We thank the reviewers for their thoughtful and thorough reviews. Point-by-point responses to the reviewers' comments are attached below.

Response to Reviewer #2 comments

_____

Cavity ringdown (CRD) and cavity enhanced (CE) spectroscopy are established techniques for measuring aerosol optical properties. Some of these instruments use broadband light sources to acquire the sample's extinction spectrum, thus enabling the aerosol extinction to be separated from structured gas phase absorptions (in this case from NO_2). Here the authors combine a CE system with an integrating sphere (IS) to additionally access aerosol scattering measurements – although this has been done before in single-wavelength cavity systems, to my knowledge this work is the first to incorporate an IS into a broadband CE system. The authors have tested their instrument in the lab using size-selected polystyrene aerosol; they also show some convincing examples of ambient aerosol and NO_2 measurements which they compare with commercial instruments. The instrument development aspects of the work have been done well. There are deficiencies in other aspects, however, which need to be addressed before the work is suitable for publication: not placing the work into the wider scientific context [1] nor appropriately citing other previous work [3,17,24,27,32], inadequate or ambiguous explanation about how the authors did the work or drew their conclusions from the data [2,4,5,6,7,8,28,33,36], and English language issues [9].

Comments
[1] The authors need to do more to place their work into context. The Introduction would benefit from a brief discussion of the atmospheric and climate relevance of aerosol scattering versus absorption, and hence establish why it is desirable to have an instrument that can quantify both these aerosol optical properties. This scene-setting information needs to come before the otherwise good & thorough review of existing instrumentation. At the other end of the paper, the Conclusions section should include examples of how the authors propose this new instrument could be applied to make atmospheric measurements, to address current uncertainties etc.

We have added following discussion in the "*Introduction*" section.

Atmospheric aerosols influence climate by modifying the Earth's energy balance through absorption and scattering of the incoming solar radiation (direct effects), changing the cloud properties and abundance (indirect effects), and changing the thermal structure of the atmosphere and the surface energy budget (semi-direct effects) (Ghan and Schwartz, 2007; Stier et al., 2007).

This radiative forcing (RF) capacity, characterized by the aerosol single scattering albedo (SSA) and its complex refractive index (RI), is mainly determined by the aerosol optical properties (scattering, absorption and extinction). The evaluation of the impact of aerosols on climate requires thus accurate, widespread and unbiased quantification of the particles optical properties as a function of the solar radiation wavelength, of their chemical composition and size distribution. However, despite several decades of research, the uncertainties of aerosol RF effects on the Earth's climate remain important (almost equal to the magnitude of the aerosol forcing) (Boucher et al., 2013).

Development of appropriate and well adapted measurement techniques for real time in-situ measurement of aerosol optical properties is an important step towards a more accurate and quantitative understanding of the aerosol climate effect (Strawa et al., 2003; Thompson et al., 2008).

References:

Ghan, S. J., and Schwartz, S. E.: Aerosol properties and processes - a path from field and laboratory measurements to global climate models, B. Am. Meteorol. Soc., 88, 1059-1083, 2007.

Stier, P., Seinfeld, J. H., Kinne, S., and Boucher, O.: Aerosol absorption and radiative forcing, Atmos. Chem. Phys., 7, 5237-5261, doi:10.5194/acp-7-5237-2007, 2007.

Strawa, A. W., Castaneda, R., Owano, T., Baer, D. S., and Paldus, B. A.: The measurement of aerosol optical properties using continuous wave cavity ring-down techniques, J. Atmos. Ocean. Technol., 20, 454–465, 2003.

Thompson, J. E., Barta, N., Policarpio, D., and DuVall, R.: A fixed frequency aerosol albedometer, Opt. Express, 16, 2191–2205, 2008.

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S.K., Sherwood, S., Stevens B., and Zhang, X.Y.: Clouds and Aerosols. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, 2013.

We have added the following discussion in the "*Conclusions*" part.

The instrument's sensitivity and specificity demonstrated in the present work show the potential of the developed Albedometer for field observation on different platforms (ground

observation networks, aircraft mapping, etc), by benefiting from its capacity of distinguishing of aerosol extinction from trace gas absorption. In addition, simultaneous measurements of aerosol scattering and extinction coefficients enable a potential application for retrieval of particle number size distribution and for faster retrieval of aerosols' complex RI. Moreover, unlike PAS technique, the measurement methods employed by the present albedometer are not affected (or much less) by RH, and are hence well suitable for the measurements of aerosol optical properties at high RH, in particular for the determination of complex RI of light-absorbing aerosols (such as black carbon and brown carbon) at high RH.

