



Laboratory Comparison of Aerosol Optical Property Measurement Techniques

Paola Massoli^(1,2), Tahllee Baynard^(1,2), Daniel Lack^(1,2), Edward R. Lovejoy⁽²⁾, and A. R. Ravishankara^(2,3)

(1) Cooperative Institute for Research in the Environmental Sciences (CIRES), University of Colorado, Boulder, CO, USA

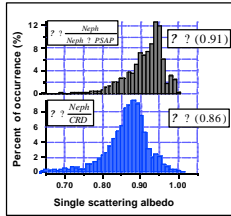
(2) NOAA Earth System Science Laboratory, 325 Broadway, Boulder, CO, USA

(3) Department of Chemistry and Biochemistry, University of Colorado, Boulder, CO, USA

Contact: Paola.Massoli@noaa.gov



Aerosol Optical Properties (AOP)



Extinction (s_{ep}), scattering (s_{sp}), absorption (s_{ap})
Single Scattering albedo (?); $? = s_{sp} / s_{ep}$

MOTIVATION FOR THIS STUDY

Values obtained from different instrument combinations can systematically differ (see NEAQS-ITCT 2004 data). These differences are significant for climate forcing estimates

GOAL

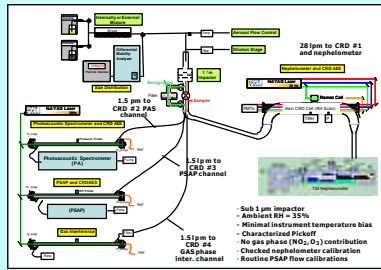
Investigate instrument biases for AOP measurements in relation to aerosol type and composition

Evaluate the uncertainty for the derived ? values

NEAQS-ITCT unpublished data:
CRD-AES (NOAA - Baynard et al.),
Nephelometer (U of Illinois - Rood et al.),
PSAP (PMEL/UW - Covert et al.)

AOP Inter-comparison Study

SET UP



MEASURED AOP QUANTITIES

- Extinction (s_{ep}), 532 nm (NOAA CRD-AES)
 $?s_{ep} = 1\%$ (532 nm, sub-1 μ m, dry) [1]
- Scattering (s_{sp}), 550 nm (TSI 3563 Nephelometer)
 $?s_{sp} = 4-7\%$ (550 nm, sub-1 μ m, dry) [2]
Angle of integration - 7°-170° (truncation), size dep. correction (Å)
- Absorption (s_{ap}), 530 nm (Radiance Research PSAP)
 $?s_{ap} = 20-30\%$ (sub-1 μ m, dry) [3]
Response to non absorbing aerosols (apparent absorption)

AEROSOL TYPES (sub-1 μ m, dry RH < 10%)

Calibration Polystyrene Latex Spheres (PSS) = Non dyed (? > 0.99)

Dyed (? = 0.85 - 0.90)

Non Absorbing (? > 0.99) = Inorganic salts (ammonium sulphate, spherical)

Organic di-carboxylic acids (succinic, adipic)

Absorbing (? < 0.99) = Nigrosin Dye (black water soluble pigment, spherical)

LOOK AT

Spherical Particles

Internal/External Mixtures

Range of ?

Non-volatile/semi-volatile

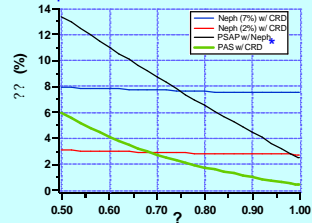
Computed ? values

$?_{CN} = s_{sp} / s_{ep}$ (CRD, NEPH)

$?_{PN} = s_{sp} / (s_{sp} + s_{ap})$ (PSAP, NEPH)

Predicted uncertainty in ?

$?_{ep} = 25 \text{ Mm}^{-1}$, det. limit = 0.1 Mm^{-1}



Data Processing (based on non absorbing aerosols)

s_{sp} = truncation error, and normalization to s_{ep} (0-2%)

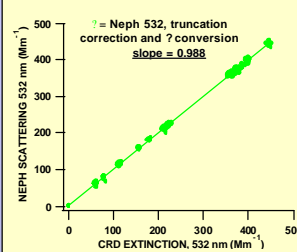
s_{sp} = wavelength conversion (550 nm to 532 nm)

s_{sp} = 'apparent absorption' correction (0 to 2 % of s_{sp})

* PAS = Photo Acoustic Spectrometer, [4]

Non absorbing Aerosols ($\text{NH}_4)_2\text{SO}_4$

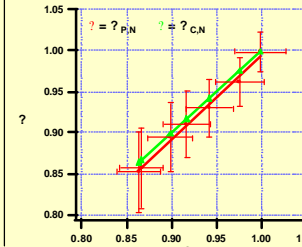
Integrated CRD-NEPH system's $s_{ep} - s_{sp} < 2\%$; ? $s_{sp} = 2-3\%$



For non absorbing aerosols
 $s_{ep} = s_{sp}$

? $C_N = s_{sp} / s_{ep}$
(CRD-AES and NEPH)
IS THE REFERENCE ?
IN THIS STUDY

Calibration 450 nm PSS - non absorbing and absorbing (10% dye)



