

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 #2

Received and published: 5 February 2009

This manuscript describes airborne measurements of SO₂ and other anthropogenic trace gases above the western edge of Europe. While this is not the first study of Asian pollution over Europe, it does provide the first measurements of Asian SO₂ above Europe that I am aware of. The subject is appropriate for ACP and I think the paper should be published after it has been revised according to my recommendations below.

Most of my recommendations are stylistic in nature, but I have one major concern that needs to be addressed. The authors seem convinced that the SO₂ is mainly of Chinese origin, which they base on the FLEXPART results and the measured SO₂/NO_y

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ratio. FLEXPART does show that the plume comes from China, but it also comes from Korea and Japan. While China has a higher SO₂/NO_x emissions ratio, the authors have provided no evidence that this ratio should be preserved or be proportional to the SO₂/NO_y ratio measured by the Falcon. With all the various SO₂ and NO_x removal processes that can be encountered during 8-10 days of transport, why can't Japanese emissions with a low SO₂/NO_x ratio undergo removal processes that remove a greater proportion of NO_x (or NO_y) than SO₂, which would increase the SO₂/NO_y ratio? If the authors can't rule out this possibility then they have no compelling evidence that the plume is mainly of Chinese origin. All they can say is that it is most likely from East Asia, and that it likely has a large influence from China. The title should be changed to read "East Asian SO₂ pollution....";

The abstract states that the DLR mission was part of INTEX but according to: Overview of the summer 2004 intercontinental chemical transport experiment - North America (INTEX-A) Author(s): Singh HB, Brune WH, Crawford JH, et al. Source: JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES Volume: 111 Issue: D24 Article Number: D24S01 Published: DEC 16 2006

INTEX was a NASA study conducted over North America during July 1-August 15, 2004. The DLR mission in the present study does not fall under the INTEX scope, but it may fall under the ICARTT umbrella experiment as described in:

International Consortium for Atmospheric Research on Transport and Transformation (ICARTT): North America to Europe - Overview of the 2004 summer field study Author(s): Fehsenfeld FC, Ancellet G, Bates TS, et al. Source: JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES Volume: 111 Issue: D23 Article Number: D23S01 Published: DEC 14 2006

page 1380 The authors have inaccurately portrayed the results of Arnold et al 1997, which concluded that the SO₂ measured by the Falcon most likely came from the northeast USA, and only said that perhaps it came from metal smelting. But in the

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present study the authors interpret Arnold et al. 1997 as concluding that the SO₂ probably originated from a Canadian NI-Cu smelting industry complex near Sudbury. There is no evidence from Arnold et al [1997] that Sudbury was the source (Sudbury isn't even mentioned) and the summary of the Arnold et al findings needs to be corrected.

page 1380 line 6 Here the authors state that they have found an even more extreme case of long range SO₂ transport, which seems to imply that the SO₂ measurements in 2004 were far greater than those observed by Arnold et al. 1997. In fact the SO₂ in Arnold 1997 was far greater (3 ppbv), so the authors need to be clear that the extreme nature of the 2004 measurement is the distance traveled. When discussing Arnold et al please mention the SO₂ mixing ratios observed.

page 1386 line 11 I don't agree with the interpretation of Figure 7 The authors state that most of the air masses that contributed to the plume originated in regions with SO₂/NO_x ratios of 1-3 mol/mol. But to my eye it looks like only about half of the regions have this ratio. Many locations such as Japan, Korea and southern China have ratios less than 1. I think that the authors are placing far too much emphasis on linking the measured SO₂/NO_y ratio to the emitted SO₂/NO_x ratio. Given the long travel times and all the various removal processes of SO₂ and NO_x, I find it very unlikely that the measured SO₂/NO_y ratio can be used to effectively indicate air mass source region. And on page 1386 line 14 the authors state that China is the main source of the SO₂, but they have not provided enough evidence to rule out Korea or Japan. Please sum up the SO₂ tracer in Figure 7 for China, Japan and Korea and report the percent of the SO₂ from each country. FLEXPART is the only tool you have to indicate whether China is the main source, and even then the model results are subject to transport errors. I think all you can say is that east Asia appears to be the most likely source of the SO₂, and that the FLEXPART tracer and the measured SO₂/NO_y ratio imply China would contribute more than Japan or Korea, but I don't think you can definitely say that China is the main source. Please change the title and conclusions accordingly.

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Page 1387 and Figure 10 It seems from the discussion that the SO₂ tracer is from North American as well as Chinese sources. Please make this point clear in the text.

page 1378 line 17, change to SO₂ stems from fossil fuel combustion, The NI-CU smelting....

page 1378 line 19, change to compounds such as dimethyl...

page 1379 line 6 please be clear that to form acid rain SO₂ must first become sulfate. Also please mention that sulfate, in addition to affecting the Earth's albedo through cloud formation also has a direct effect on the albedo.

Page 1381 all of the relative error and accuracy values are reported as positive values which implies a positive bias. Do you mean to use +/- instead of just + ?

Page 1381 Was O₃ not measured on this flight?

page 1379 line 6 planet's

page 1379 line 23 Jacob et al 1999 is cited as an example of measured pollution transported from Asia to North America. But this was purely a modeling study and did not discuss any measurements of transported pollution plumes. Good examples would be papers from the 2002 ITCT experiment such as:

Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) and Pacific Exploration of Asian Continental Emission (PEACE) experiments: An overview of the 2002 winter and spring intensives Author(s): Parrish DD, Kondo Y, Cooper OR, et al. Source: JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES Volume: 109 Issue: D23 Article Number: D23S01 Published: NOV 12 2004

A case study of transpacific warm conveyor belt transport: Influence of merging airstreams on trace gas import to North America Author(s): Cooper OR, Forster C, Parrish D, et al. Source: JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES Volume: 109 Issue: D23 Article Number: D23S08 Published: JUL 1 2004

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Gas-phase chemical characteristics of Asian emission plumes observed during ITCT 2K2 over the eastern North Pacific Ocean Author(s): Nowak JB, Parrish DD, Neuman JA, et al. Source: JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES Vol-ume: 109 Issue: D23 Article Number: D23S19 Published: JUL 21 2004

page 1380 line 1 say mesoscale convective systems mainly in summer; as such systems can also occur in spring, and deep convection can occur year round in some places

page 1383 line 9-10 change to running mean over 30 data points;

page 1383 line 16 using the word preferably implies that the plume preferred its origin to be China. Instead say stemmed mainly from China;

page 1383 most of lines 18-21 is not necessary because the figure description should be covered in the figure caption

page 1384 line 14 as the source

page 1385 line 15-16 change to This column residence time can also be thought of as the approximate probability that an air parcel reached the measurement location.;

page 1385 lines 18-20 delete this sentence as the SO₂ release location is not indicated by Figure 6a but by 6c

page 1385 line 22 actually the footprint isn't averaged over the lowest 150 m but summed over the lowest 150 m.

page 1385 line 25 change preferably to mainly

page 1386 line 7 is of Asian origin

Table 3 Which year are these data from?

Figure 1 Please indicate the portion of the flight track that contained the Chinese SO₂

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plume

Figure 4 for SO₂ use the same units as in Figure 3

Figure 6 The images are far too small and the map is barely visible. Please improve accordingly.

Figure 8 replace footprint with location

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9, S134–S139, 2009

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