

1 **Supplementary information of**
2 **“A key process controlling the wet removal of aerosols: new**
3 **observational evidence”**

4

5 Sho Ohata¹, Nobuhiro Moteki^{1*}, Tatsuhiro Mori¹, Makoto Koike¹, and Yutaka Kondo²

6

7 ¹Department of Earth and Planetary Science, Graduate School of Science, The
8 University of Tokyo, Tokyo, Japan.

9 ²National Institute of Polar Research, Tokyo, Japan.

10

11

12 ***Corresponding author**

13 N. Moteki, Department of Earth and Planetary Science, Graduate School of Science,
14 The University of Tokyo, Hongo 7-3-1, Bunkyo-ku, Tokyo, 113-0033, Japan (e-mail:
15 moteki@eps.s.u-tokyo.ac.jp).

16

17 Last modified on August 24, 2016

18

19

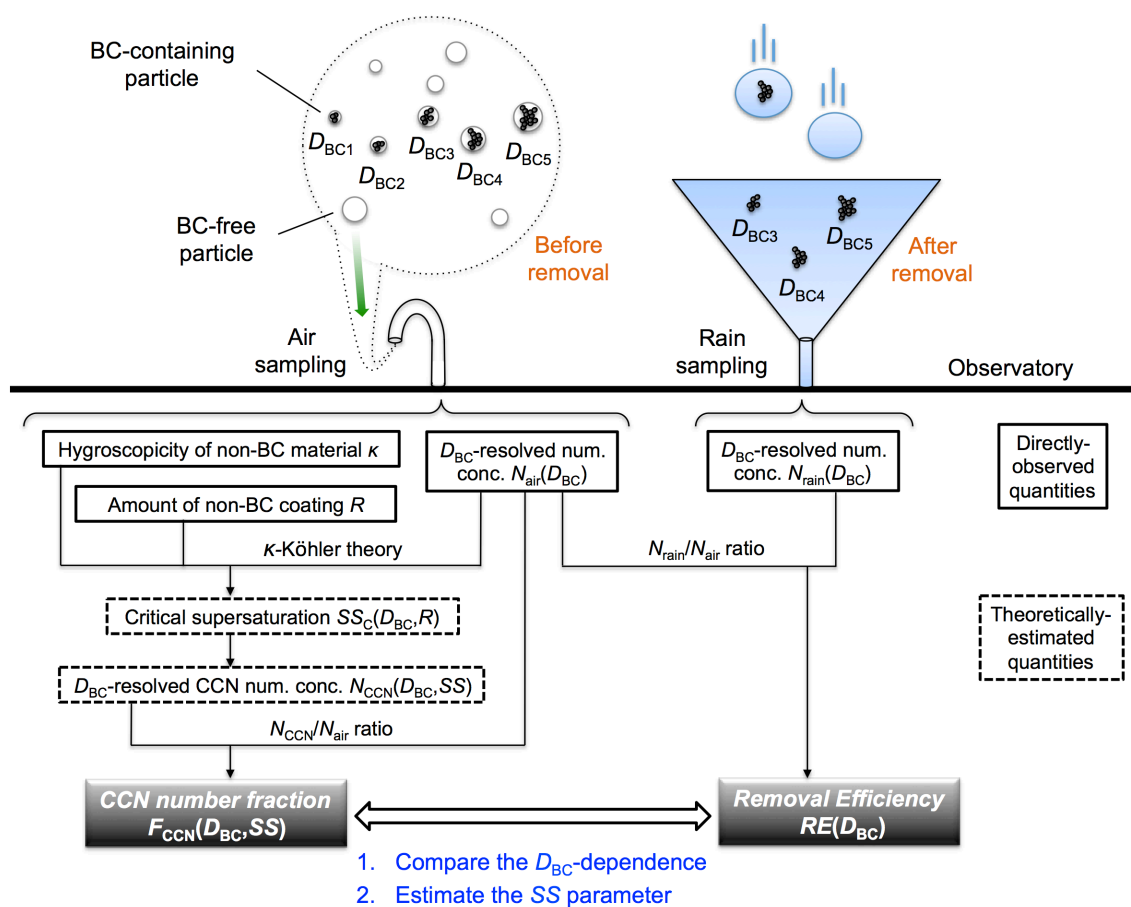
20

21 **Supplementary Table S1. Symbols used in this paper.**

Symbols	Definitions
D_{BC}	Mass equivalent diameter of BC
$F_{bc-imp}(D_{BC})$	D_{BC} -resolved number fraction of BC in air that will be scavenged by impaction with rain droplets below cloud