[2] Definitions of the terms involved in the key equation 1 on page 2989 were inadequate. Hence how the authors apply equation 1 for their measurements is unclear. The total extinction coefficient of an atmospheric sample is the sum of aerosol extinction, gas absorption and Rayleigh scattering by the gas: alpha_total = alpha_aerosol_ext + alpha_gas + alpha_Rayleigh. However if, as here, both the I(lambda) and reference I0(lambda) spectra are obtained in air, Rayleigh scattering is present in both spectra, and cancels. Consequently the measured absorption (alpha_meas = alpha_aerosol_ext + alpha_gas) is equal to the lower line of equation 1 but without the alpha_Rayleigh term. The measured extinction coefficient (alpha_meas) is also smaller than the gas sample's total extinction (alpha_total) by the amount equal to the Rayleigh extinction of air, due to the way the I0 reference spectrum was acquired. For this reason Washenfelder et al (2013) added a Rayleigh term for N_2 back into their version of equation 1 (their I0 reference gas was N2). Conversely, when the I(lambda) and reference I0(lambda) spectra are obtained in different gases (for example to measure the mirror reflectivity), the relevant quantity is the *difference* in Rayleigh scattering in the two gases, and this could be a positive (e.g. SF_6) or a negative (e.g He) quantity, see Fig 4b. Much more explanation is required here by the authors.

Thank the reviewer for the detailed comments. We have modified the first paragraph of section 3.2 as follows:

In IBBCEAS approach, wavelength resolved aerosol extinction can be calculated using the following equation (Fiedler et al., 2003; Washenfelder et al., 2008; 2013):

$$\alpha_{Total\ Ext}(\lambda) = \alpha_{Aerosol\ Ext}(\lambda) + \alpha_{Gas\ Abs}(\lambda) + \alpha_{Gas\ Rayleigh}(\lambda)$$
$$= R_L \left(\frac{(1-R(\lambda))}{d} + \alpha_{Gas\ Rayleigh}(\lambda) \right) \left(\frac{I_0(\lambda) - I(\lambda)}{I(\lambda)} \right) \quad (1)$$

where three components included in the measured total extinction $\alpha_{Total\ Ext}(\lambda)$: $\alpha_{Aerosol\ Ext}(\lambda)$, $\alpha_{Gas\ Abs}(\lambda)$ and $\alpha_{Gas\ Rayleigh}(\lambda)$ correspond to the aerosol extinction, gas phase absorption and Rayleigh scattering by the gas, respectively. R_L is the ratio of the total cavity cell length to the real cell length containing air sample when the cavity mirror is purged with gas flow. $R(\lambda)$ is the mirror reflectivity, d is the distance between two cavity mirrors, $I_0(\lambda)$ and $I(\lambda)$ are the

light intensities transmitted through the cavity without and with air samples, respectively. In our experiment, both the $I_0(\lambda)$ and $I(\lambda)$ spectra were more conveniently obtained in N_2 or air, the gas Rayleigh scattering was presented in both spectra and hence cancelled. The measured extinction can be rewritten as follows (Washenfelder et al., 2013):

$$\alpha_{Ext,Meas}(\lambda) = \alpha_{Aerosol\ Ext}(\lambda) + \alpha_{Gas\ Abs}(\lambda) = R_L \frac{(1-R(\lambda))}{d}\left(\frac{I_0(\lambda)-I(\lambda)}{I(\lambda)}\right) \qquad (2)$$

[3] Page 2989 line 20 to Page 2990 line 5 seems to be an abridged version of text drawn from Section 3.1 of Washenfelder et al, AMT, 6, 861, 2013. This Washenfelder 2013 work is not cited in this section of text; instead the Washenfelder 2008 work that is cited here is a carry-over from the text drawn from Washenfelder 2013. Additionally, the text and analysis approach in 2995 lines 13-25 is rather similar to section 3.3.1 of Washenfelder at al 2013 (which isn't credited on page 2995).

The reference of Washenfelder et al., 2013 is added in the revised version on pages 2989 and 2995.

[4] Three of the claims for the instrument's precision and accuracy in Section 3.3 looked optimistic (or at least, they represent very best case scenarios). [a] 2993 line 7 "lowest detection limit of 0.07 Mm^{-1} (on the scattering channel) with an optimum integration time of 459 s". It may be true that these values correspond to the very lowest individual data point around the minimum of the Allan variance plot in Fig 5(b). However the data points are quite scattered here, so the authors should choose values representative of the collection of data points around the minimum. Also, the minimum in the Allan plot for the extinction channel has an integration time of only 54 s. So it's not obvious what practical use a 459 s (but very sensitive) measure of the scattering would be if there is no equivalent extinction measurement – making both measurements together is one of the selling points of this new instrument. [b] Line 19 "the drift of the LED intensity is not included (in considering the accuracy of the extinction measurements)". This is an awkward omission because the Allan plot in Fig 5 says that, to achieve optimal performance, the I0 reference spectrum should be re-acquired every 54 s. Frequently re-recording the I0 reference severely limits how much time can be devoted to ambient air measurements. Indeed it may be impractical to frequently re-acquire I0 because it will take time for the N_2 flow (or zero air) to flush the cavity. Comment #2 by Reviewer #1 makes a related point. [c] Line 21 "The mean uncertainty in the determined (1-R) was less than 1%". I don't see how the authors can claim the (1-R) measurement's uncertainty is smaller than the uncertainties in the quantities on which this measurement relies. The uncertainties in the Rayleigh cross sections are 1% for N_2 and 3% for SF_6 [see 2991 line 8].