Test aerosols (spherical, known size)
good to verify instrument agreement

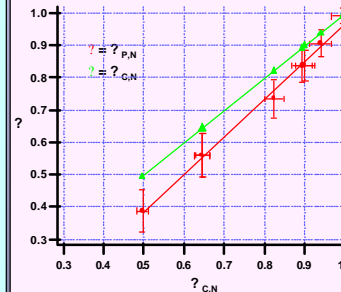
External mixture ranging from 100% non dyed to 100% dyed covering the typical atmospheric aerosol ? range

AMMONIUM SULPHATE /NIGROSIN MIXTURES (AS/NSN)

Proxy for complex inorganic/organic atmospheric aerosols mixtures

Case 1) External mixture AS/NSN

1% solutions of pure NSN and pure AS mixed by using two atomizers



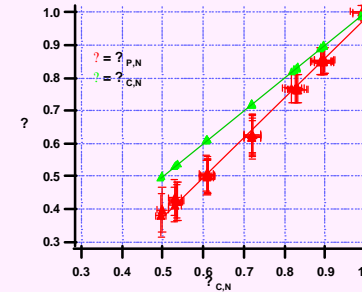
Good agreement between ? C_N and ? P_N for the non absorbing aerosols in both cases

? P_N lower than ? C_N , discrepancy linearly increasing with the NSN % independently on filter transmission

Not significant differences between external vs internal AS/NSN mixtures

Case 2) Internal mixtures (AS/NSN)

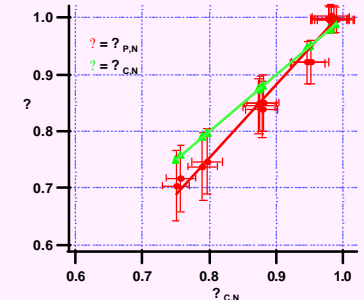
Mixtures of known NSN fraction (pure AS to pure NSN)
-Good repeatability of ? values



NON ABSORBING ORGANIC ACIDS/NSN

Di-carboxylic (succinic, adipic) acids are non absorbing, water soluble semi-volatile compounds, commonly found in ambient aerosols

Range of NSN amount is 0 to 40%



Similar trend of ? C_N vs ? P_N as for AS/NSN mixture
Lower than expected ? C_N values for pure succinic acid

?? (? $C_N - ? P_N$) @ ? = 0.86 (atmospheric range)

PSS	?? = 0.01
AS/NSN external	?? = 0.05
AS/NSN internal	?? = 0.045
ORGANICS/NSN	?? = 0.02

Overestimation of absorption by NEPH, PSAP respect to the reference CRD, NEPH

Significant discrepancies between ?? values between calibration spheres and mixtures

Dependence of the applied correction on mixture is a generalization possible?

Systematic instrument biases need to be investigated under controlled conditions with laboratory generated aerosols to obtain detailed and specific correction factors before moving to atmospheric cases

For ? = 0.86, ?? = 0.05 is a too big uncertainty for accurate calculations of radiative aerosol forcing

SUMMARY

The use of integrated CRD, NEPH approach can reduce the propagated uncertainty in ? and help evaluating filter based absorption measurement technique, e.g., PSAP

Good closure between CRD, NEPH and PSAP, NEPH for non absorbing aerosols

Discrepancies still remaining for absorbing aerosols indicate that recommended correction factors cause significant biases in AOP measurements - need to account for dependence of such factors on aerosol size, morphology and composition

Need to improve AOP measurement techniques and methods

References

- Baynard, T., A. Pettersson, E. R. Lovejoy, S. S. Brown, H. Osthoff, D. Lack, S. J. Ciciora, B. Dube and A. R. Ravishankara (2005), "Direct Measurement of Atmospheric Aerosol Optical Extinction using Cavity Ring-down Spectrometry: Field Instrument Development." *submitted to Aerosol Science and Tech.*
- Anderson, T. L. and J. A. Ogren (1998), "Determining Aerosol Radiative Properties using the TSI 3563 Integrating Nephelometer." *Aerosol Science and Technology* 29(1): 57
- Bond, T. C., T. L. Anderson, and D. Campbell (1999), "Calibration and Intercomparison of Filter-Based Measurements of Visible Light Absorption by Aerosols." *Aerosol Science and Technology* 30(6): 582
- Lack, D., E. R. Lovejoy, T. Baynard, A. Pettersson and A. R. Ravishankara (2006), "Aerosol Absorption Measurement using Photoacoustic Spectroscopy: Sensitivity, Calibration, and Uncertainty Developments." *Aerosol Science and Technology* 40(1): 697

Acknowledgements

NOAA's Climate Program
for Project Funds

Disclaimer

This article was prepared by the Cooperative Institute for Research in Environmental Sciences (CIRES) with support in part from the National Oceanic and Atmospheric Administration, U.S. Department of Commerce, under cooperative agreement NA17OZ02 and other grants. The statements, conclusions, and recommendations are those of the author(s) and do not necessarily reflect the views of the National Oceanic and Atmospheric Administration or the Department of Commerce.