$F_{CCN}(D_{BC}, SS)$	D_{BC} -resolved number fraction of BC in air that will be activated as CCN at SS
$F_{ic-imp}(D_{BC})$	D_{BC} -resolved number fraction of BC in air that will be scavenged by impaction with cloud or rain droplets in cloud
κ	Hygroscopicity parameter of non-BC aerosols in air
$N_{air}(D_{BC})$	D_{BC} -resolved number concentration of BC in air
$N_{CCN}(D_{BC})$	D_{BC} -resolved number concentration of BC in air that will be activated as CCN at SS
$N_{rain}(D_{BC})$	D_{BC} -resolved number concentration of BC in rainwater
R	Shell-to-core diameter ratio (relative coating amount) of each BC particle in air
$RE(D_{BC})$	D_{BC} -resolved removal efficiency; $N_{rain}(D_{BC}) / N_{air}(D_{BC})$
SS	Supersaturation of water vapour
$SS_c(D_{BC}, R)$	D_{BC} -resolved critical supersaturation of BC in air
SS_{est}	An SS value providing the best agreement of D_{BC} -dependence between $F_{CCN}(D_{BC})$ and $RE(D_{BC})$

22

23



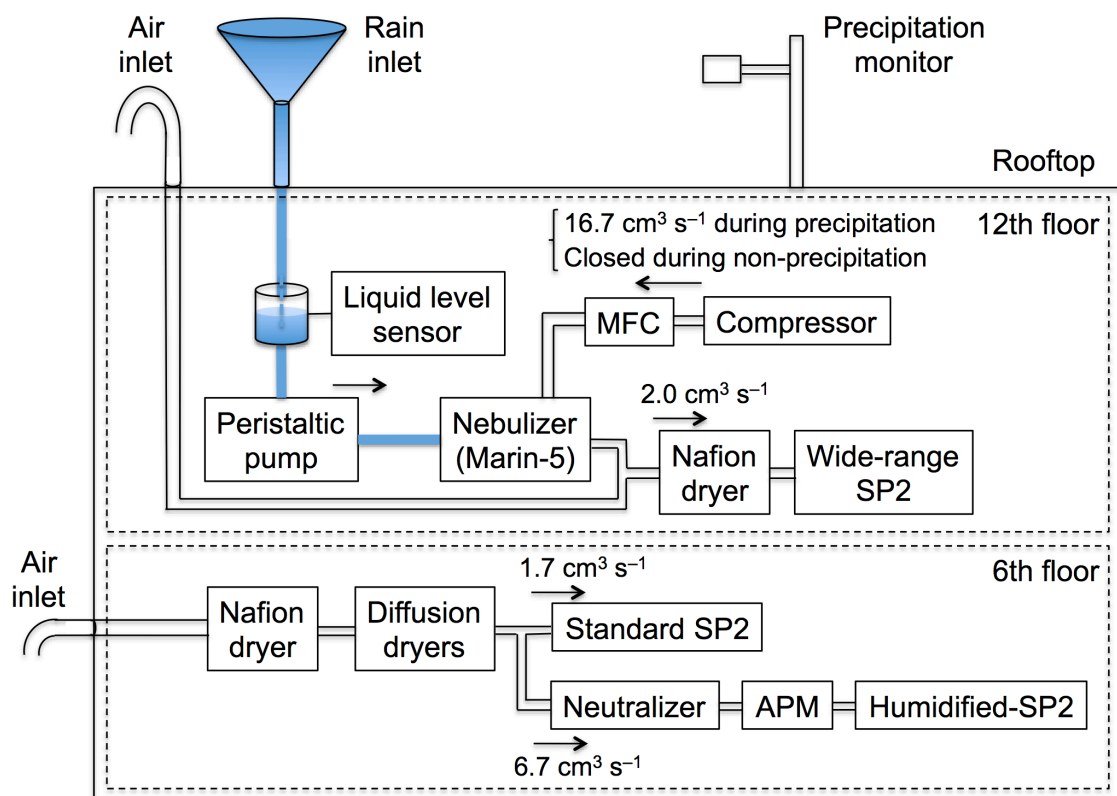
24

25

26 **Supplementary Figure S1. A flow chart illustrating the sampling, measurements,**