[a] In fact, each datum was acquired with an integration time of 9 s. The data points are a little scattered here, which is caused by the short integration time. We did not acquire the data with longer time to smooth the measured signals because we would like to have more data for Allan variance analysis. Measurement of aerosol extinction and scattering coefficients can be made with the same time interval. When higher sensitivity is required, longer data integration time can be used. In this case, data acquisition mode is different. For example, the scattering coefficient can be measured with an acquisition time of 432 s for one datum and the extinction can be measured with 54 s integration time for one datum, and the average value of 8 data was treated as one measured extinction coefficient (within 432 s).

[b] We have included the omission to the sentence: "the drift of the LED intensity is not included in considering the accuracy of the extinction measurements".

[c] The cavity was flushed with N_2 and SF_6 at 1.5 Lmin^{-1} rate for 40 min for each species, until the transmitted light intensity attained a stable value. Ten different pairs N_2 and SF_6 transmission spectra under stable condition were used for mirror reflectivity determination, and then 10 values of the mirror reflectivity were averaged. The mean value used as mean mirror reflectivity and the relative mean error of (1-R) was less than 1%.


[5] 2996 line 8 onwards. It would be preferable if the authors summarised the previous RI measurements on PSL aerosol before they state "our results [delete "were"] agree with: : :" The previous reports cited in the text are for different wavelengths from 470 nm used here. So establishing the agreement requires some interpolation – I would like to see a plot showing the real and imaginary parts of RI versus wavelength from the previous reports and this paper's new measurement to illustrate the interpolation process and the resultant agreement. An earlier study from these authors (Zhao et al., Analytical Chem, 2013, which is not referenced in this section of text) found the RI to be 1.625 + i0.038 for PSL between 442-472 nm. The authors should include their earlier measurement in the interpolation plot, and discuss any difference with the present result. (A related minor point from line 14: "interpolation of their data". Whose data? - Nikolov & Ivanov? Should the interpolation not also include all the available data?). Earlier, 2995 line 2 describes PSL as "non-absorbing", yet the previous reports and one of the two fits from this work show RI has a small absorbing part. Are we now reaching consensus that PSL absorbs weakly?

PSL is commonly considered as non-absorbing particles (Petzold et al. 2013, Washenfelder et al. 2013).

We have rewritten this paragraph:

Despite a number of studies previously performed, the differences between the retrieved RI values still span a range of about 5% in the visible spectral region which is mainly due to the experimental difficulty in particulate measurements, in particular due to sample to sample differences depending on the nature of the preparation (Miles et al. 2010).

Washenfelder et al. (2013) reported a RI value of $m = 1.633 + i0.005$ at $\lambda = 420$ nm. Chartier and Greenslade (2012) provided a value of $m = 1.72 + i0.005$ at $\lambda = 355$ nm, Rudich and co-workers (Abo Rizip et al. 2007; Lang-Yona et al. 2009), Bluvshtein et al. (2012) found a value of $m = 1.597 + i0.005$ at $\lambda = 532$ nm. Miles et al. (2010) published a value of $m = 1.627 + i0.0005$ at $\lambda = 560$ nm. Nikolov and Ivanov (2000) reported a value of $m = 1.617 + i0$ at $\lambda = 436$ nm and $m = 1.606 + i0$ at $\lambda = 486$ nm. Our results of $m = 1.679 + i0.015$ (retrieved from the scattering channel) and $m = 1.674 + i0$ (retrieved from the extinction channel) obtained at $\lambda = 470$ nm agree with the reported RI values as shown in Fig. 9. These values are a little larger than our previous result of $m = 1.625 + i0.038$, which was probably caused by the large inner volume of the albedometer, hence longer residual time and larger conglomeration effects on small diameter particles. The larger particle loss leads to under-estimation of the particle number concentration, and over-estimation of the extinction and scattering cross-sections. In comparison with the data of Nikolov and Ivanov after interpolation to the same wavelength ($m = 1.61 + i0$), the difference between the RI values is about 4 %, within the tolerance of the instrumental accuracy (4% for scattering and 5% for extinction measurements, 3% for particle concentration measurement), which confirmed that the used calibration method for the determination of the cavity mirror reflectivity $R(\lambda)$, the scattering parameter K', and the parameter R_L (determined by calibration too) was suitable for the aerosol optical properties measurement.
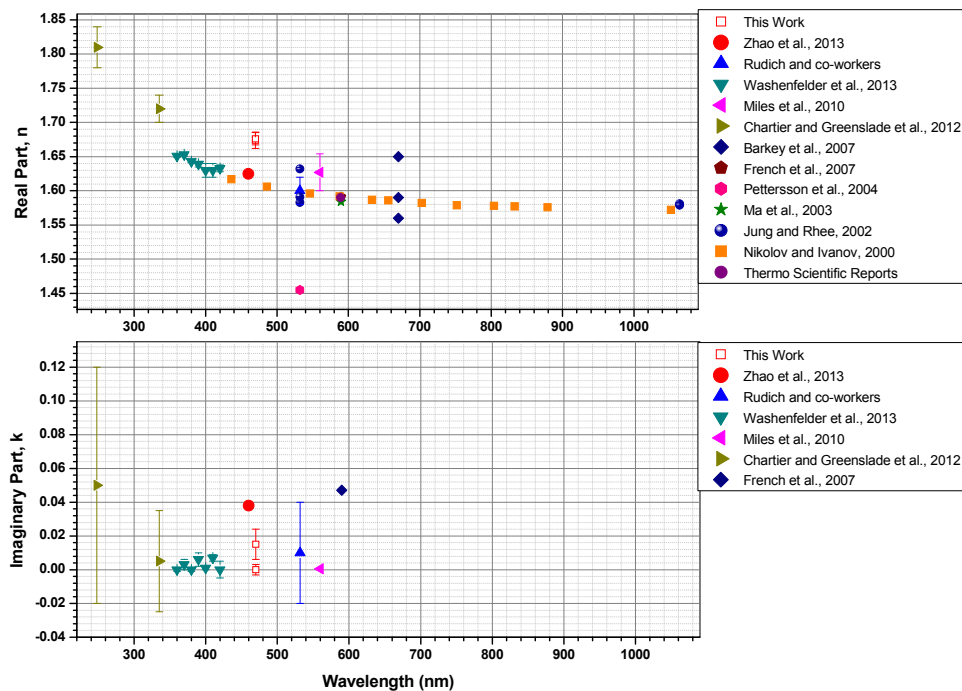
Survey of the measured value of real and imaginary part of the refractive index versus wavelength for PSL.

