Renmin Yuan et al.,

A new method for measuring the imaginary part of the atmospheric refractive index structure parameter in the urban surface layer

ACPD-14-21285-2014

The authors appreciate the three referees for their constructive comments and suggestions. The manuscript has been revised accordingly. Our point-by-point responses to these comments are provided below. The comments of the referees are printed in black and our responses following each comment in blue.

Answer to the first anonymous referee's comments:

The atmospheric refractive index structure parameter (ARISP) is studied from both theoretical and experimental perspectives. The real part of ARISP is closely related to the strength of atmospheric turbulence whereas its imaginary part determines the absorption for radiative transfer. Because of the importance, the findings reported in this paper should be useful contributions to atmospheric physics literature.

Overall, the manuscript can be easily understood. But the manuscript in its present form needs some mandatory major revisions before it is accepted for publications.

We thank the referee for his/her positive comments.

Major issues:

1) The use of English in the manuscript needs to be substantially improved. There are a number of grammatical errors and awkward phrases or sentences in the manuscript.

We've tried our best to improve the English writing in the revised manuscript, but also the revised version of the manuscript was reviewed by a native English speaker.

2) The originality of the theoretical development in the manuscript is ambiguous. Specifically, in Section 2 "Theory" it is not clear which part is the authors' original contribution. For example, it seems that the formulas and relevant explanations in Sections 2.1 and 2.2 are taken from the literature. If this is true, please delete these sections and cite the original papers.

We have deleted Section 2.1and 2.2, and keep two sub-sections Section 2.3 and 2.4, one is for the relationship between log-intensity variance and the imaginary part of the ARISP, and the other for the relationship between the structure function and the imaginary part of the ARISP. We deleted some existing equations. However, in order to ensure the continuity of the content and the readability of the article, we kept some existing equations with references. This paper is to illustrate the process of decomposing the signals measured by a large-aperture scintillometer (LAS) into high- and low-frequency parts, which are due to the real and imaginary parts of the ARISP respectively. The contribution of the imaginary part of ARISP to the received light intensity fluctuation is figured out. Then we obtained the expressions for the variance and structure function of log-intensity caused by the imaginary part of ARISP. Based on these relationships, a practical expression for the imaginary part of ARISP is deduced and can be applied to measurement data. Compared to the method proposed by Nieveen (Nieveen et al., 1998) for estimating the refractive index structure parameter of imaginary part, this method is more objective, and can give the estimation of the outer length scale of turbulence.

We have rewritten and rearranged Section 2. Please see Page 4-10.

3) For Sections 2.3 and 2.4, trivial technical details seem unnecessary. To enhance the clarity of the manuscript, Sections 2.3 and 2.4 should be rewritten.

We have rewritten Sections 2.3 and 2.4

Please see Pages 4-10.

4) In the conclusions, the value of the findings of this study should be clearly stated. The current statement about the value of this study is too generic. It should be more specific.

We have rewritten this part. In the revised manuscript, the significance of the imaginary part of ARISP is emphasized, and the potential applications of LAS measurement on aerosols are discussed. In the discussions, the differences between long-path absorption spectroscopy and the absorption measurement are also discussed due to their common ground in aerosol monitoring.

Please see Pages 15-17.

<u>Answer to the second anonymous referee's comments</u>:

Major Comments:
The most innovative parts of this paper are: 1) the derivation of a rather simpler expression of the imaginary part of the atmospheric refractive index structure parameter (ARISP) (also, the equation to calculate transverse wind velocity), based on results of some original papers; 2) the aerosol concentration, e.g. in an urban area, could be obtained accordingly with ordinary LAS (Large Aperture Scintillometer) observations. This would extend the LAS usages in some cities to environmental monitoring. Generally the manuscript was properly written, with clear theoretical derivations, as well as carefully designed/ operated experiments. The paper is appropriate to the journal Atmospheric Chemistry and Physics (ACP). However, the present manuscript still needs some revisions before it can be accepted for publication.

We thank the referee for his/her positive comments.

We've tried our best to improve the English writing in the revised manuscript, but also the revised version of the manuscript was reviewed by a native English speaker.

