The paper on "Systematic analysis of tropospheric NO2 long-range transport events detected in GOME-2 satellite data" by Zien et al. presents a comprehensive analysis of long range transport events over oceans from 5 years of satellite observations of NO2.

While individual LRT events have been investigated as case studies before, the study of Zien et al. provides, to my knowledge, the first systematic analysis of LRT of NO2 from satellite data. The paper is well written and considers various aspects of NO2 LRT like frequency, meteorological conditions, or NO2 fluxes, both on the basis of selected case studies as well as for statistical means. Nevertheless, some aspects remain unclear or inconsistent.

I recommend publication on ACP after considering the major and minor issues listed below.

1. Major issues

Impact

The study is performed thoroughly and comprehensively. However, it is quite descriptive in focus.

For publication in ACP, I am missing a discussion of the impact of LRT on atmospheric chemistry and ozone production over oceans and arctic regions. The authors should extent the respective discussion qualitatively, and might even think of ways how to become more quantitative in their conclusions.

NOx/PAN

NOx combines NO and NO2, with varying NO2/NOx ratios, and NOx is also converted (temporarily or eventually) to other nitrogen containing species (NOy), in particular PAN. This is mentioned in the introduction, but especially the discussion of PAN is rather short, while PAN is probably a key player for the LRT of NOx.

Conversion between NO and NO2 and between NOx and NOy affects the NO2 signal observed from satellite. E.g., while an uplifted plume might contain only few NO2 (but a lot of NO and PAN), the total NO2 might increase if the plume is sinking down due to PAN decomposition and the shift of NOx from NO towards NO2. Also the effective NOx lifetime might be considerably longer than 4 days due to temporary conversion into reservoir species.

These effects have to be discussed and kept in mind for the interpretation of e.g. the day-to-day changes of total NO2, the calculation of fluxes, etc.

Cloud data

The authors mention FRESCO+ cloud data. In their study, however, they calculate a cloud fraction on their own. I do not understand the motivation for this procedure.

As the authors note, "even small cloud fractions have a strong impact on the air-mass factor". Especially for low cloud fractions, the determined CF value according to eq. 6 strongly depends on the a-priori cloud-free reflectivity, which is just taken from MERIS without further discussion.

The authors should clearly motivate their choice of an "own" cloud product, need to discuss its uncertainties, have to compare it to FRESCO cloud fractions, and should discuss reasons for and impact of possible differences.

Oddly enough, in section 6, FRESCO cloud fractions are used instead of the CF from eq. 6, which is quite inconsistent.

In addition to cloud fractions, FRESCO+ provides cloud pressure as well. This information is not considered at all in this study. However, if the NO2 plume is actually located inside the cloud, as assumed, the cloud pressure directly provides NO2 plume altitude information! This should be discussed, and the FRESCO cloud pressure for the identified plumes has to be compared to the plume heights inferred from back-trajectories.

Lightning

The authors discuss lightning as kind of a side phenomenon which sometimes occurs but is generally irrelevant. While this is probably true in general (i.e. anthropogenic NOx emissions are far higher than the NOx produced from lightning), situations might be systematically different for the investigated LRT events. In this context, it would be very helpful to include lightning observations from continuous, global lightning networks like WWLLN in the systematic analysis.

Wenig et al., 2003, report on thunderstorms coinciding with the transport event originating in South Africa. The same is the case for the example discussed in 5.2: Figure 1 displays the flashes detected by WWLLN, which are coinciding with the NO2, at least on July 9. In addition, FRESCO CP (Figure 2) reveals very high clouds South from Madagascar (far above the plume heights given in table 1).

Also for the case study discussed in 5.3, WWLLN detects a considerable amount of flashes, also over land (see Fig. 3), which coincide well with the NO2 plume.

Besides the production of LNOx, which is indeed hard to quantify and cannot easily be discriminated from the LRT NOx, the role of convective systems, e.g. for the initial uplift of BL NOx into the free troposphere, or the impact on the accuracy of the back-trajectories, has to be discussed.

