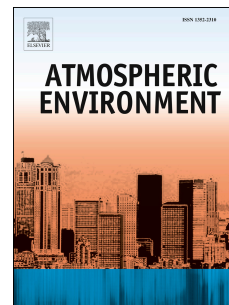


# Accepted Manuscript

Long-term variation of the source of sulfate deposition in a leeward area of Asian continent in view of sulfur isotopic composition

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1 Long-term variation of the source of sulfate deposition in a leeward area of Asian  
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16  
17 **Abstract:**

18 A large emission of air pollutants from the Asian continent has caused  
19 transboundary air pollution, especially in northeastern Asia. This paper evaluates  
20 sulfate deposition at a leeward area of Asian continent, i.e., the Nagaoka observation  
21 station located along the Sea of Japan. We have monitored atmospheric sulfate  
22 deposition and its sulfur isotopic ratio for 28 years at the station. The sulfur isotopic  
23 ratios of non-sea-salt sulfate ( $\delta^{34}\text{Snss}$ ) ranged from 0.0 to +6.2‰. The isotopic ratios  
24 of local emission and Chinese coal sulfur showed negative and positive values,  
25 respectively. Several statistically significant trends were detected on the deposition of  
26 non-sea-salt sulfate ( $\text{nss-SO}_4^{2-}$ ) during the study period. The decrease of  $\text{nss-SO}_4^{2-}$   
27 deposition since the middle of 1980s was considered to have been caused by local  
28 anthropogenic  $\text{SO}_2$  emission that showed relatively low  $\delta^{34}\text{Snss}$  values during the  
29 period. The increase of  $\text{nss-SO}_4^{2-}$  deposition from the end of 1990s to the second half  
30 of 2000s was interpreted to have been caused by the change in  $\text{SO}_2$  emission in China  
31 because the  $\delta^{34}\text{Snss}$  values increased during the period with the winter values getting  
32 closer to the averaged value of Chinese coal sulfur. The decreasing trend of  $\text{nss-SO}_4^{2-}$   
33 deposition from the middle of 2000s was likely affected by reduction of Chinese  $\text{SO}_2$   
34 emission judging from the decrease in  $\delta^{34}\text{Snss}$  values in the period. Mass balance  
35 calculations suggested that sulfur released by coal combustion in China during 1990s  
36 contributed by about 40% of annual total sulfur deposition in Nagaoka, and its

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