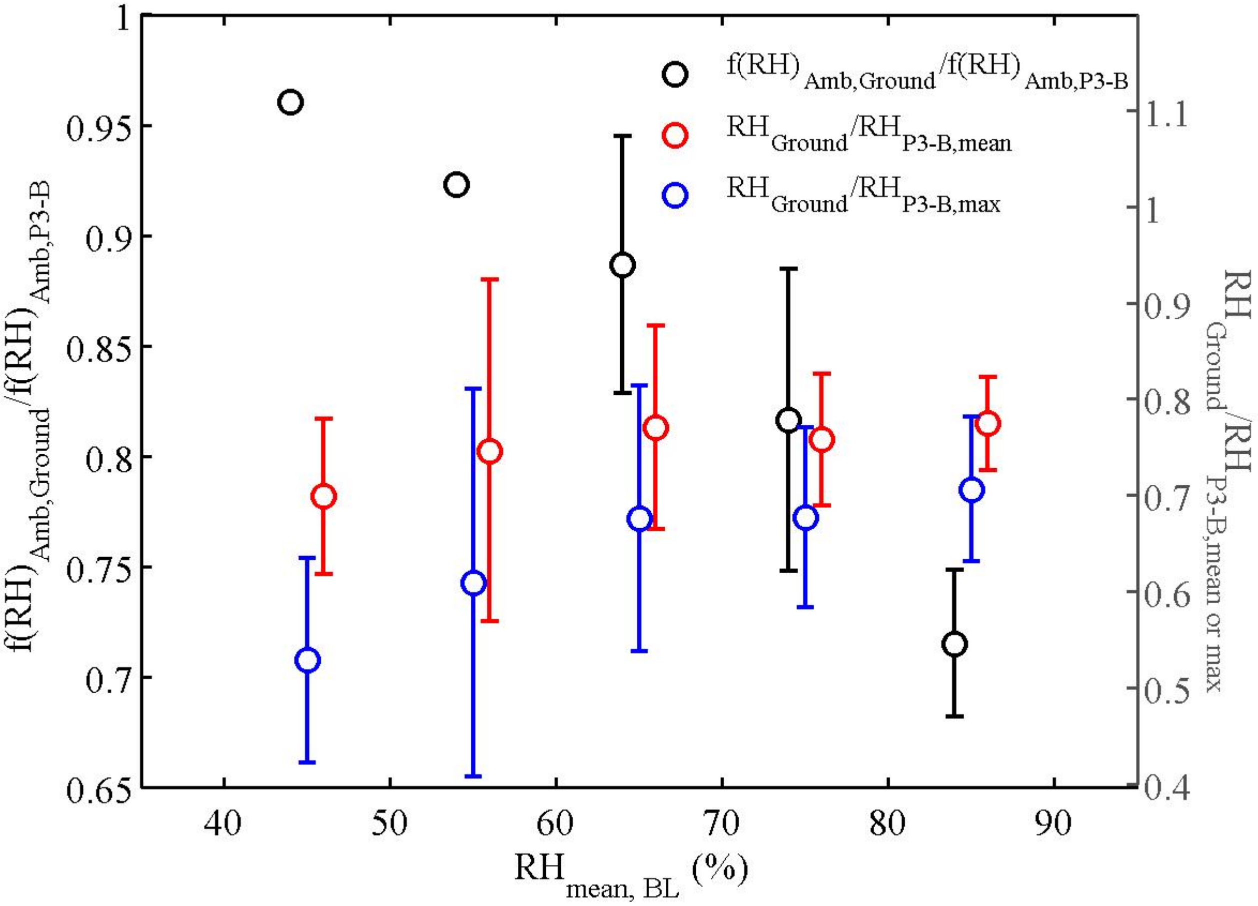


Figure 4 : Ratio of the $f(RH)_{amb,ground}$ to $f(RH)_{amb,P\text{-}3B}$, ratio of the RH averaged throughout the column to the RH measured at the ground, and the ratio of the maximum RH value observed throughout the column to the RH measured at the ground as a function of the averaged relative humidity measured during the P-3B profiles within the BL.

P23440 lines 2-4 "The measurements were performed during one month and show that the aerosol mass concentrations ($PM_{2.5}$) measured at the surface (EPA ground sites) are driving the AOD." In section 5.1.2 it seems like you are trying to make the argument that AOD is driven by RH. please clarify

This has been clarified in the manuscript. In the section 5.1.2 the authors show that the larger values of the AOD were driven by the water uptake. The figure 3 shows the AOD and the PM2.5 time series and both tendencies show strong similarities.