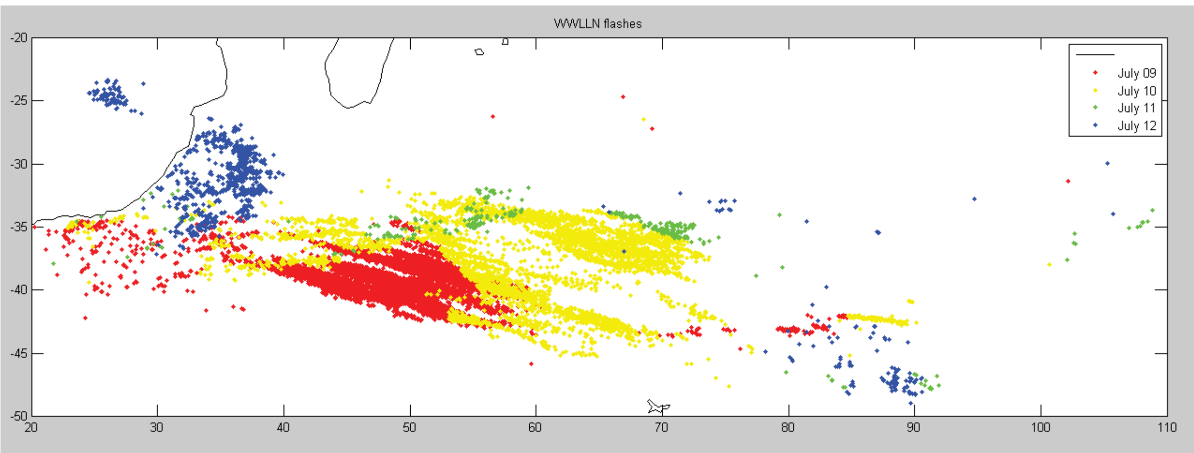

Figure 1: Flashes detected by WWLLN for July 9-12 2008.

