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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.

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(1) GOME NO_2 enhancements and cloud interference

On 8 November, the densest clouds were actually located in those regions not overpassed by GOME. Thus, no extensive cloud masking was applied, but it can be seen that clouds were masked out east and south of Greenland (around 40 W).

On 10 November, the filament was in a region with relatively little clouds. An exact comparison between Figures 5d and 8 is not possible, because GOME passes over any location at about 10 a.m. local time, whereas the infrared image shown is from 12 UTC. Because of the rapid synoptic evolution, cloud scenes change rapidly, rendering a direct comparison between clouds from geostationary satellites and the plume position from GOME unreliable. Therefore, the cloud masking was based directly on cloud detection by GOME.

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Interactive Discussion
Discussion Paper

Considering the potential influence of clouds on the NO_2 observations two major effects have to be considered: a) NO_2 below or deep inside the cloud is shielded, and b) NO_2 (directly) above the cloud is enhanced.

Thus, it is not a priori clear whether clouds lead to an over- or underestimation of the NO_2. To quantify the errors introduced by these two effects, the exact vertical distributions of both clouds and NO_2 would have to be known at an accuracy that cannot be achieved using the data at our disposal.

Therefore, we carried out a sensitivity study for a worst-cases scenario for effect b) assuming a thin NO_2 layer immediately above a layer of clouds at 3-5 km altitude. This scenario yields an overestimate of NO_2 by our retrieval algorithm of less than a factor of 2, not enough to explain the observed NO_2 plume. Note also that, due to the cloud masking, maximum actual cloud cover in the pixels shown is 50%, thus reducing this maximum possible cloud effect. An independent argument against a large NO_2 overestimate due to clouds is that the strongest NO_2 signals are not seen above the densest clouds, but over pixels with relatively little cloud cover.

Even though the exact vertical distribution of clouds and NO_2 are both unknown, it is very likely that clouds formed in the very same airmass that was lifted from the surface and contained the NO_x. Thus, most of the NO_x would likely be in-cloud, rather than above-cloud. In this case, effect a) could even lead to an underestimate of the NO_2 columns.

In summary, we agree with the reviewer that clouds lead to inaccuracies in our NO_2 retrieval, but we can rule out that the observed filament is entirely (or to a very large extent) a cloud artifact. It may even be possible that, because of effect a), we have underestimated the NO_2 vertical columns.

We will present parts of the above discussion in a new subsection of the revised paper version.

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(2) Lightning

NLDN data is not normally available for free, and therefore we had to restrict our analysis to a limited region. However, the region (which is larger than the section shown in Figure 9) was choosen such that it covers the main inflow to the express highway. As explained in the paper, there was some lightning, but by far not enough to explain the observed NO_2 amounts.

We have only very limited information about off-shore lightning. No OTD data were available, but LIS data are available for the region south of about 40^o N. There was indeed some lightning detected, associated with the hurricane moving north. However, according to the limited data available from LIS, the lightning appears to not have been particularly strong. We have performed an additional lightning tracer simulation using the daily positions of the hurricane from 2-4 November, and we found that the lightning NO_x produced in the hurricane was transported far south of the express highway, incompatible with the detected NO_2 filament. We will describe this additional lightning tracer simulation in a short paragraph in the final version of the paper. Therefore, our conclusion remains that we have ruled out lightning as a possible source of the NO_x.

(3) Anthropogenic influence

We completely agree that FLEXPART cannot be used to reliably determine the amount of NO_x from anthropogenic sources transported during this event (or in the climatology), and this was not the aim of this study. All the model was used for was to show that anthropogenic emissions do indeed enter the express highway (and in quantitities compatible with the observations), and that, to the best of our knowledge, other plausible sources for the NO_x observed in the express highway can be excluded. All we then want to say is that the observed NO_x must be due to anthropogenic emissions in North America. Thus, for the purpose of this paper, it is not very important whether we assume a 2-day lifetime of the NO_x, or assume a shorter lifetime, or take a fixed percentage of the NO_x leaving the boundary layer (which by itself is highly variable,

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and we thus do not see how we can improve the accuracy of our simulation without explicitly simulating the chemistry!). Any model-based quantification of the amount of NO_x actually transported should use chemistry transport models. We definitely would welcome simulations of this event with chemistry transport models!

However, we take your (and the other reviewer's) point and will re-phrase various parts of the manuscript, in order to more clearly show the limitations of our study.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 2101, 2003.