

**INVESTIGATION OF AEROSOL HYGROSCOPICITY,  
ITS CAUSATIVE FACTORS, AND IMPACT AT AN  
URBAN SITE**

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**DISSERTATION  
ON  
INVESTIGATION OF AEROSOL HYGROSCOPICITY,  
ITS CAUSATIVE FACTORS, AND IMPACT AT AN  
URBAN SITE**

*by*

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**Submitted**

**in fulfilment of the requirements of the degree of Doctor of Philosophy  
to the**



**INDIAN INSTITUTE OF TECHNOLOGY DELHI  
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*Dedicated to my Creator, Allah.*

## **CERTIFICATE**

This is to certify that the dissertation entitled ‘Investigation of Aerosol Hygroscopicity, its Causative Factors, and Impact at an Urban Site’ being submitted by Zainab Arub to the Indian Institute of Technology Delhi, for the fulfillment of the requirements for the award of Doctor of Philosophy in Civil Engineering is a record of bonafide research work carried out by her. She has worked under my supervision and has fulfilled the requirements, which to my knowledge, has reached the requisite standard for the submission of the thesis. The research report and results presented in this thesis have not been submitted, in part or full, to any other university or institute for the award of any degree or diploma.

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## ABSTRACT

This is in an age of rampant climate change. There are huge uncertainties in the quantification of effective radiative forcings due to the interaction of aerosols with both clouds and solar radiations as well as precipitation by GCMs (Global Climatic Models). This work is a small effort to contribute to reduce these uncertainties in the long run. This was made possible by (a) adding to the estimates of hygroscopicity parameter ( $\kappa$ ) and cloud condensation nuclei (CCN) in response to the dearth of such data, especially in the Indian sub-continent, and (b) by adding to the scientific understanding of these parameters. These have been accomplished as a result of two campaigns, both carried out at New Delhi, which is a highly polluted continental site in southern Asia home to anthropogenic emissions locally and long-range transfer of pollutants.

The first campaign was a long-term study from Jan 2017- Mar 2018 based on time-resolved estimates of  $\kappa$  and CCN in the PM<sub>1</sub> (Particulate Matter upto 1 $\mu$ m; refers to aerosol particles with an aerodynamic diameter within 1  $\mu$ m regime) size range obtained from chemical composition (NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, and organics that constitute the Non-refractory PM<sub>1</sub> (NRPM<sub>1</sub>) species) and size distribution, for the first time. Air mass originating from southern Asia (SA) was the most polluted with high Cl<sup>-</sup> and organic loading. SA was followed by the Bay of Bengal air mass (BB) with higher organics and NO<sub>3</sub><sup>-</sup> with contamination exceeding that of the Arabian Sea air mass (AS). The dominant sources included biomass-burning and emissions from thermal power plants, industries, and vehicles. The average  $\kappa$  was reported to be 0.3 (ranging from 0.13-0.77) for all the air masses. The source activities by way of impacting the aerosol chemical composition impacted  $\kappa$ 's diurnal pattern. SA > BB > AS was seen in terms of (a) NRPM<sub>1</sub> loading, (b) number concentrations ( $N_{CN}$ ) in the bulk, Aitken ( $N_{Aitken}$ ), and accumulation ( $N_{Accu}$ ) modes, (c) means of CCN number concentration ( $N_{CCN}$ ; 3669–28926 cm<sup>-3</sup>) and, (d) the activated fraction ( $a_f$ ; 0.19–0.87), for supersaturations (SS) ranging from 0.1- 0.8 %. Size impacted  $N_{CCN}$  more than chemical composition directly, being governed by  $N_{Aitken}$  or  $N_{Accu}$  depending on SS and critical diameter ( $D_c$ ).  $a_f$  was ruled by the geometric mean diameter (GMD). The world's highest  $a_f$  (0.71±0.14 for the most dominant sub-branch of the SA air mass- R1- at 0.4 % SS) for a continental site is reported owing to very low  $D_c$  (25–130 nm, for 0.1- 0.8 % SS). Indirectly,  $N_{CCN}$  and  $a_f$  were impacted by the chemical properties via their impact on the diurnal behaviour of  $N_{Aitken}$ ,  $N_{Accu}$ ,  $\kappa$ , and  $D_c$ .