References:

Abo Riziq, A., Erlick, C., Dinar, E., and Rudich, Y.: Optical properties of absorbing and non-absorbing aerosols retrieved by cavity ring down (CRD) spectroscopy, Atmos. Chem. Phys., 7, 1523–1536, doi:10.5194/acp-7-1523-2007, 2007.

Barkey, B., Paulson, S. E., and Chung, A.: Genetic Algorithm Inversion of Dual Polarization Polar Nephelometer Data to Determine Aerosol Refractive Index, Aerosol Sci. Tech., 41, 751-760, 2007.

Chartier, R. T. and Greenslade, M. E.: Initial investigation of the wavelength dependence of optical properties measured with a new multi-pass Aerosol Extinction Differential Optical Absorption Spectrometer (AE-DOAS), Atmos. Meas. Tech., 5, 709–721, doi:710.5194/amt-5195-5709-2012, 2012.

French, R. H., Winey, K. I., Yang, M. K., and Qiu, W. M.: Optical properties and van der Waals-London dispersion interactions of polystyrene determined by vacuum ultraviolet spectroscopy and spectroscopic ellipsometry, Aust. J. Chem., 60, 251–263, 2007.

Jung, C. and Rhee, B.K.: Simultaneous determination of thickness and optical constants of polymer thin film by analyzing transmittance, Appl. Opt. 41, 3861-3865, 2002.

Lang-Yona, M., Rudich, Y., Segre, E., Dinar, E., and Abo-Riziq, A.: Complex refractive indices of aerosols retrieved by continuous wave-Cavity Ring Down Aerosol Spectrometer, Anal. Chem.,81, 1762–1769, 2009.

Ma, X. Y., Lu, J. Q., Brock, R. S., Jacobs, K. M., Yang, P., and Hu,X. H.: Determination of

complex refractive index of polystyrene microspheres from 370 to 1610 nm, Phys. Med. Biol., 48, 4165–4172, 2003.

Miles, R. E. H., Rudic, S., Orr-Ewing, A. J., and Reid, J. P.: Influence of uncertainties in the diameter and refractive index of calibration polystyrene beads on the retrieval of aerosol optical properties using Cavity Ring Down Spectroscopy, J. Phys. Chem. A, 114, 7077–7084, 2010.

Nikolov, I. D. and Ivanov, C. D.: Optical plastic refractive measurements in the visible and the near-infrared regions, Appl. Optics, 39, 2067–2070, 2000.

Pettersson, A., Lovejoy, E. R., Brock, C. A., Brown, S. S., and Ravishankara, A. R.: Measurement of aerosol optical extinction at 532 nm with pulsed cavity ring down spectroscopy, J. Aerosol Sci., 35, 995–1011, 2004.

[6] 2998 line 4 onwards and Fig 9. The logic behind trying two different wavelength windows to retrieve NO$_2$ concentrations was not explained; the conclusions to be drawn from Fig 9 weren't explained either. The DOAS literature contains many investigations into establishing the optimum wavelength window for retrieving molecular absorption signals; there are analogous tests for broadband CE systems, for example, Chapter 3 of "Cavity Ring-down Spectroscopy: Techniques and Applications", Berden & Engeln (editors) Wiley 2009. The net result is that NO$_2$ concentrations retrieved from long and short fitting windows ought to be the same (although the uncertainty on the fitted NO$_2$ concentration will change depending on the data quality included in the fitting windows). That different NO$_2$ concentrations were produced in Fig 9(a) suggests an instrumental or spectral fitting issue, potentially with determining the wavelength dependence of the mirror reflectivity, or a subtle change has happened in the LED's output, or the polynomial used in the DOAS fit doesn't completely account for the aerosol's extinction thus generating an interference for NO$_2$. The authors need to be clear about their reasons for showing Fig 9 and the conclusions to be drawn.