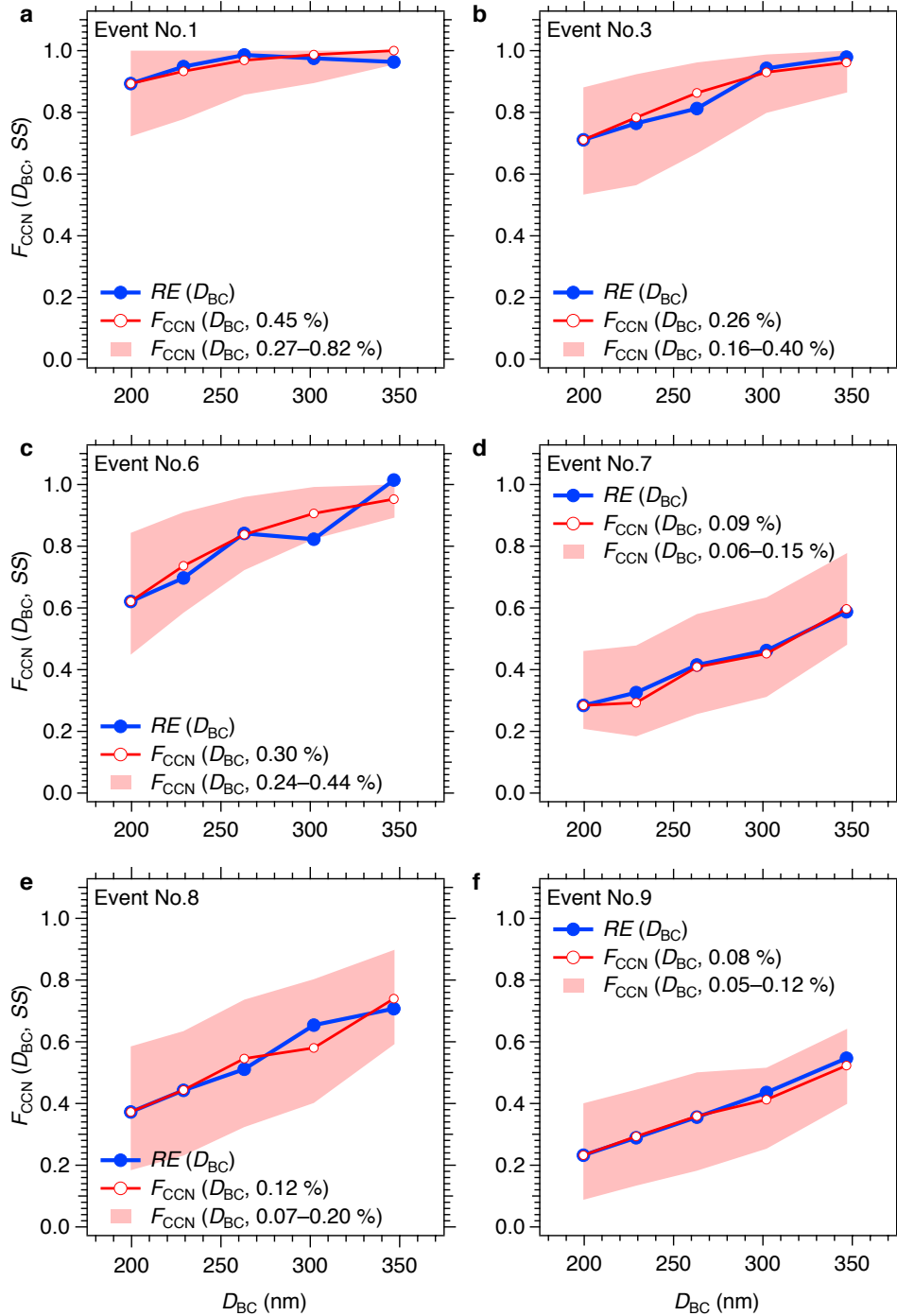
27 **and data analysis in our observational method. See the main text for details.**

