

Interactive comment on “Impact of Chinese SO₂ emissions on submicron aerosol concentration at Mt. Tateyama, Japan” by K. Osada et al.

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We thank Dr. Collaud Coen for very constructive comments with respect to the overall clarity of the article. Modified words or sentences are highlighted as yellow in the text.

Comment: This study describes the year to year variation of free tropospheric submicron aerosol measured in Japan, a leeward area of China. Ten years trend of submicron aerosol concentration, SO₂ emission in China, simulated total SO₄²⁻ and Chinese contribution to SO₄²⁻ are presented. Clear correlation between the trend of Chinese contribution of SO₄²⁻ and aerosol concentration was found for December-January and Mars-April periods. This paper reports a very interesting long-term trend analysis of free tropospheric aerosol, some more detailed calculation of the observed trends would however contribute to obtain a clearer analysis and allow comparison with other mea-

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suring sites.

Response: To the revised manuscript, we added additional detailed information (Tables 1–3) related to trend analysis and its discussion.

Comment: Trend analysis: The trend analysis has to be better described. The 5% significance level is cited only once, but values of the slopes with confidence levels are never given. I think that a least mean square analysis (that is probably what was done) with clear confidence level calculation (see Weatherhead et al., 1998 and 2000) should be performed. The slopes, standard deviation or confidence levels for each month and each variable (aerosol concentration, SO₂, total and Chinese contribution to SO₄²⁻) should be then reported in a table. I would also suggest that the trend analysis should be done for each month and not for period of 1, 2 or 3 months. The aerosol concentration is usually a lognormally distributed variable, so that all the analysis requiring a normal distribution should not be applied or only applied to the logarithm of the data. Non-parametric trend analysis methods (such as Mann-Kendall analysis) can otherwise be applied (see Collaud Coen et al., 2007). Whatever the method used is, this point has to be clarified in the paper.

Response: We added results of statistical analyses including slopes, standard deviations, and confidence levels to Tables 1 and 3 of the revised manuscript. We also tested trends using Mann–Kendall analysis, as presented in Table 2. We added references that were necessary for trend analysis.

Comment: Correlation between SO₄²⁻ and aerosol concentration trends: All the explanations given by the authors to explain the correlation between the Chinese contribution to SO₄²⁻ and submicron aerosol volume data are very clear. Fig. 6 also shows an increase (at least after 2005) of SO₄²⁻ in summer, when there is no increase of submicron aerosol volume data. The author should explain why this SO₄²⁻ summer increase does not influence the aerosol trend.

Response: We added possible explanations of the difference between simulation re-

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sults and observation results.

Comment: Other proxies: The authors clearly shows that no change in precipitation are detected for the analyzed period. As described in section 2, NO_x, CO, NMVOC, BC, OC and NH₃ are also included in the simulation. Is there any other correlation between trends of these chemical species and aerosol trends? A small insight in these possible correlations would be very valuable.

Response: We added discussion to the text and a new figure showing other components in the model.

Detailed comments: Abstract: - Line 10: "trends at 5% level": you probably mean 5% significance level or 95% confidence level. Please clarify.

Response: We corrected the text as suggested.

2. Observation, data treatment and numerical model: - Second paragraph: the fact that nighttime data from 2400 to 0500 are representative of free tropospheric conditions (Osada et al., 2003) should also be briefly described.

Response: We added a brief description of diurnal variation at this site.

- p. 5 second paragraph: "we performed simulations": of what? please clarify.

Response: We modified the text in the revised manuscript.

3. Results and discussion 3.1 Temporal variations of submicron aerosols: - first paragraph lines 5-6: The slight increasing tendency of winter minima is not really visible in Fig. 2.

Response: We modified the text of the revised manuscript to include values of winter minima.

- second paragraph line 3: "and low variation in winter" - line 4: In that analysis . . . + delete "in that study"

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Response: We corrected these.

line 7: "with faster movements": not clear.

Response: We modified the text.

- third paragraph: The authors should specify how the trends were calculated and slopes of all the trends have to be given (see global comments). Idem for Fig. 7

Response: We rewrote the text and added additional discussion and tables.

3.2 Factors relating to increasing trend and seasonal preference in winter-spring: - third paragraph: is the seasonality of total and Chinese components of SO₄²⁻ due to a seasonality in the emission of SO₂ or to a seasonality of transport?.

Response: Seasonal variation was not given in emission data. Therefore, these seasonal variations are attributed to the seasonality of transport. We added this to the text in the revised manuscript.

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