The reasons for showing Fig. 7 and 9 were that :
We would like to show how IBBCEAS method could provide a useful tool for simultaneous and selective quantitative measurements of both aerosol extinction and trace gas concentrations in ambient air.

Why did we use different wavelength windows to retrieve NO$_2$ concentration?
At first, we did the fit to the data from the full spectral window for retrieval of both NO$_2$ concentration and aerosol extinction coefficient. We found that the retrieved NO$_2$ concentration was larger than the data from the NOx analyzer under high aerosol extinction condition (as shown in Fig. 9a).

This was maybe caused by the big oscillation-like in baseline at λ>475 nm and the absorption structure of aerosol around 465 to 470 nm, which affects the fit quality. The worse data quality related to the oscillation in baseline at λ>475 nm is due to the operation of the Albedometer on the edge of the cavity bandwidth. In addition, the polynomial used in the DOAS fit did not completely account for the aerosol's extinction (aerosol absorption feature was observed around 465 to 470 nm), as shown in Fig. 7(a). Using an appropriate spectral region, the good data retrieval is obtained (as shown in Fig. 7(b)). The fitted NO_2 concentrations with appropriate spectral region agreed well with the NOx analyzer measurement (as shown in Fig. 9(a)).
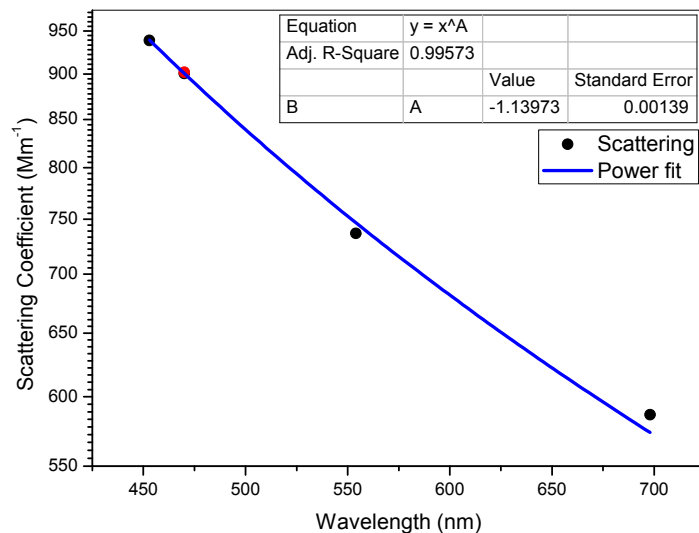
We have made following changes in the text :
2997 line 17-20 : Under higher aerosol loading condition (Fig. 7(a)), the detection sensitivity deteriorated and the NO_2 concentration retrieved from the full window was interfered by the big oscillation-like in baseline at λ>475 nm due to the cavity bandwidth edge and the incomplete account of the aerosol's extinction by the polynomial fit (ambient aerosol absorption structure was observed around 465 to 470 nm). This structure was not observed under lower aerosol loading condition (Fig. 7(c)).

2998 line 3-7 : An enlarged drawing of the NO_2 measurement comparison in two selected periods (10:00–15:00 LT on 18 April for high aerosol load condition and 6:00–14:00 LT on 19 April for low aerosol loading) is shown in Fig. 9. NO_2 concentrations retrieved from different fit windows are also shown in the figure. An appropriate choice of spectral region is very important for accurate data retrieval (Fig. 7(b), (d)).

[7] 2998 lines 10-20. The TSI nephelometer's 554 nm channel was not used to calculate the Angstrom exponent – why not? Surely an extra data point would allow the Angstrom exponent to be better constrained.

Instead of using the 554 nm channel to calculate the Ångström exponent, we determined Ångström exponent using scattering coefficients at two wavelength (as reported in Refs. Anderson and Ogren, 1998; Massoli et al., 2009) in order to well constrained the Ångström exponent. We used 3 data (black) to determine the Ångström exponent curve (fit) and then deduced the scattering coefficient at 470 nm from the fitted Ångström exponent curve, as shown in the follow figure. The deduced scattering coefficient at 470 nm is 900.6 Mm^{-1} in comparison with the value of 902.0 Mm^{-1} (red dot) calculated with Eq. (6) (page 2998, line 17). The difference was smaller than 0.2%.

The plot shows Scattering Coefficient (Mm^{-1}) versus Wavelength (nm) with a power fit.

Equation	y = x^A			
Adj. R-Square	0.99573			
			Value	Standard Error
B	A		-1.13973	0.00139

- ● Scattering
- — Power fit

References:

Anderson, T. L. and Ogren, J. A.: Determining aerosol radiative properties using the TSI 3563 integrating nephelometer, Aerosol Sci. Tech., 29, 57–69, 1998.

Massoli, P., Murphy, D. M., Lack, D. A., Baynard, T., Brock, C. A., and Lovejoy, E. R.: Uncertainty in light scattering measurements by TSI nephelometer: results from laboratory studies and implications for ambient measurements, Aerosol Sci. Tech., 43, 1064–1074, 2009.