The first study was based on the most prevalent assumption of ammonium salts being the dominant salts at continental sites other than NaCl at a marine location. It is hypothesized that this assumption may lead to a huge underestimation of  $\kappa$  for a polluted site subject to heavy mineral dust loading like New Delhi. Accordingly, the second campaign which is also a long-term study was conducted from Dec 2012–2014 wherein detailed chemical speciation of PM<sub>2.5</sub> aerosols of inorganic ions (NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and Cl<sup>-</sup>) and organic carbon (OC) was carried out based on filter-paper measurements of day and night. The validation of this hypothesis necessitated the quantification of mineral dust on  $\kappa$ . To accommodate the components of mineral dust in  $\kappa$  estimation, two new mathematical approaches differing in the sequence of salt formation based on Mann-Kendall's test were coined. The inclusion of mineral dust components resulted in an average increment of 57% and 63.5% in  $\kappa$  for day and night respectively.  $\kappa$  which was inclusive of mineral dust was inter-compared with those deduced using other analytical methods. This approach challenges the traditional  $\kappa$  estimation  $\kappa$  based on the dominance of ammonium salts while ignoring (a) the other components, their individual and collective impact on  $\kappa$ , and (b) meteorological parameters like temperature (T) and Relative Humidity (RH). The impact of mineral dust on  $\kappa$  decreased with an increase in water-soluble organic carbon to organic carbon ratio. The consideration of insoluble calcium salts lessened the effect of mineral dust on the one hand, while on the other hand resulted in MgCl<sub>2</sub> formation which led to an increase in the estimated  $\kappa$ .  $\kappa$ 's highest value was reported as 0.82 (on 23/7/2014 during the night). The significant contribution of mineral dust to  $\kappa$  was further verified for the cities of Durg and Kanpur in India, and Xian and Beijing in China for PM<sub>1</sub> and PM<sub>2.5</sub> aerosols.

The high values of  $\kappa$ ,  $N_{CCN}$ , and  $a_f$  in the PM<sub>1</sub> regime can severely impact Delhi's rainfall, the radiation budget, and aerosol indirect effect, and require a thorough investigation to quantify this impact. The high impact of mineral dust on  $\kappa$  at polluted sites as seen in the PM<sub>2.5</sub> regime for Delhi, and both PM<sub>1</sub> and PM<sub>2.5</sub> regimes for the other cities can be of crucial relevance for CCN estimation and thus impact precipitation quantification by GCMs, lead to visibility deterioration and affect the indirect radiative forcing.

## सारांश

यह बड़े पैमाने पर जलवायु परिवर्तन का युग है। बादलों और सौर विकिरणों के साथ एरोसोल की परस्पर क्रिया के कारण प्रभावी विकिरण बल के साथ-साथ जीसीएम (वैश्विक जलवायु मॉडल) द्वारा वर्षा के परिमाणन में भारी अनिश्चितता है। आगे चल कर इन अनिश्चितताओं को कम करने में योगदान देने के लिए यह कार्य एक छोटा सा प्रयास है। यह संभव हो सका (1) विशेष रूप से भारतीय उपमहाद्वीप में इस तरह के डेटा की कमी के जवाब में हाइग्रोस्कोपिसिटी पैरामीटर ( $\kappa$ ) और क्लाउड कंडेनसेशन न्यूक्लियाई (CCN) के अनुमानों को जोड़ने से, और (2) इन मापदंडों की वैज्ञानिक समझके योगदान से। इन्हें दो अभियानों के परिणामस्वरूप पूरा किया गया है, दोनों नई दिल्ली में किए गए हैं, जो कि दक्षिणी एशिया में एक अत्यधिक प्रदूषित महाद्वीपीय स्थल है, और स्थानीय रूप से मानवजनित उत्सर्जन और प्रदूषकों के लंबी दूरी के हस्तांतरण के अधीन है।