P23440 lines 14-16 "...which highlight different tendency as a function of the presence and the optical thickness of the elevated aerosol layer." This phrase doesn't make sense

We have reworded the phrase. "The contribution of the aerosol present within the BL to the total AOD is used to constrain the relationship between the AOD_{550nm} *and* $PM_{2.5}$*. As a result, different AOD versus* $PM_{2.5}$ *slopes are observed as a function of the presence and the optical thickness of the elevated aerosol layer."*

P23440 lines 17-18 "Using the BuL instead of the BL top as the height for the aerosol layer dramatically improves the $PM_{2.5}$ estimation." Rephrase: Using the top of the BuL instead of the BL top as the height...."

Done.

P23440 lines 19-20 "The f $(RH)_{amb}$ effect on the estimation of the $PM_{2.5}$ is secondary compared to the BL contribution and induced an error factor of 1.6. Comparision of the observed f $(RH)_{amb,P\text{-}3B}$ and the calculated f $(RH)_{amb,ground}$ show that the errors are lower than 10% when the RH within the BL is lower than 55% while the errors are larger than 19% when RH within the BL is larger than 75 %." Rephrase. This isn't clear. Presumably you mean the error in the calculation of PM2.5. you should say what direction the error is – over or under estimate of PM2.5. Related to my comments in the text – the more important thing is humidity and perhaps this entire section should be re-evaluated.

The statement has been rephrased to be clearer. However, $PM_{2.5}$ *retrieval from column integrated measurements is not an easy task. Indeed, this analysis shows that the aerosol vertical distribution needs to be known as well as the water uptake. Using this data set, we show that the presence of an aerosol layer above the BL lead to an overestimation of the* $PM_{2.5}$ *by a factor of 4 while the water uptake would introduce only an error up to a factor of 2. It appears thus that the aerosol vertical distribution has a larger impact than the humidity during this campaign.*

PM23440 line 27-28 *"…shows a less-pronounced improvement.."* **→ is less important for estimated PM2.5**

This has been corrected.

PM23440, line 26 "...The generally dry conditions observed throughout the study may..." did you quantify the 'dry' conditions in the main text of the manuscript? i.e., were you able to compare column average RH for the time of the study with a climatology of column average RH for the reason – 2x daily sondes are available for that sort of analysis.

The referee is right. The authors wanted to highlight here that all the flights were performed during clear sky conditions and that the presence of clouds in the scene could dramatically change the impact of the f(RH) on the PM2,5 retrievals. The sentence in the manuscript has been corrected into : 'During clear sky conditions, the f(RH) variability is found to be a second-order effect in the overall estimates. Since these results are representative of the Baltimore-Washington metropolitan area, extrapolating these results to other geographical locations must be done with care.'

References Beyersdorf is not in alphabetical order

Done.

Tables Table 1 →refers to figure 3abc, but should refer to figure 4abc. →Caption has grammatical and spelling errors.

Done.

→Add another row to table giving percentage of each profile type. →if the percentages are indeed just for the 'case study profiles' then I would recommend deleting this table or changing so the percentages are for the profiles of each type.

Done.

Table 2 →Caption has grammatical errors →Why not include all profile sites in this table? →having all blanks for Beltsville is not useful – put a note at the bottom of the table explaining why no values for Beltsville

Done.

' (a) The profiles performed over Beltsville were limited by the air traffic controlers to 1.5 km which may have caused an underestimation of the AOD_{550nm} *and limited the exploration of the BuL.'*

Figure 1 – it might be helpful to put a scale on the map as well as lat/long coordinates

The latitude and longitude already appear on the figure axis.

Figure 3 – (a) include a regression on the plot (b) color code the dots by wavelength – e.g. blue dots=440 nm, green dots=500 nm and red dots=675 nm. Also it's not clear the value of including all wavelengths on the plot – you could probably do just one of them and state that similar results were observed for all three wavelengths.

The AOD comparison (AERONET and in-situ) for all wavelengths shows good agreement, which is an important result, so we prefer to include all wavelengths in the figure.

Figure 4 what wavelength of extinction in figures 4abc and what wavelength of AOD in figure 4d?

550nm. This has been clarified in the figure caption.