[8] 3019 Ambient aerosol Fig 11(a). The albedometer's scattering measurements are consistently higher than the nephelometer if the extinction is large, but are consistently below the nephelometer if the extinction is small – why is this? This effect causes the correlation plot's gradient in Fig 11b (=1.126) to depart from the idea value of 1.00. Actually, the correlation plot looks slightly curved to me.

This effect was probably due to particle loss. Fine particle loss due to conglomeration effects was larger in our system (as shown in following figure). Under small extinction condition, fine particles are dominant which leads to a underestimation of scattering and extinction coefficients. Appropriate choice of the flow rate could further minimize the particle loss (von der Weiden et al. 2009). Under large extinction condition, large diameter particles are dominated. The truncation error of the TSI 3563 nephelometer caused the under-estimation of the scattering coefficient.

We investigated particle loss vs. their size. The laboratory generated NaCl particles were used for the evaluation. The particle loss was determined with two CPCs (a CPC 3775 installed at the entrance of the cavity and a CPC 3776 at the exit). The particle loss was determined by

the difference in particle concentration measured by these two CPCs, respectively, after considering the diluting effect of the purging gas. For particle diameter larger than 300 nm, the particle loss could be ignored.
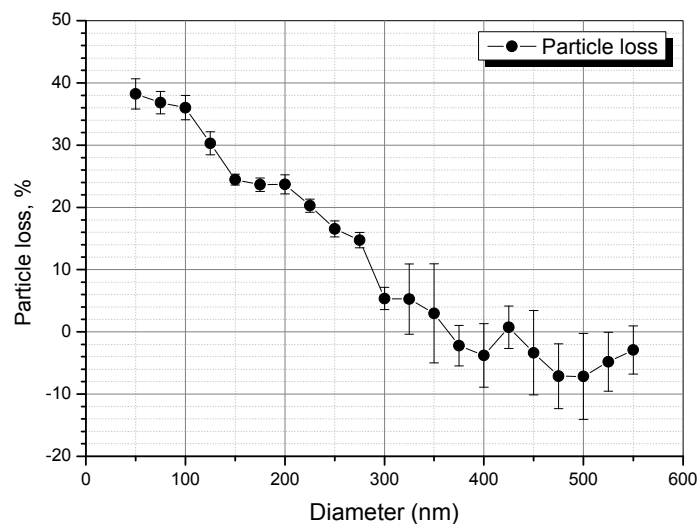
Reference:
von der Weiden, S.-L., Drewnick, F., and Borrmann, S.: Particle Loss Calculator – a new software tool for the assessment of the performance of aerosol inlet systems, Atmos. Meas. Tech., 2, 479-494, doi:10.5194/amt-2-479-2009, 2009.

[9] I agree with Reviewer #1 that, although improved, language issues remain. There are numerous examples of missing "the" or "a", problems with singular versus plural, and missing punctuation. [Some examples: p2983 line 9 "Since [the] aerosol extinction is the sum of [the] absorption and scattering coefficients...". Line 24 "...multiple scattering by [the] filter medium, and [the] angular distribution of [the] scattered light". Line 28 "For... highly light[-]absorbing[,] organic aerosol loading[s], the bias in filter-based light absorption measurement[s] may be larger than 100%." Page 2984 line 13..."such as [the] TSI 3563 integrating nephelometer...". Lines 15-20 "underestimation of scattering coefficient[s]... scattering measurement[s] using a nephelometer... Correction factor[s] for the truncation errors..."]. The authors sometimes form a sentence's subject by grouping several nouns together, and this makes the sentence's meaning unclear or ambiguous. [For example 2984 line 28: "Optical extinction cell approach, limited by the detection sensitivity, is of practical use only in...." would read better as "Optical extinction measurements made in [single pass?] cells are limited by the detection sensitivity and are of practical use only in..."]. The errors are too many to expect a reviewer to list them. The manuscript requires comprehensive proof-reading by the authors and, I suggest, careful checking by the editorial office.

We tried our best to carefully check the English usage.

Technical comments & corrections:

[10] 2982 line 11: ...”averaged [not integrated] value over a narrow bandwidth”.

DONE.

[11] Line 13: ... “averaged scattering “. Unclear: averaged over what? – the wavelength range stated in the previous sentence?

... averaged over the spectral region of 465–474 nm ...

[12] Line 18: Be clear that the good agreement with RI in previous papers applies to the laboratory tests on PSL in the previous sentence.

DONE. The retrieved refractive index (RI) <u>of the PSL particles</u> from the measured scattering and extinction efficiencies agreed well with the values reported in the previously published papers.

[13] Line 23 ”aerosol scattering coefficient[s] and NO₂ trace [gas] concentration[s]”

DONE.

[14] 2984 line 8. Move the Langridge et al citation from line 11 to line 8 where it is first referenced. Explain why they concluded that the PAS technique is not well-suited to measurements at high RH.