28



29
30
31
32
33
34

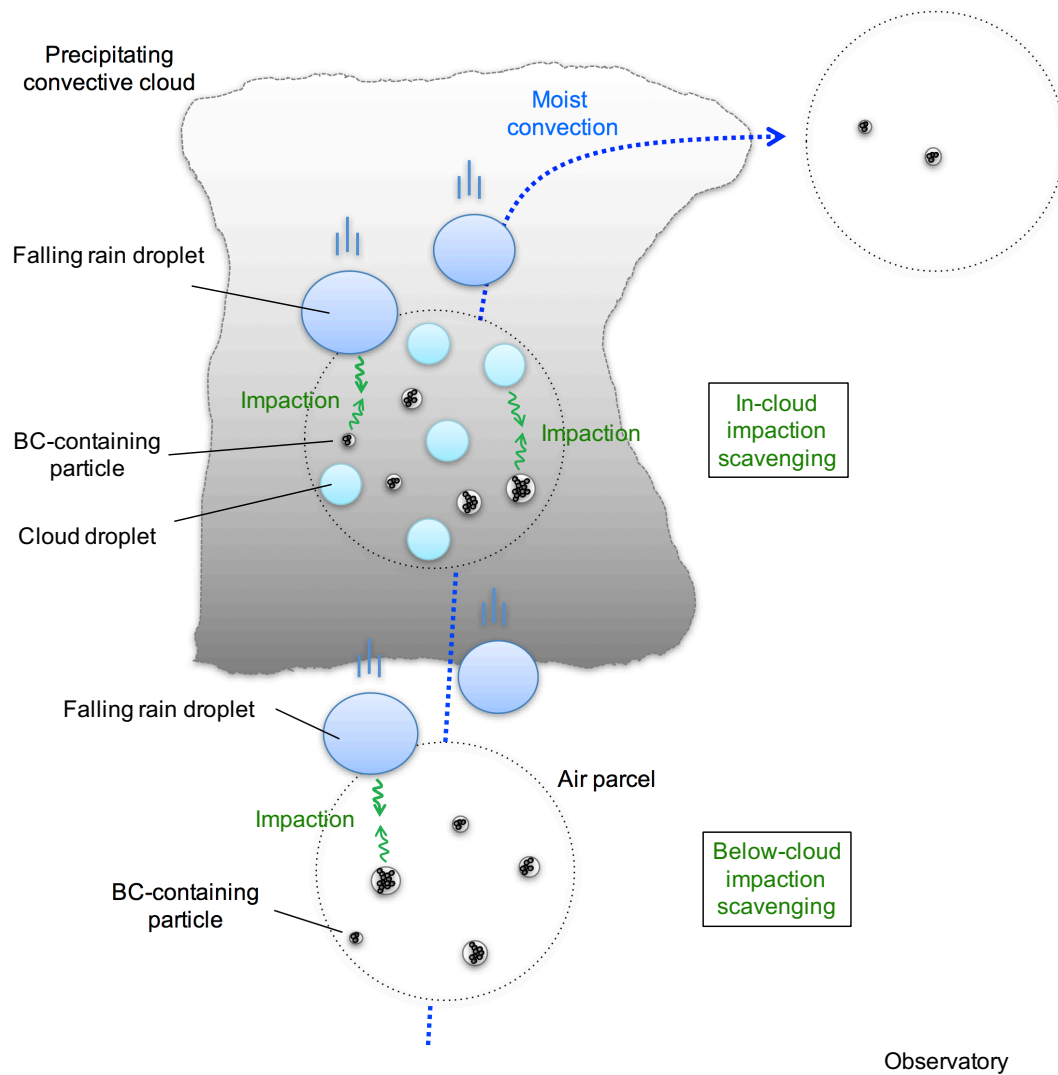
Supplementary Figure S2. Experimental setup. The flow rates are given for room temperature ($\sim 298 \text{ K}$) and pressure ($\sim 1013 \text{ hPa}$). MFC, mass flow controller; SP2, single particle soot photometer; APM, aerosol particle mass analyser.