Comments for revision:
1) Some inconsistencies in, for instance, 'Conclusions' and 'Abstract'. While the 'Conclusions' part emphases aerosol effects on the imaginary part of the ARISP, the 'Abstract' also stress the effect of 'trace gases' (with selected wavelength of LAS). The scintillometer used by the authors (with wavelength 620 nm), as well as the most popular LAS's used in recent decades (with light wavelength about 850 nm to 880 nm), are all working in the atmospheric windows. These would be improper for the assessment of trace gases.

Indeed, there are some inconsistencies in 'Conclusions' and 'Abstract'. The study offers a theoretical framework, which is to measure the imaginary part of ARISP by a large-aperture scintillometer. We choose the wave band of atmospheric window with a wavelength of 620 nm, and then the imaginary part of ARISP reflects the characteristics of aerosols. This is the goal of our current work. So we mentioned this point in both the abstract and the conclusion. The theoretical framework of the paper can also be applied to the assessment of trace gas. If we choose a wavelength for absorbing a certain trace gas, then the measured imaginary part of ARISP would reflect the characteristics of this trace gas. For example, a wave with a wavelength of 940 nm can be used to monitor water vapor.

In the revised manuscript, we deleted the part about the assessment of trace gas in the abstract, and only talked about the application to the assessment of trace gas in the conclusions as a discussion.

Please see Lines 29-29 Page1, Lines 12-15 Page 17.

2) The theoretical derivation, particularly for section 2.3 and 2.4, is a little lengthy.

We have rewritten and rearranged Section 2. In the revised manuscript, just two sub-sections are kept; one is for the relationship between log-intensity variance and the imaginary part of the ARISP, and the other for the relationship between the structure function and the imaginary part of the ARISP.

Please see Section 2 in Pages 4-10.

Some formulas particularly unused symbols are better to be deleted (e.g. the 4F*(k,L) term in Eq. (1)). While the symbols used should be described clearly.

We revised the equations and gave clear descriptions for the symbols.

Please see Line 5 Pages 4.

3) In several places it mentioned that 'the LAS observations are performed at the height of 24.5 m'. However, the scintillometer used is actually a slant path (one side 18.5 m, another 24.5 m). An effective path height is better to be used.

We conducted two experiments. During the experiments, the transmitting and receiving parts are nearly at the same height, namely, the light beam is almost horizontal. In the first experiment, the transmitting and receiving terminals are both at the 10^{th} floor, which is 18.5m higher than the reference plane to validate the LAS with conventional measurements. In the second experiment, the transmitting and receiving terminals are both at the 12^{th} floor, which is 24.5m higher than the reference plane. The purpose of the first experiments is to ensure the reliability of our instrument and compare the results with the other models. In the revised version, the results of the first experiment are deleted according to the referee's comments.

4) The English writing in this manuscript need to be carefully revised. Following are only a few examples: Page 21286, line 22: 'trace gas' better to be 'atmospheric trace gases'; Page 21289, line 14-22: The symbols used in Eq. 1 need to be described precisely, e.g., k is the wave number of the light wave used; z is the position along the propagation path; etc.

Thank you for some specific revising comments. We've already made revisions on the syntaxes that are confusing, such as the light wave number (as η) and turbulence wavenumber (κ). We now use x to represent the horizontal transmission position.

We've tried our best to improve the English writing in the revised manuscript, but also the revised version of the manuscript was reviewed by a native English speaker.

Page 21289, line 23-24: 'temperature' is also 'conservative'? 'passive scalars with their sources at the surface'?

It is often assumed that if there is not source or sink of heat, and no change of phase happens except at the surface, temperature can be considered as conservative. But also, temperature can be taken as passive.

The "passive" means that, turbulence exerts force on the aerosols (or temperature) and no force from the aerosols (or temperature) is done to the turbulence.

There is something confused with the statement 'passive scalars with their sources at the surface', and we modified the statement.