We have added the following statement on Page 2984 line 10 onwards:

"They concluded that the PAS is not a technique well suited for the measurement of aerosol absorption at high RH due to the impact of water evaporation on PAS signal, ...".

[15] 2985 line 12 "this method still involves"...

DONE.

[16] Line 25. Dial reference is 2010, not 2012.

DONE.

[17] 2986 line 25: use of the single citation to Zhao et al 2013 (wrongly!) implies that Zhao were the first to use DOAS to separate aerosol extinction from extinction by molecular absorbers in a gas sample. Either cite the earlier work from other groups, or delete the Zhao reference and refer readers back to the works cited a few lines above.

Following references were added here :

Berden, G. and Engeln, R. (Eds.): Cavity Ring-Down Spectroscopy: Techniques and Applications, Wiley-Blackwell, 2009.

Fayt, C., De Smedt, I., Letocart, V., Merlaud, A., Pinardi, G., and Van Roozendael, M. : QDOAS Software user manual:http://uv-vis.aeronomie.be/software/QDOAS/index.php (last access : 14 May 2012), 2011.

Gherman, T., Venables, D.S., Vaughan, S., Orphal, J., and Ruth, A.A.: Incoherent broadband cavity-enhanced absorption spectroscopy in the near-Ultraviolet : application to HONO and NO2, Environ. Sci. Technol., 42, 890-895, 2008.

Kraus, S. and Geyer, A.: DOASIS Jscript programming description. http://www.iup.uni-heidelberg.de/bugtracker/projects/doasis (last access : 12 May 2012), 2001.

Platt, U., Meinen, J., Pöhler, D., and Leisner, T.: Broadband Cavity Enhanced Differential Optical Absorption Spectroscopy (CE-DOAS) – applicability and corrections, Atmos. Meas. Tech., 2, 713-723, doi:10.5194/amt-2-713-2009, 2009.

Platt, U. and Stutz, J.: Differential Optical absorption spectroscopy : principles and applications, Springer, 2008.

Thalman, R. and Volkamer, R.: Inherent calibration of a blue LED-CE-DOAS instrument to measure iodine oxide, glyoxal, methyl glyoxal, nitrogen dioxide, water vapour and aerosol extinction in open cavity mode, Atmos. Meas. Tech., 3, 1797-1814, doi:10.5194/amt-3-1797-2010, 2010.

[18] 2987 lines 14,19 & 22 give dimensions in cm units, whereas the dimensions are shown

in mm units on Fig 1. Please make consistent.

DONE. All the dimensions were changed to mm units.

[19] Line 23. In addition to the 1.5 L min-1 flow rate, please provide the residence time of the gas sample inside the instrument. This also relates to point #2 raised by Reviewer #1 and my point [4b].

The residence time was about 200 s for the present albedometer (with a flow rate of 1.5 L min^{-1} and a total volume of ~ 1.9 L of the instrument including the truncation reduction tubes).

[20] 2988 line 1. "spectral resolution 0.4 nm". Is this FWHM? How was it measured? Is this resolution the same over the spectrometer's full 412-487nm range?

Yes, it is FWHM. It was measured with a low-pressure mercury lamp. The resolution is the same over the spectrometer's full 412 - 487 nm range.

[21] 2989 line 1. What is "r"? Where is this defined?

r is the radius of the hole presented at each pole of the hemisphere for the passage of the probe light beam.

[22] 2990 line 1. RL is defined here as the ratio of the total length of the cavity to the length of the cavity occupied by the sample. Yet 2991 line 12 states RL was measured using a 42 ppbv NO$_2$ sample. Which is correct?

Yes, R_L is the ratio of the total cavity length to the length of the cavity occupied by the sample. R_L could be determined using an absorber with known extinction (such as a dilute concentration of NO_2), or geometrically measured based on the assumption that aerosols follow the gas flow path and are not present in the purge volumes (Washenfelder et al., 2013). In this work, R_L was determined from the absorption measurement of 42 ppbv NO_2 with and without mirror purges.

[23] Line 6. "Broadband wavelength measurement of extinction coefficient by IBBCEAS provides..." needs rephrasing.

Broadband extinction measurement with IBBCEAS provides a robust method for simultaneous and selective quantitative measurement of both aerosol extinction and absorbing trace gases concentrations using a single instrument.

[24] 2991 line 2. Add Moosmuller et al (Aerosol Sci Tech 2005) to the reference list. This group was one of the first to use Rayleigh scattering in different gases to determine cavity mirror reflectivity.

DONE.

[25] Line 14. "The aerosol scattering coefficient, alpha_scat, is proportional..." (not the "scattered light intensity").

DONE.

[26] Line 15. Needs a clear statement about whether the CCD spectrometer used to measure I_trans is the same spectrometer used in the IBBCEAS measurements.

DONE. The CCD spectrometer used to measure the transmitted intensity is the same as used in the IBBCEAS measurements.

[27] Line 20. The unfortunate positioning of the Strawa 2003 and Thompson 2008 citations at the end of this sentence implies those works impacted only on correcting a minor truncation error. In fact, Strawa et al produced equation 3, and Thompson et al then added the K' term.

