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Sources of chemical species in rainwater during monsoon and non-monsoonal periods over two mega cities in India and dominant source region of secondary aerosols

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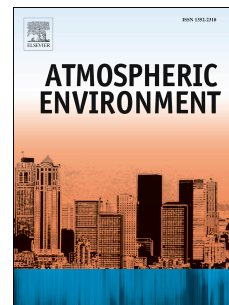
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1 **Sources of chemical species in rainwater during monsoon and non-**  
2 **monsoonal periods over two mega cities in India and dominant source**  
3 **region of secondary aerosols**

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14

15 **Abstract**

16 Samples of rainwater (RW) were collected to characterize the chemistry and sources in two  
17 representative megacities at Pune (Southwest) and Delhi (Northern) India from 2011 to 2014  
18 across two seasons: monsoon (MN) and non-monsoon (NMN). Collected RW samples were  
19 analyzed for major chemical constituents ( $F^-$ ,  $Cl^-$ ,  $SO_4^{2-}$ ,  $NO_3^-$ ,  $NH_4^+$ ,  $Na^+$ ,  $K^+$ ,  $Ca^{2+}$ , and  
20  $Mg^{2+}$ ), pH and conductivity. In addition, bicarbonate ( $HCO_3^-$ ) was also estimated. The mean  
21 pH values of the RW were  $> 6$  at Pune and  $< 6$  at Delhi and 4% and 26% were acidic,  
22 respectively. The mean sum of all measured ionic species in Pune and Delhi was 304.7 and  
23 536.4  $\mu eq/l$ , respectively, indicating that significant atmospheric pollution effects in these  
24 Indian mega cities. Both the  $Ca^{2+}$  and  $SO_4^{2-}$  were the dominant ions, accounting for 43%  
25 (Pune) and 54% (Delhi) of the total ions. The sum of measured ions during the NMN period  
26 was greater than the NM period by a factor of 1.5 for Pune (278.4: NM and 412.1: NMN  
27  $\mu eq/l$ ) and a factor of about 2.5 for Delhi (406 and 1037.7  $\mu eq/l$ ). The contributions of  $SO_4^{2-}$   
28 and  $NO_3^-$  to the RW acidity were  $\sim 40\%$  and  $60\%$ , respectively, at Pune and correspondingly,  
29 36% and 64% at Delhi. The concentrations of secondary aerosols ( $SO_4^{2-}$  and  $NO_3^-$ ) were  
30 higher by a factor of two and three when the air masses were transported to Pune from the  
31 continental side. At Delhi, the concentrations of  $SO_4^{2-}$ ,  $NO_3^-$ ,  $Ca^{2+}$ , and  $Mg^{2+}$  were  
32 significantly higher when the air masses arrive from Punjab, Haryana, and Pakistan indicating  
33 the greater atmospheric pollution over the Indo-Gangetic Plain. Positive matrix factorization  
34 was applied to the source apportionment of the deposition fluxes of these ions. Three factors  
35 were obtained for Pune and four for Delhi. The sources at Pune were secondary aerosols from

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