The wavelength we used in our experiments is at the atmospheric window region, and the absorption was caused by aerosols. We assume the source of aerosols is on the ground, and there's no production of new aerosol particles. Aerosol observations in the urban area also show that the aerosol particles and the molecule of gas follow the same law of scalars such as temperature and water vapor density, which is that the aerosol number density fluctuation spectrum follows the "-5/3" law and the co-spectrum of aerosol number density and wind speed follow the "-4/3" law [Martensson et al., 2006; Vogt et al., 2011]. Therefore, we can regard the aerosols as a conservative and passive scalar.

Please see Lines 16-24, Page 4.

Page 21290, line 15: 'by the real part', of what? Page 21297, line 2: 'data process' should be 'data processing'.

For the question “Page 21290, line 15: ‘by the real part’, of what?”, it is by the real part of the ARISP. We modified the text.

Please see Line 20 Page 5, Line 5 Page 10.

Page 21302, line 27-28: On the date and time, better to be‘...at09:00LT,15 Jan 2014, and at 12:00 LT the next day’.

According to the comments, we have rewritten the date and time as the format.

New references:

Martensson, E. M., E. D. Nilsson, G. Buzorius, and C. Johansson (2006), Eddy covariance measurements and parameterisation of traffic related particle emissions in an urban environment, *Atmos. Chem. Phys.*, *6*, 769-785.

Vogt, M., E. D. Nilsson, L. Ahlm, E. M. Martensson, and C. Johansson (2011), Seasonal and diurnal cycles of 0.25-2.5 mu m aerosol fluxes over urban Stockholm, Sweden, *Tellus Series B-Chemical And Physical Meteorology*, *63*(5), 935-951, doi:10.1111/j.1600-0889.2011.00551.x.

Answer to the third anonymous referee’s comments:

Overview:

This paper presents possibly useful advances in the application of large-aperture scintillometry (LAS) to determine refractive index structure parameters. The authors claim to present a new method to separate the contributions of atmospheric absorption (the imaginary part of the refractive index structure parameter) and atmospheric scintillation (the real part of the refractive index structure parameter). It appears that there is some new theoretical development, amongst a great deal of already published theory; the authors need to clearly distinguish their new derivations from earlier published work.

We thank the referee for his/her positive comments.

We made revisions on the theoretical part of the article, deleted the some existing equations. In the revised manuscript, we keep two sub-sections for theory Section 2.3 and 2.4; one is for the relationship between log-intensity variance and the imaginary part of the ARISP, and the other for the relationship between the structure function and the imaginary part of the ARISP. We deleted some existing equations.

This paper is to illustrate the process of decomposing the signals measured by a large-aperture scintillometer into high- and low- frequency parts, which are attributed to the real and imaginary parts of the ARISP respectively. The contribution of the imaginary part of ARISP to the received light intensity fluctuation is figured out. Then we obtained the expressions for the variance and structure function of log-intensity caused by the imaginary part of ARISP. Based on these relationships, a practical expression for the imaginary part of ARISP is deduced and can be applied to measurement data. Compared to the method proposed by Nieveen (Nieveen et al., 1998) for estimating the refractive index structure constant of imaginary part, this method is more objective, and can give the estimation of the outer length scale of turbulence.

We have rewritten and rearranged Section 2. Please see Page 4-10.

This is a potentially improved method to reject the unwanted influence of absorption on the scintillometer measurement, without relying on spectral separation (analyzing the spectrum of scintillation), enabling more accurate measurement of the real part of the refractive index structure parameter, and therefore better estimation of the sensible heat flux. Furthermore, this opens the new opportunity to measure the spatial variation of atmospheric absorption along the long measurement path, which may have applications in aerosol monitoring, and potentially trace gas monitoring. The paper could be improved by further discussion of these applications, and the subtle differences between long-path absorption spectroscopy and the absorption measurement presented here.

In the revised manuscript, we presented discussions on the potential applications of this method on the measurement for the flux of aerosols. The paper proposed a theoretical framework, which may have potential ability to obtain the flux by measuring the imaginary part of ARISP using a large-aperture scintillometer. If the wavelength used is not absorbed by gases, then it reflects the characteristic of aerosol. If the wavelength used is within a certain trace gas' absorption interval, then it reflects the characteristics of this trace gas.