पहला अभियान जनवरी 2017- मार्च 2018 का एक दीर्घकालिक अध्ययन था, जो आधारित था  $\kappa$  और CCN के समय-समाधान अनुमान पीएम<sub>1</sub>(पार्टिकुलेट मैटर 1 $\mu$ m तक; 1 माइक्रोन शासन के भीतर वायुगतिकीय व्यास वाले एरोसोल कणों को संदर्भित करता है) आकार सीमा में, प्राप्त की गई पहली बार रासायनिक संरचना (NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, और ऑर्गेनिक्स जो की गैर-दुर्लभ PM<sub>1</sub> (NRPM<sub>1</sub>) प्रजातियां हैं) और आकार वितरण के माध्यम से। दक्षिणी एशिया (SA) से उत्पन्न वायु द्रव्यमान अत्याधिक Cl<sup>-</sup> और कार्बनिक लोडिंग के साथ सबसे प्रदूषित था। SA के बाद बंगाल की खाड़ी का वायु द्रव्यमान (BB) था, अत्यधिक कार्बनिक पदार्थों और NO<sub>3</sub><sup>-</sup> के साथ, जिसका संदूषण अरब सागर वायु द्रव्यमान (AS) से अधिक था। प्रमुख स्रोतों में बायोमास-बर्निंग और थर्मल पावर प्लांट, उद्योगों और वाहनों से उत्सर्जन शामिल थे। सभी वायु द्रव्यमानों के लिए औसत  $\kappa$  0.3 (0.13-0.77 के बीच) बताया गया। एरोसोल रासायनिक संरचना को प्रभावित करने के माध्यम से स्रोत गतिविधियों ने  $\kappa$  के दैनिक प्रतिरूप को प्रभावित किया। SA > BB > AS देखा गया निम्नलिखित के लिए (1) NRPM<sub>1</sub> लोडिंग, (2) संख्या सांद्रता थोक में ( $N_{CN}$ ), एटकेन ( $N_{Aitken}$ ), और एक्युमुलेशन ( $N_{Accu}$ ) मोड के संदर्भ में, (3) औसत CCN संख्या एकाग्रता ( $N_{CCN}$ ; 3669-28926 cm<sup>-3</sup>), और (d) सक्रिय अंश ( $a_f$ ; 0.19-0.87),सूपरसैचुरेशन (SS) 0.1- 0.8 % के लिए। आकार ने  $N_{CCN}$  को प्रत्यक्ष रूप से रासायनिक संरचना से अधिक प्रभावित किया, जो SS और महत्वपूर्ण व्यास ( $D_C$ ) के आधार पर  $N_{Aitken}$  या  $N_{Accu}$  द्वारा नियंत्रित होता है।  $a_f$  ज्यामितीय माध्य व्यास (GMD) द्वारा शासित था। एक महाद्वीपीय स्थल के लिए दुनिया का सबसे उच्चतम  $a_f$  (0.71±0.14 SA वायु द्रव्यमान की सबसे प्रमुख उप-शाखा- R1- 0.4 %

SS पर) बहुत कम  $D_C$  (25-130 nm, 0.1- 0.8 % SS) के कारण सूचित किया गया है। परोक्ष रूप से,  $N_{CCN}$  और  $a_f$  रासायनिक गुणों से प्रभावित थे,  $N_{Aitken}$ ,  $N_{Accu}$ , और  $D_C$  के दैनिक व्यवहार पर उनके प्रभाव के माध्यम से।

