Characteristics of Brown Carbon Present in Ambient Aerosols Over Different Regions in India

A Thesis

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By

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DECLARATION

I, Mr. Rangu Venkata Satish Kumar, S/o R Tirupatha Chari, a resident of PRL, Ahmedabad-380009 hereby declare that the research work incorporated in the present thesis entitled "**Characteristics of Brown Carbon Present in Ambient Aerosols Over Different Regions in India**" is my work and is original. This work (in part or in full) has not been submitted to any University for the award of a degree or a diploma.

I have properly acknowledged the material collected from the secondary sources wherever required, and I have run my entire thesis on the anti-plagiarism software namely, "**iThenticate**."

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CERTIFICATE

I feel great pleasure in certifying that the thesis entitled "**Characteristics of brown carbon present in ambient aerosol over different regions in India**" by Mr. Rangu Venkata Satish Kumar has been completed under my guidance.

He has completed the following requirements as per the Ph.D. regulations of the University:

(a)Completion of the Course Work as per the university rules.

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(c)Regular submission of six monthly progress reports.

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(e)Publication of at least one research paper in a refereed research journal.

I am satisfied with the analysis of data, interpretation of results and conclusions drawn. I recommend the submission of the thesis.

Date:2018

Name and Designation of the supervisor

Dr. Neeraj Rastogi

Associate Professor

Countersigned by

Dedicated to Nanna, Amma
and Grand Parents

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ABSTRACT

Brown carbon (BrC) is a part of organic carbon (OC) which efficiently absorbs light at near UV and visible region, and it is characterized by an absorption spectrum that smoothly increases from the visible to UV wavelengths. BrC immersed in cloud droplets can absorb light and facilitate evaporation/dispersion of clouds. It contributes ∼35% of the direct radiative forcing warming caused by carbonaceous aerosols. However, to better assess the effects of BrC on air quality and climate, a proper understanding of its sources and characteristics on temporal and spatial scales is very important. In this study, we have characterized atmospheric BrC using both offline and online techniques. With an aim to understand the characteristics of BrC over different regions dominated by different emission sources and/or meteorological conditions, many regions (Patiala, Delhi, Kanpur, Ahmedabad, Shillong, and Port Blair) and sampling periods were selected for particulate matter smaller than 2.5 µm aerodynamic diameter ($PM_{2.5}$) sampling. Absorption coefficient at 365 nm is used as a general measure of water-soluble (($b_{abs\_365\_Water}$) and methanol soluble ($b_{abs\_365\_Methanol}$) BrC. Further, BrC characteristics were investigated concerning its sources, chemical composition, optical properties, and atmospheric evolution. Results from this study suggest that primary emissions from biomass burning (BB) and fossil fuel burning (FFB) are the major sources of highly absorbing BrC, and BB emits significant fraction of water-insoluble BrC. Further, secondary BrC and aged/oxygenated OA are found to be relatively less absorbing. BrC absorption decreases as the day progresses achieving a minimum during afternoon hours, which is ascribed to photo-bleaching/volatilization of BrC. It is also shown that BrC composition is not uniform throughout the day. Our observations suggest that BrC chromophores are a variable mixture of at least HULIS and nitrogen-containing organic compounds over the study regions. Mass absorption efficiency (MAE) of water-soluble and methanol-soluble BrC is found to be significant compare to that of elemental carbon over most of the study sites. These results have important implications in estimating regional radiation budget.

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LIST OF ABBREVATIONS

AAE	Absorption Ångström Exponent
AOD	Aerosol Optical Depth
BB	Biomass Burning
BBOA	Biomass Burning Organic Aerosol
BC	Black Carbon
BrC	Brown Carbon
CCN	Cloud Condensation Nuclei
CO	Carbon Monoxide
EC	Elemental Carbon
FFB	Fossil Fuel Burning
FID	Flame Ionization Detector
HOA	Hydrocarbon like Organic Aerosol
HYSPLIT	Hybrid Single Particle Langrangian Integrated Trajectory
IGP	Indo-Gangetic Plain
IPCC	Intergovernmental Panel on Climate Change
KHP	Potassium Hydrogen Phthalate
MAE	Mass Absorption Efficiency
MODIS	Moderate Resolution Imaging Spectroradiometer

NDIR	Non Dispersive Infrared Detector
NIOSH	National Institute of Occupational Safety and Health
NOAA	National Oceanic and Atmospheric Administration
NO	Nitrogen Oxide
NO2	Nitrogen Dioxide
OA	Organic Aerosol
OC	Organic Carbon
OM	Organic Matter
OPAC	Optical Properties of Aerosol and Clouds
PC	Pyrolyzed Carbon
PM	Particulate Matter
RH	Relative Humidity
SOA	Secondary Organic Aerosol
SVOOA	Semivolatile Oxygenated Organic Aerosol
TC	Total Carbon
TOC	Total Organic Carbon
TSP	Total Suspended Particulates
VOC	Volatile Organic Carbon
WSIS	Water Soluble Ionic Species
WSOC	Water Soluble Organic Carbon

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List of Publications:

1. A. Singh, N. Rastogi, A. Patel, **R. V. Satish**, D. Singh, Size-Segregated Characteristics of Carbonaceous Aerosols over the Northwestern Indo-Gangetic Plain: Year Round Temporal Behaviour. *Aerosol Air Qual. Res.* **2016**, *16* (7), 1615–1624 DOI: 10.4209/aaqr.2016.01.0023.

2. **R.V. Satish**, P.M. Shamjad, N.M. Thamban, S. N. Tripathi and N. Rastogi, Temporal Characteristics of Brown Carbon over the Central Indo-Gangetic Plain. *Environ. Sci. Technol.* **2017**, acs.est.7b00734 DOI: 10.1021/acs.est.7b00734.

3. P. M Shamjad, **R. V**. **Satish**, N. M Thamban, N. Rastogi, Tripathi, S. N. Absorbing Refractive Index and Direct Radiative Forcing of Atmospheric Brown Carbon over Gangetic Plain. *ACS Earth Sp. Chem.* **2018**, *2* (1), 31–37 DOI: 10.1021/acsearthspacechem.7b00074.

4. N. Rastogi, A. Singh, **R. V. Satish**, Characteristics of sub-micron particles coming from a big firecrackers burning event: Implications to atmospheric pollution. *Atmos. Pollut. Res*., 2018 (accepted, in press).

5. **R. V**. **Satish** and N. Rastogi, On the Use of Brown Carbon Spectra as a Tool to Understand Their Broader Composition and Characteristics: A Case Study from Crop-residue Burning Samples, 2018 (Under review)