The revised manuscript discussed the difference between the method proposed in this paper and long path absorption spectroscopy. As far as we know, the long path absorption spectroscopy can measure attenuation of narrow band and obtain gas concentration. The goal of our study is to measure the imaginary part of the ARISP using a LAS, and it is possible to obtain the aerosol flux. The LAS can be used to measure the high-frequency fluctuation of light intensity to obtain some information including atmospheric temperature, crosswind, aerosol and other turbulence characteristics. The advantage of the spectroscopy is to attain the narrow spectral information. But the aerosol exhibits strong wide band absorption in visible region, so, if the spectroscopy can sample the attenuated light at high rate at every narrow band, then more parameters (for example, size, and chemical composition) of aerosols may be retrieved using the current method.

Please see Lines 16-32 Page 16.

The presentation is reasonably clear, but the English needs to be improved in numerous places.

We've tried our best to improve the English writing in the revised manuscript, but also the revised version of the manuscript was reviewed by a native English speaker.

It is unclear from the paper, whether a fundamentally new approach has been derived, or more likely a variation on the already known method of spectral separation for the absorption and scintillation contributions to the measured spectrum. The auto-correlation analysis presented essentially fits two asymptotic lines to the time-delay auto-correlation – I challenge the authors to show how this is fundamentally different to determining the 'corner' frequencies shown in Fig.1, by fitting the dashed and solid lines shown – as described by the authors 'analyzing the spectrum of scintillation' (p.21301).

To my knowledge, there's no literature that clearly decompose the signals measured by large-aperture scintillometer into high and low frequency parts. Absorption was ever discussed on the influence to fast frequency part and it is considered as disturbance and removed (Solignac, 2012). Spectral separation and time-delay auto-correlation are two methods that can be used in separating the signals. The two methods are introduced and applied, and results by the two methods are same. In the revised manuscript, the method of time-delay auto-correlation is deleted in order to avoid repetition.

Based on the decomposition of the signal, the contribution from the imaginary part of ARISP is obtained, and then the expressions for the variance and structure function are deduced. So a new practical expression for imaginary part of ARISP is given.

The method used to determine the 'corner' frequency: linear regression is conducted to the rapid-change part. Practically, the data points from the fifth to the 15th point are fitted for the high frequency part; then the data points from the 400th (this scale is larger than the receiver diameter) to the 10000th is fitted for the low frequency part. The y-coordinate of the intersect point (shown in Fig. 4a in the original manuscript) is the energy of the low frequency part. According to practice, the results by this method are pretty stable.

Solignac, P. A., Brut, A., Selves, J. L., Beteille, J. P., and Gastellu-Etchegorry, J. P.: Attenuating the Absorption Contribution on C-n2 Estimates with a Large-Aperture Scintillometer, Bound-Lay. Meteorol., 143, 261-283, 10.1007/s10546-011-9692-3, 2012.

The authors have not demonstrated improved results over the aforementioned spectral analysis, using the time-delay auto-correlation, and only one 20 min period is analysed by the two separate methods; therefore, it has not been shown that the time-delay auto-correlation method has any advantage. It might be claimed that the measurement of the outer length scale of turbulence is a valuable benefit; however, the experimental performance of this measurement is not properly evaluated (again only one result is quoted, which may be considered to be approximately the measurement height, a 'rule of thumb' approximation of the outer length scale). The authors should estimate the uncertainty of their measurements. Overall, very few data are shown – Fig.4 uses a single 20 min data period, and Fig.6 & Fig. 7 show the same 24 hour period.

Time-delay auto-correlation decomposes the large aperture scintillating signals into high and low frequency parts. This is relatively easy and also able to get a pretty objective and stable value. Certainly, spectral separation can generate the same result. Comparing to the spectral separation, time-delay auto-correlation has no special advantage. We have removed the discussion about time-delay auto-correlation method in the revised manuscript.