We have moved this sentence (line 16-20) to line 25.

[28] 2993 line 6. Explain how the long-term drift value (smaller than 2 Mm^{-1}) was quantified from Fig 5. (Don't presume the reader should do this him/herself).

Longer-term drift of the instrument was observed and smaller than 2 Mm^{-1} (as shown in Fig. 5).

[29] 2997 line 12-13. Perhaps it's just clumsy phrasing, but "[The] large fit error observed around 475-481nm" is caused by noise in the data, not a problem with the spectral fit.

We agree with the reviewer's comment and our statement was in the same sense:
"The large fit error observed around 475–481 nm was due to the low signal-to-noise ratio (SNR) data related to low light transmission from the cavity."

[30] 2998 line 3 "tolerance of the NOx detection sensitivity (1 ppbv)" – for which instrument?

For the used NOx analyzer.

[31] Line 14 "zero adjusting" – of what?

Zero adjusting of baseline for the scattering coefficient measurements ...

[32] 2999 line 6 onwards. The authors note that aerosol extinction measurements by cavity methods require very stable light sources, and that LEDs are therefore a good choice. Either here in the Conclusions section or on 2986 line 16 where they cite Fiedler et al 2003, the authors ought also to cite Ball et al (Chem Phys Lett 2004) who were the first to demonstrate cavity enhanced absorption using an LED light source. The Fiedler work used an arc lamp which, according to this Conclusions section, the authors consider is less favourable than an LED.

We have add the reference of Ball et al., 2004 to Page 2986 line 18 and Page 2999 line 6.

Page 2999 line 6 :
The cavity enhanced methods require very stable light sources, and LED is a promising new type of light source, with long life time and low energy consumption. It is more compact than commonly used broadband arc lamps.

Reference:

Ball, S.M., Langridge, J.M., and Jones, R.L.: Broadband cavity enhanced absorption spectroscopy using light emitting diodes, Chem. Phys. Lett., 398, 68-74, 2004.

[33] 3012 Fig 4. The meanings of the "X offset" and "Y offset" annotations are unclear. How these offsets are used in the analysis needs to be clearly explained in the text. Please check the equation of the best fit line – are the exponents correct?

The annotation of "X offset" was modified with : X offset : Scattering intensity by internal surfaces. The annotation of "Y offset" was deleted, since Y axis is the absolute Rayleigh scattering coefficients of each gas.

We made mistake in the unit here. We fitted the data in the unit of "cm^{-1}", but the figure showed the results in [Mm^{-1}]. The equation of the best fit line should be written as : $y = -186.5 (\pm 4.2) + 1.52 (\pm 0.03) \times 10^7 \, x$.

[34] 3014 Fig 6. It is very difficult to discern the different symbol shapes in panel (a). Also check the value of the best fit line's intercept = 4.23x10^-9 Mm-1 doesn't look right.

DONE. The symbol shapes in panel (a) were modified.
We made mistake here. It should be 4.23×10^{-9} cm^{-1} (0.423 Mm^{-1}). The fit result should be : $y = 0.42 (\pm 0.76) + 0.96 (\pm 0.03) \, x$.

[35] 3015 Fig 7. Provide the +/- uncertainties on the fitted NO_2 concentration. Also provide the water concentrations from fitting spectra III of each panel.

DONE.

[36] 3016 Fig 8. Are the relative humidity measurements in panel (a) from a commercial instrument (what type?) or are they measured by IBBCEAS using the fitted water absorptions (in Fig 7)? Which wavelength window was used for the NO_2 retrievals in panel (b)? There is a lot of scatter in the SSA data points, presumably because the integration time of the measurement is quite short – these data will probably look better if they are shown as averages over a longer integration time. Are SSA values around 0.9 reasonable for ambient aerosol? The figure caption attributes differences between the scattering measured by the albedometer and the TSI instrument to larger truncation errors in the TSI instrument. This

The relative humidity was measured using the internal relative humidity sensor of the TSI 3563 integrating nephelometer.

The narrow fit window (444-467 nm) was used for the NO_2 retrievals in panel (b).

Yes, the scatter in the SSA data points was caused by a short integration time. We would like to show high temporal resolution measurements of the SSA parameter.

The SSA value around 0.9 was reasonable. For instance, the SSA values of ambient air measured by Thompson et al. (2008) and Dial et al. (2010) were about 0.9. However, in biomass burning seasons, the ambient air was more absorbing and our measurement results showed a SSA value as low as 0.75. The developed albedometer will be further tested via intercomparison with commercial measurement instruments.

We did not make correction for the scattering data from the TSI nephelometer. For ambient air measurements, this correction needs the following information: measured Ångström exponent, aerosol size distribution and complex refractive index. Unfortunately, we did not have the information on size distribution and the RI in this study.

As shown in Fig. 3, for 1 μm diameter particles, the truncated fractions of total scattering was about 10% with a truncation angle of 7º. This value was increased to 20% for particles of diameter of 1.5 μm. The differences (with a slope of 1.13) between scattering coefficients measured by the albedometer and the TSI instrument was mainly caused by the larger truncation errors in the TSI instrument.