पहला अध्ययन एक समुद्री स्थान पर NaCl के अलावा अन्य महाद्वीपीय स्थलों पर अमोनियम लवण के प्रमुख लवण होने की सबसे प्रचलित धारणा पर आधारित था। यह अनुमान लगाया गया है कि इस धारणा से नई दिल्ली जैसे भारी खनिज धूल लोडिंग के अधीन प्रदूषित साइट के लिए  $\kappa$  का एक बड़ा कम आंकलन हो सकता है। तदनुसार, दूसरा अभियान जो एक दीर्घकालिक अध्ययन भी है, दिसंबर 2012-2014 से आयोजित किया गया था जिसमें  $PM_{2.5}$  एरोसोल के अकार्बनिक आयनों ( $NH_4^+$ ,  $Na^+$ ,  $K^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $SO_4^{2-}$ ,  $NO_3^-$ , और  $Cl^-$ ) और कार्बनिक कार्बन (OC) की विस्तृत रासायनिक विशिष्टताएं थीं, जो दिन और रात के फिल्टर-पेपर माप के आधार पर किया गया था। इस परिकल्पना की पुष्टि के लिए  $\kappa$  पर खनिज धूल की मात्रा का ठहराव आवश्यक हो गया।  $\kappa$  अनुमान में खनिज धूल के घटकों को समायोजित करने के लिए, मान-केंडल के परीक्षण के आधार पर लवण निर्माण के क्रम में भिन्न दो नए गणितीय दृष्टिकोण गढ़े गए। खनिज धूल घटकों को शामिल करने से दिन और रात के लिए  $\kappa$  में क्रमशः 57% और 63.5% की औसत वृद्धि हुई।  $\kappa$  जिसमें खनिज धूल शामिल थी, अन्य विश्लेषणात्मक तरीकों का उपयोग करके निकाले गए  $\kappa$  के साथ तुलना की गई। यह दृष्टिकोण अमोनियम लवण के प्रभुत्व के आधार पर पारंपरिक अनुमान को चुनौती देता है, जबकि (1) अन्य घटकों,  $\kappa$  पर उनके व्यक्तिगत और सामूहिक प्रभाव, और (2) तापमान (T) और सापेक्ष आर्द्रता (RH) जैसे मौसम संबंधी मापदंडों की अनदेखी करते हैं। पानी में घुलनशील कार्बनिक कार्बन से कार्बनिक कार्बन अनुपात में वृद्धि के साथ  $\kappa$  पर खनिज धूल का प्रभाव कम हो गया। अघुलनशील कैल्शियम लवणों के विचार ने एक ओर खनिज धूल के प्रभाव को कम किया, जबकि दूसरी ओर  $MgCl_2$  के गठन के परिणामस्वरूप अनुमानित  $\kappa$  में वृद्धि हुई।  $\kappa$  का उच्चतम मान 0.82 (रात के दौरान 23/7/2014 को) बताया गया था।  $\kappa$  में खनिज धूल के महत्वपूर्ण योगदान को भारत के दुर्ग और कानपुर शहरों और चीन के जियान और बीजिंग में  $PM_{10}$  और  $PM_{2.5}$  एरोसोल के लिए और अधिक सत्यापित किया गया।

$PM_{10}$  आकार सीमा में,  $N_{CCN}$ , और  $a_f$  के उच्च मान दिल्ली की वर्षा, विकिरण बजट और एरोसोल अप्रत्यक्ष प्रभाव को गंभीर रूप से प्रभावित कर सकते हैं, और इस प्रभाव को मापने के लिए गहन जांच की आवश्यकता है। प्रदूषित स्थलों पर  $\kappa$  पर खनिज धूल का उच्च प्रभाव, जैसा कि दिल्ली के लिए  $PM_{2.5}$  आकार सीमा में देखा गया है, और अन्य शहरों के लिए  $PM_{10}$  और  $PM_{2.5}$  दोनों आकार सीमाओं में, CCN आकलन के लिए महत्वपूर्ण प्रासंगिक हो सकते हैं और इस प्रकार GCM द्वारा वर्षा की मात्रा को

प्रभावित कर सकते हैं, दृष्टता में गिरावट का कारण बन सकते हैं और अप्रत्यक्ष विकरण बल को प्रभावित कर सकते हैं ।

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