Figure 5 what wavelength of AOD? →Why no R2 for the blue equation? →what does the black font equation represent? All points or points not included in the blue fit?

The wavelength used to calculate the AOD is 550nm, which has been clarified in the caption. The black font equation is associated with the grey fit. It corresponds to the comparison of all the PM2.5 data with the all the retrieved AOD values. The authors used the blue color to distinguish the cases when fine aerosols (Dp < 100nm) are observed and to highlight the non-linearities of the $PM_{2.5}$*- AOD relationship associated. The goal of the blue equation was to estimate the errors due to these non-linearities. The* R^2 *has been added to the figures.*

Figure 6 caption does not match plot. If plot is correct please provide AOD wavelength

We apologize for this oversight. The caption has been corrected to match the plot.

'$PM_{2.5}$ *measured from the EPA ground sites (Beltsville, Edgewood and Fairhill) as a function of the* AOD_{550nm} *calculated using extinction profiles performed by the P-3B when an elevated aerosol layer aloft the BL was observed (layer aloft, red squares) or absent (well mixed, black dots).'*

Figure 7 **"Fig. 7. Ambient AOD measured by the P-3B as a function of the Volume concentration weighted by the BL height and the f** $(RH)_{amb}$**. The color code represents the BL contribution to the AOD and the size of each dot correspond to the BL height. The red line is coresponding to the linear fit of cases where the the AOD contribution of the BL is higher than 75 %." →please provide AOD wavelength. →lower case Volume →volume conc. weighted by BL(z) and f(RH) is a function of AOD the way the plot is presented →coresponding is spelled wrong, and rephrase: 'The red line corresponds to...' →two 'the's in front of AOD in last sentence →why are there 4 different plots →why does only one of the plots have a fit line? →figures should be described more clearly in the caption for each of the abcd labels.**

The caption has been corrected. All figures are slightly different from each other. Figure 7a and c represent the PM2.5 weighted by the BL (BuL) height and the $f(RH)_{amb}$ *(a and c respectively) as a function of the ambient* AOD_{550nm} *measured by the P-3B. Due to some technical issues, the PM2.5 measurements were not as numerous as the volume concentrations performed aboard the P3-B. The authors decided to use the volume concentrations (Figure b and d) in order to increase the robustness of the comparison.*

Figure 8 "Fig. 8. Comparison of the Ångström exponent between 450 and 700nm (a), and the f (RH) (b) averaged within the BL (Boundary Layer) and the BuL (Buffer Layer) at the different DISCOVERAQ sites (Beltsville, Padonia, Fairhill, Aldino, Edgewood and Essex). The black line corresponds to the 1 : 1 line and the gray area represents the 10% variability." →Rewrite caption. Very confusing. Something like: Comparison of aerosol properties averaged within the BL and the BuL for all DISCOVER-AQ sites (a) the Ångström exponent between 450 and 700nm (b) f(RH). The black line corresponds to the 1 : 1 line and the gray area represents the 10% variability." →what wavelength f(RH)? →Label axes better. How about something like Ångström in BL and Ångström in BuL or subscript the BL and BuL →presumably this is f(RH=80) not f(RH)amb. Should clarify in caption and text. →provide linear fit equation and correlation coefficient so readers can have feel for correlation.

The caption has been corrected per the reviewer's suggestions.

' Comparison of the aerosol properties (Ångström exponent between 450 and 700nm (a), and the $f(RH=80)_{550nm}$ *(b)) averaged within the BL (Boundary Layer) and the BuL (Buffer Layer) for all the DISCOVER-AQ sites (Beltsville,*

Fairhill, Aldino, Edgewood and Essex). The black line corresponds to the 1:1 line and the gray area represents the 10% variability.'

Figure 9 "Fig. 9. Frequency of the AOD retrieved from ambient extinction coefficient measured aboard the P-3B and the water fraction (WF, %) associated with observed dry (P-3B) and ambient (P-3B) AOD." →what wavelength AOD? →presumably the black line is the water fraction? →why do you need to say dry and ambient P3 AOD? Can't you just say ambient?

The caption has been modified per the reviewer's suggestions.
'Frequency of the AOD_{550nm} *retrieved from ambient extinction coefficient measured aboard the P-3B and the water fraction (WF in percent, black line) calculated using the dry and ambient* AOD_{550nm} *measured aboard the P3-B (see Equation 6).'*

Figure 10 what wavelength AOD?
550nm. This has been corrected in the figure and in the caption.