Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts. This journal is © The Royal Society of Chemistry 2014



## **Supplementary Information**



3

4 Fig. S1 The co-variability of OC and nss-SO<sub>4</sub><sup>2-</sup> during paddy-residue burning emissions
5 (October–November) in the Indo-Gangetic Plain (IGP) in: (a) 2008 and (b) 2010.

6

The farmers utilize Bentonite sulphur (pure elemental S  $\approx$  90%; a fertilizer) for high paddy-7 crop yield in the IGP. The OC is a major component from paddy-residue burning emission and 8 its co-variability with nss-SO<sub>4</sub><sup>2-</sup> (Supplementary Fig. S1) suggests the origin of SO<sub>4</sub><sup>2-</sup> in ambient 9 aerosols via rapid oxidation of sulphur dioxide gas (SO<sub>2</sub>) involving heterogeneous reactions of 10 aerosols as suggested by Lammel and Leip (2005)<sup>1</sup> following its emission from the paddy-11 residue (containing pure elemental S) burning. The ambient atmospheric conditions and chemical 12 reactivity leading to the formation of  $SO_4^{2-}$  are in sharp contrast to the chamber-based 13 experiments. It is, thus, relevant to study particulate concentrations of SO<sub>4</sub><sup>2-</sup> in ambient aerosols 14 and their impact on atmospheric chemistry and human health. This is a first comprehensive 15 study, conducted from a site located in the Indo-Gangetic Plain, to assess the temporal variability 16

- 17 and impact of agricultural-waste burning emissions on the concentrations of  $SO_4^{2-}$  under ambient
- 18 atmospheric conditions.





Fig. S2 Scatter plot between the nss-K<sup>+</sup> and nss-Ca<sup>2+</sup> infers about the K<sup>+</sup> derived from paddyresidue burning emissions (K<sup>+</sup><sub>BB</sub>) in the IGP.

22

The minimum nss-(K<sup>+</sup>/Ca<sup>2+</sup>) mass ratio of 7 (lower envelop) corresponds to crustal 23 composition. However, the upper envelop (nss-K<sup>+</sup>/nss-Ca<sup>2+</sup> mass ratio of 95) corresponds to 24 mixed contribution from the paddy-residue burning emissions and mineral dust. Thus, it can be 25 inferred that the nss-(K<sup>+</sup>/Ca<sup>2+</sup>) mass ratio of 88 (= 95 - 7) is associated with the paddy-residue 26 burning emissions in the IGP. It is important to state here that in an earlier study, the nss-27  $(K^+/Ca^{2+})$  maximum mass ratio of 10 was attributed for biomass burning emissions, owing to a 28 negligible nss-(K<sup>+</sup>/Ca<sup>2+</sup>) minimum ratio of 0.12 (10 - 0.12  $\approx$  10, two orders of magnitude 29 difference).<sup>2</sup> In this study, the difference between maximum and minimum nss-(K<sup>+</sup>/Ca<sup>2+</sup>) mass 30 ratio has been considered to represent the paddy-residue burning emissions. 31

32

33 The following equations have been used to determine the  $K^+_{BB}$ :

$$K_{BB}^{+} = (nss - K^{+}) - (K_{Dust}^{+})$$
(1)

$$nss - K^{+} = K_{aerosol}^{+} - 0.037 * Na_{aerosol}^{+}$$
(2)

$$K_{Dust}^{+} = 7 * [(nss - Ca^{2+}) - Ca_{BB}^{2+}]$$

$$Ca_{BB}^{2+} = \frac{nss - K^{+}}{88}$$
(3a)
$$Ca_{BB}^{2+} = \frac{nss - K^{+}}{88}$$
(3b)

$$nss - Ca^{2+} = Ca_{aerosol}^{2+} - 0.038 * Na_{aerosol}^{+}$$
(4)



40

41 Fig. S3 The co-variability of OC and K<sup>+</sup><sub>BB</sub> during paddy-residue burning emissions (October–
42 November) in the Indo-Gangetic Plain (IGP) in: (a) 2008 and (b) 2010. BB refers to biomass
43 burning emissions.

44

Recently, it has been emphasized to assess the temporal co-variability of K<sup>+</sup> and OC to utilize the K<sup>+</sup>/OC as a tracer of biomass burning emissions in atmospheric aerosols. <sup>3, 4</sup> In this study, the OC and K<sup>+</sup><sub>BB</sub> concentrations exhibit a temporal co-variability (Supplementary Fig. S3), suggesting the use of K<sup>+</sup><sub>BB</sub>/OC ratio as a tracer of paddy-residue burning emission.

## 49 **References**

- G. Lammel and A. Leip, *Environmental Science and Pollution Research*, 2005, 12, 213220.
- 52 2 C. A. Pio, M. Legrand, C. A. Alves, T. Oliveira, J. Afonso, A. Caseiro, H. Puxbaum, A.
  53 Sanchez-Ochoa and A. Gelencśer, *Atmos. Environ.*, 2008, 42, 7530-7543.
- 54 3 Y. Cheng, G. Engling, K. B. He, F. K. Duan, Y. L. Ma, Z. Y. Du, J. M. Liu, M. Zheng 55 and R. J. Weber, *Atmos. Chem. Phys.*, 2013, **13**, 7765-7781.
- 56 4 X. Zhang, A. Hecobian, M. Zheng, N. H. Frank and R. J. Weber, *Atmos. Chem. Phys.*,
  57 2010, 10, 6839-6853.