As the referee pointed out, using this method can give the value of the outer scale of turbulence. Up until now, there is no study on getting the outer scale of turbulence based on scintillation. We have conducted some experiments to measure the outer scale over the urban underlying surface. The results showed that the outer length scale of turbulence depends on height, stability, etc. Because we only talk about the imaginary part of ARISP in the manuscript, the results for outer scale of turbulence will be discussed in other paper. So we did not present the uncertainty of measurements. In this paper, the outer scale of turbulence is only treated as a needed parameter for calculating the imaginary part of ARISP.

This manuscript gives only the results from one 20 min and two 24 hours' measurements. They are mainly for testing our proposed measuring method. More results will be given in other articles.

The LAS derived crosswind comparison is not really novel and comparison with a single cup anemometer over complex urban terrain, does not provide a scientific quantitative comparison. There is insufficient discussion of the Double-Point temperature fluctuation sensor – what is its response time? As the sensor separation is 0.8 m, it will not be able to measure the same inertial sub-range turbulence measured by the LAS; how is this frequency response mismatch dealt with? Why do this comparison, if only to say that agreement will be limited by the vastly different spatial sampling (was the LAS path length still 960 m?).

Indeed, using a LAS to get the crosswind velocity and the real part of ARIP is not novel. The original manuscript intends to ensure the reliability of our LAS experiments, so we make comparisons between a single cup anemometer and LAS measured wind speed, and the real part of the ARISP measured by DP and LAS. Generally the results are satisfying, and that means our instrument is reliable. In the revised manuscript, this part has been removed.

Please see Section 4 in Pages 13-14.

The Double-Point temperature fluctuation sensor is made by inserting two tungsten wires into an electric bridge to measure the temperature difference. Each of the tungsten wires has diameter of 5um, length of 0.8 cm, and response time of approximately 0.02s. The distance of the two arms is 0.8m, with the length within the inertial sub-range of the turbulence. In the additional comparison experiment, the LAS path length was still 960 m. The measurement of LAS is sensitive to the turbulent eddies with scale of transmitting and receiving aperture diameters; namely, it's sensitive to the eddy with 0.18m (The diameters of the lenses for transmitting and receiving are 0.18m). The scale is also within the inertial sub-range of the turbulence. From this perspective, the results from the two methods are comparable. Certainly, the turbulence measured by a LAS is the integral of the whole path, while that measured by DP is from a single position. If the turbulence is distributed uniformly, the two should be consistent. In fact, as the propagating path is not uniform, the results from a single position are different from the integral of the whole path.

Fig.5(a) is a log-log scale plot, and seeing the spread of data, the apparent noise-floor of the LAS and the curve of the data, I do not agree that the comparison is 'very good'.

We agree with the referee's opinion that the agreement of comparison is not 'very good'. In the revised manuscript, we removed the comparison in the revised version because the comparison is not really novel.

The authors ought to be able to measure and state the instrumental noise floor of the LAS (which appears to be rather poor compared to some commercial LAS instruments).

The variance ($\sigma_{lnI,Re}^{2}$) of the noise of LAS we currently built is around 2.0e-06, which is that $C_{lnI,Re}^{2}$ is less than 5e-17m$^{-2/3}$. This is indeed larger than the noise of LAS for commercial instrument（~1e-17m$^{-2/3}$）.

Please see Section 4 in Pages 13-14.

Technical Comments:

1. P.21288 Lines 16-17 please expand as this is important and useful, that the imaginary part of the ARISP contains information on inhomogeneities of the absorptions (contrast with long path spectroscopy).

The measurements of urban aerosol imaginary part carried on different places show that the mean value of urban aerosol imaginary part distributes inhomogeniously in a long period of time and a large space. In order to describe the inhomogeneity, we introduce the imaginary part structure constant of refractive index. The larger this value is, the more uneven the imaginary part of the refractive index distributed, and vice versa. We expanded the statement.

Please see Lines 14-20 Page 3

2. P.21289,L.23-4 These assumptions need careful and critical justification; the absorption media may not be conservative nor passive, and sources may be above the surface (e.g. chimney stacks)? It is unlikely that the absorption and temperature sources will have the same spatial distribution at the surface – how does this affect the application of the theory?