35

36 **Supplementary Figure S3. D_{BC} -resolved wet removal efficiency (RE) and CCN**
 37 **number fraction (F_{CCN}) of BC-containing aerosols for other precipitation events.**

38 Same as Fig. 3 in the main text but for the other 6 precipitation events. Since both the
 39 estimated $F_{ic-imp}(D_{BC})$ and $F_{bc-imp}(D_{BC})$ are much smaller than $F_{CCN}(D_{BC}, SS)$ for the size
 40 range of $D_{BC} = 185\text{--}370$ nm, they are not shown for illustrative convenience.



41

42

43 **Supplementary Figure S4. A diagram of the in-cloud and below-cloud impactation**

44 **scavenging mechanisms considered in this study.** For the estimates of in-cloud

45 scavenging number fraction F_{ic-imp} , the collision of a BC-containing particle with a

46 cloud droplet and a falling rain droplet is considered. For the estimates of below-cloud

47 scavenging number fraction F_{bc-imp} , the collision of a BC-containing particle with a

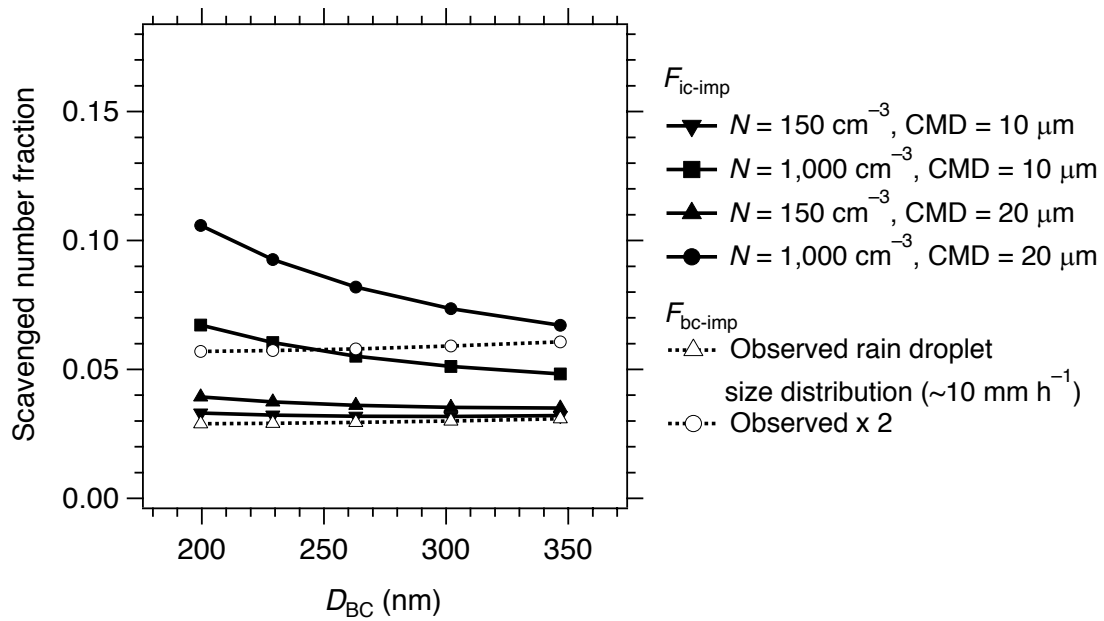
48 falling rain droplet is considered. The number size distribution of cloud droplets and the

49 residence time of an air parcel in cloud and below cloud are assumed (see the Methods

50 section).

51

52



53

54

55 **Supplementary Figure S5. D_{BC} -resolved number fraction scavenged by impaction**
 56 **in cloud (F_{ic-imp}) and below cloud (F_{bc-imp}) for a selected precipitation event.** The
 57 results for the event No. 4 are shown. The solid and dashed lines show the estimated
 58 F_{ic-imp} and F_{bc-imp} , respectively. The different markers indicate various assumptions of
 59 the number size distributions of water droplets.

60

61

62

63

64

65

66

67