

***Interactive comment on “Chinese SO₂ pollution
over Europe –
Part 1: Airborne trace gas measurements and
source identification by particle dispersion model
simulations” by V. Fiedler et al.***

Anonymous Referee #1

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General Comments:

The manuscript presents a case study of a SO₂ pollution episode over the North Atlantic on May 3, 2006. In addition, the study applied FLEXPART analysis to show that the episode can be attributed to surface emission in eastern China. Both methodology and analysis of this study is found to be limited in scope for publication in this journal. I suggest the authors should add more in-depth and convincing discussion before it can be accepted for any publication. Also the authors should consider more serious about the robustness and limitations of the data products they use before jumping into

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any statements. The discussion needs to be put more into the scientific context of other publications. Therefore I think that the paper cannot be considered for publication in its present version, and the authors should find stronger evidence to support their conclusion.

Specific comments:

(1) The lifetime of SO₂ based on reaction with the OH radical, at typical atmospheric levels of OH, is about one week. It would be much shorter when dry deposition and cloud processes are considered. Unlike other long-lived species like CO, it is unlikely for SO₂ to be transported for more than 7 days. Surface SO₂ emission in eastern China is constrained within the boundary layer. Dry deposition may remove SO₂ efficiently from the atmosphere. At a dry deposition velocity of about cm s⁻¹, the lifetime of SO₂ by dry deposition in a 1km boundary layer is about 1 day. Furthermore what kind of processes to enable SO₂ to be transferred to the middle troposphere around 5-6km? What is the role of cloud and precipitation processes prior to the flight measurement of 3 May 2006?

(2) It is not proper to infer the potential source regions of the SO₂ plume using the ratios of SO₂/NO_y and NO/NO_y, especially for long range transport study, because there are diverse sources both for NO_x and SO₂ in the atmosphere. Also such ratios will change with time when the air mass is transported away from the source. Sources for NO_x in the middle and upper troposphere are distinct from that in the low troposphere. Aircraft measured NO and NO_y in Figure 4 are unlikely from Asia and the observed SO₂/NO_y ratio can not be used to identify the source region. Satellite data of OMI SO₂ may give more information for source identification and the evolution of SO₂ concentration for this case study.

(3) Section 1, para. 2, "Pollution from Asia is mainly lifted to the upper troposphere in so-called warm conveyor belts at the eastern seaboard of Asia and subsequently transported by fast air streams in the middle and upper troposphere (Stohl,

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2004). Another transport path is the lifting by deep convection in thunderstorms and mesoscale convective systems in summer (Wild and Akimoto, 2001). What was the process or mechanism responsible for the China plume measured over the North Atlantic on 3 May 2006 in this paper?

(4) SO₂ concentration at height of 6km varied significant during the flight (Fig.1 and Fig.3). How to define a Chinese SO₂ pollution, American SO₂ pollution, or European SO₂ pollution? How to tell the high SO₂ episodes are resulted by long range transport or regional (local) mixing process?

(5) Section 3, there are 2 significant elevated SO₂ layers in Figure2, so called Chinese pollution plume and American pollution plume. Such separated 2 layers may be resulted from the flightpath of measurements and may not represent SO₂ distribution in a large scale or even a region scale. It can be seen in Figure3 that SO₂ decreases with altitude in the beginning and end of measurement when aircraft took off and landed. Even if the 2 separated elevated pollution layers did exist, the higher pollution plume may be caused by regional convective mixing from the lower plume around 2km. Also, in figure 10 FLEXPART modeled results show that the SO₂ with its age less than 2-3 days contribute lots to plume A. The authors should add more analysis to address this issue using meteorological data of the days prior to the measurements.

(6) FLEXPART provides reasonable representation of large-scale horizontal advection, but how good it is in representing sub-grid scale convections? Further, the discussion of the findings should at least mention the potential vertical transport processes (frontal uplifting, deep convection, ?) required for long range transport events. This should be discussed also with regard to what mean wind trajectories can be expected to represent. Without credible information on the vertical lifting, it is difficult to jump into any conclusion of whether it is coming from surface emissions.

(7) As far as this methodology and robustness of using trajectories models to infer the vertical motions of air mass during transport is concern, the analysis should incorpo-

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rate a large number of ensembles and rigorously consider possible injection heights to present reasonably conclusive results.

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