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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Rapid intercontinental air pollution transport associated with a meteorological bomb" *by* A. Stohl et al.

Anonymous Referee #1

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General Comments
The authors present a case of rapid intercontinental transport of air pollution associated with a meteorological bomb, which occured in November 2001. The transport is simulated by a lagrangian model and was directly observed as enhanced NO2 by the GOME instrument. The paper explores the origin of the NOx (lightning vs anthropogenic) and a climatology of how such rapid long range transport events affect transport of short lived species.

Overall this is a well written paper, well organized and dealing with an interesting topic. To my knowledge it is the first time that meteorological bombs have been linked to transport of short-lived pollutants. I have concerns on three central points. The first point regards the confidence in the detection of NO2 enhancement by GOME and the influence of clouds, the authors have not provided enough information to assess the

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possible enhancement of NO2 columns due to interference by clouds. Figures 3 and 8 demonstrate that clouds were indeed pervasive. The second point deals with ruling out lightning as a source of the observed NOx. With the information presented in the paper, I do not think that the authors have convincingly ruled out lightning (the dataset used for lightning was very limited in time and space). My final concern (which is similar to the referee 2's concern) regards the likely overestimate of the effect of anthropogenic pollution as the authors haven't taken into account that only a small fraction of the NOx produced in the U.S. will be exported as NOx. More details on these three points are given below.

Specific Comments
(1.) GOME NO2 enhancements and cloud interference.

The NO2 columns were not corrected for clouds and scenes with large cloud fractions have been excluded. The authors themselves aknowledge that a 10the retrieved NO2 column (which is the type of enhancement in columns in the meteorological bomb transport case!). Figures 3 and 8 show the extensive presence of clouds. Based on Figure 3 I would have expected the areas south-east of Hudson bay (location of C2) and at -40E longitude (along the cloud band of the bomb) to have been excluded from the GOME analysis, yet they appear not to have been masked on Figure 5b. Were these high thin clouds?

The authors claim that most of the relevant scenes were cloud free. Yet, by comparing Figure 8 and Figure 5d it looks to me like the enhanced NO2 off of Iceland is located right were there is a cloud band. Could it be that it is the cloud that is detected and not these high NO2 concentrations? If the clouds were below the NO2 then the columns might be largely overestimating the actual NO2. I suggest a more in depth investigation and discussion of possible cloud contamination for the enhanced NO2 over the oceans.
(2.) Lightning.

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Interactive Comment

The authors rule out the lightning source based on very limited information on lightning flashes. Why only consider the region north of 40N and east of 100W? I would expect most of the lightning activity (if any) to occur further south over the warm waters of the gulf stream, gulf of mexico, or in marine thrunderstorms. Jeker et al. [JGR, 105, 3679-3700, 2000] discussed such situations for November 1997, and presentes observations of the resulting high NOx produced over the North Atlantic. Similarly observations of high NOx levels over the N. Atlantic (and low levels of anthropogenic tracers) were observed repeatedly during SONEX and indicate a large influence of lightning over that region in the fall [Liu et al., 1999; Thompson et al. 1999; Allen et al 2000; Crawford et al. 2000].

The lightning observations from the NLDN network would only be able to capture a few percent of this lightning over the water, but satellite observations possibly could observe this. Have the authors investigated whether satellite lightning information is available for their time period? If no data is available for that time period, an estimate of lightning flashes for November from the OTD detector could be a useful addition.

Given the limited amount of information used, the authors' estimate is thus likely to be a (possibly large) underestimate of the lightning influence. The authors haven't made a strong enough case to say that 'lightning can be ruled out as the main source of NO2 detected by GOME', as stated in the text and conclusions.
(3.) Anthropogenic influence.

The flexpart model is a powerful tool and allows to highlight many of the detailed characteristics of transport. However it does not include any chemistry and assumes a simple 2-day lifetime for NOx. Thus any conclusions in terms of magnitude of the NOx transported should be taken with big caveats. This applies in particular to transport from antropogenic NOx sources.

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As stated by the other referee, most of the NOx emitted over the U.S. will have a short lifetime in the boundary layer (12 hours) and thus will be rapidly converted to HNO3 and deposited nearby. A number of modeling studies have examined the amount of NOy exported from the U.S. boundary layer. In particular, Liang et al. [1998] have shown that during the fall about 10l would expect the results from the flexpart model to largely overestimate (perhaps by one order of magnitude) the amount of NOx exported from the boundary layer, especially as the simulated NOx is already much too high in the boundary layer over the U.S. compared to the GOME observations. As suggested by the other referee one way to improve the calculation would be to implement a 10the free troposphere.

Thus, given the limitations of the tool used and lack of corroborating observations (such as observations of enhanced concentrations of other tracers of anthropogenic influence), I don't think that the authors can justify their conclusions as to the unambiguous assignment of the enhanced NO2 to transport of anthropogenic pollution. This also extends to their assessment of the 2-3 pptv NOx enhancements due to these express highways. These estimates are highly uncertain.
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Interactive comment on Atmos. Chem. Phys. Discuss., 3, 2101, 2003.