It is often assumed that if there is not source or sink of heat, and no change of phase happens except at the surface, temperature can be considered as conservative. But also, temperature can be taken as passive.

The "passive" means that, turbulence exerts force on the aerosols (or temperature) and no force from the aerosols (or temperature) is done to the turbulence.

The wavelength we used in our experiments is at the atmospheric window region, and the absorption was caused by aerosols. We assume the source of aerosols is on the ground, and there's no production of new aerosol particles. Aerosol observations in the urban area also show that the aerosol particles and the molecule of gas follow the same law of scalars such as temperature and water vapor density, which is that the aerosol number density fluctuation spectrum follows the "-5/3" law and the co-spectrum of aerosol number density and wind speed follow the "-4/3" law [Martensson et al., 2006; Vogt et al., 2011]. Therefore, we can regard the aerosols as a conservative and passive scalar.

It is not required that the absorption and temperature sources will have the same spatial distribution at the surface. As long as the absorption media is conservative and passive, the scalar turbulence rules can be applied to the absorption media.

According to the comments, it should be careful to give some assumption, and in the revised manuscript we added some references.

Please see Lines 16-24, Page 4.

New references:
Martensson, E. M., E. D. Nilsson, G. Buzorius, and C. Johansson (2006), Eddy covariance measurements and parameterisation of traffic related particle emissions in an urban environment, *Atmos. Chem. Phys.*, *6*, 769-785.
Vogt, M., E. D. Nilsson, L. Ahlm, E. M. Martensson, and C. Johansson (2011), Seasonal and diurnal cycles of 0.25-2.5 mu m aerosol fluxes over urban Stockholm, Sweden, *Tellus Series B-Chemical And Physical Meteorology*, *63*(5),

935-951, doi:10.1111/j.1600-0889.2011.00551.x.

3. Justify the assumption of isotropic turbulence in the urban environment (p.21290, L.9-10).

Usually the heights of the transmitting and receiving units are on the 12^{th} floor. The signal measured by large-aperture scintillometer has a larger weight on the middle part of the propagating path which is at the height of about 24.5m over the reference plane, so we can think that it satisfies the isotropy assumption.

Please see Lines 22-24 Page 10.

4. P.21292, L.10, please justify this assumption – it appears that the variances caused by the real and imaginary parts of the ARISP will be highly correlated because of their dependence on atmospheric turbulence and crosswind speed, as shown by Eq.4 and Eq.6?

The real and imaginary parts depend highly on the atmospheric turbulence and crosswind speed, but their contributions on the measured light intensity fluctuation have high and low frequency parts. The fluctuations of high and low frequency are not correlated; namely, they are independent.

Please see Lines 3-8 Page 6.

Editorial Comments

1. P.21286, Line 25 change 'line-sight' to 'line-of-sight'.

   Done.

   Please see line 2 Page 2.

2. L.26 – rephrase, turbulence alone does absorb light.

   Done.

   The scattering of atmospheric turbulence, gas molecules and aerosol particles, as well as the absorption of gas molecules and aerosol particles.

   Please see lines 2-4 Page 2.

3. P.21287, L.6 change to 'intensity in the receiving plane'; change 'a distance' to 'some distance'.

   Done.

   Please see line 9 Page 2 and line 4 Page 2

4. L.11 change 'measure' to 'determine' (since this step is dependent on similarity theory, and is not a direct measurement).

   Done.

5. L.19 change to 'contribution of absorption'.

   Done.

6. P.21288, L.29 change to 'Finally, a brief conclusion is presented'.

   Done.

7. P.21290, L.9 (and elsewhere) change 'isotopic' to 'isotropic'.

   Done.

8. P.21293, L.1 change to 'commonly used'.

   Done.

9. Other numerous minor errors in the English language need to be addressed.

We’ve tried our best to improve the English writing in the revised manuscript, but also the revised version of the manuscript was reviewed by a native English speaker.

Finally, the authors thank the three referees for their constructive comments that help us to greatly improve the clarity and the quality of the manuscript. We sincerely hope our answers can relieve doubts and give a better description of our work.