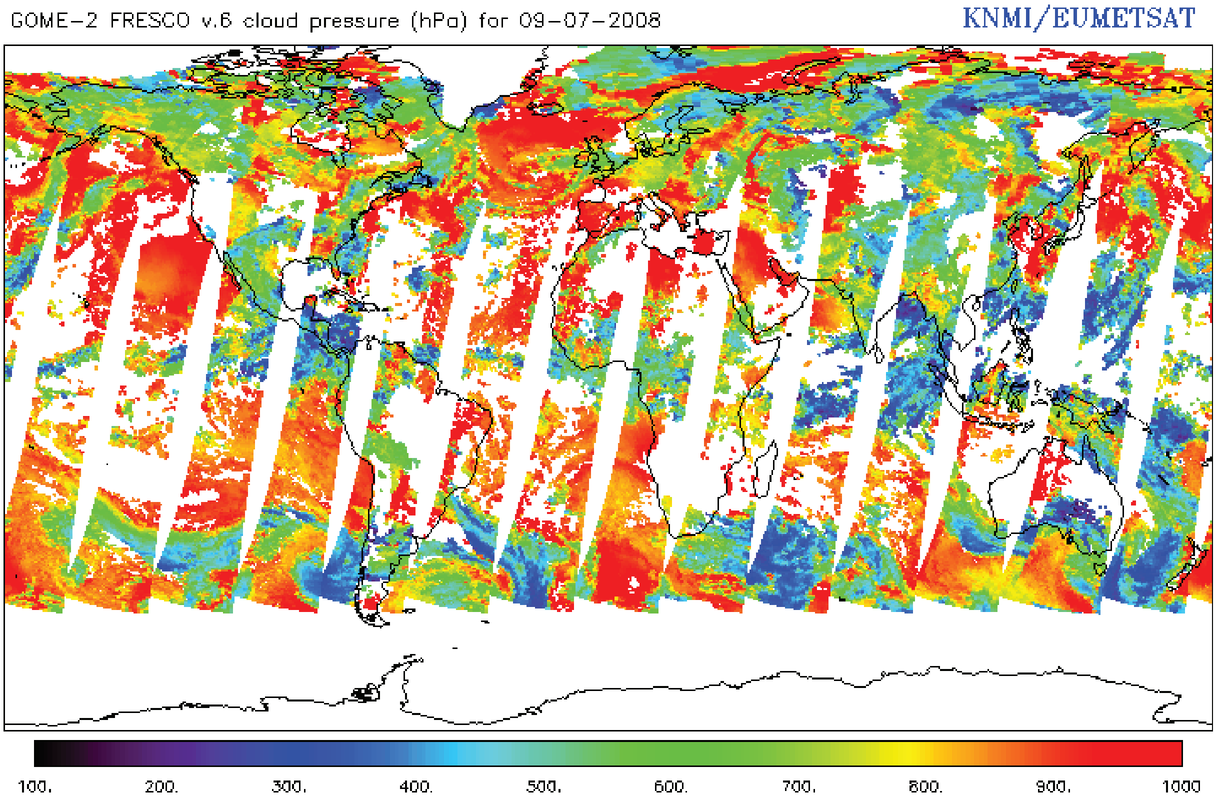

Figure 2: FRESCO cloud pressure on 9 July 2008.
Source: http://www.temis.nl/fresco/gome2_v6/2008/20080709_p.gif

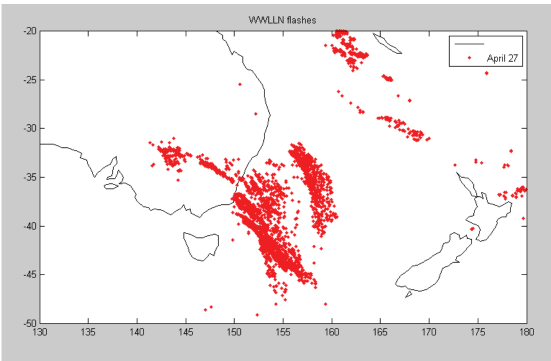

Figure 3: Flashes detected by WWLLN for April 27 2008.

2. Minor comments:

30947/15: Add power plants.

30947/24: The NOx lifetimes reported by Beirle et al., 2011, are considerably shorter than 8 hours for most Megacities.

30948/5: “up to four days” → “up to several days”

30948/6: “due to lower concentrations of radical species” – and due to higher NO/NOx ratios!

30948/7: “For it to occur” → “For its occurrence”

30948/21: PAN plays probably a key role for the long-range transport of NOx. Thus it should be discussed in more detail and perhaps also earlier in this paragraph.

The conversion of NOx into PAN (and back) also hampers the deduction of the NOx lifetime.

30949/1: What is “common satellite data”?

30949/15-17: The discussion of NOx effects on Ozone is quite short and vague.

Secion 2 is quite detailed and might be shortened. E.g. the explanation of DOAS (30951/24-30952/17) might be replaced by a reference to Richter et al., 2011.

30955/5: The reference to Eskes and Boersma in this context is strange, as in this study, clouds are treated as Lambertian reflectors, i.e. multiple scattering effects are ignored! There are several other studies which have discussed the different cloud effects, and show Block AMFs similar to Fig. 2.

30958/22: Ships are “concentrated” NOx emitters on the open ocean!

30959/13: “developed”

30960/3-4: This is a too absolute statement: There might be reasons for plumes over oceans without LRT, e.g. a burning oil platform, or strong thunderstorms.

30960/8: n_{seed} \times \sigma

Figure 4 is meant to illustrate the selection procedure, but misses several aspects. The identification of plumes consists of “seeds”, which are either “merged” or “discarded”, with additional “iterations” and changing thresholds (n_seed versus n_member). All these steps and the different pathways for candidate pixels should be illustrated examplarily.

30960/25: Which kind of instrumental artefact could be interpreted as a LRT plume?

30960/4: FRESCO CTP yields information of the cloud altitude, which is assumed to be the same as the NO2 plume altitude.

30962/3: Which area was chosen for averaging?

30962/6: “All plumes … are discarded”: It would be interesting to know how many plumes have been discarded by this criterion.

30962/12: 10e15 molec/cm2 is actually *larger* than the threshold value given in line 4!

30964/1: To avoid misunderstandings, I propose to add “as long as the NO2 plume is within the cloud”.

30964/12: FRESCO provides cloud altitude information!?

30965/7: “high”→”higher”

30965/12: “we perform”→”performed”

30965/14: “eventually”→”possibly” or “probably”

30965/17: “so that stray pixels …”: I propose to skip this.

30967/15-16: I do not understand why high emission rates and long lifetimes should be obstructive for the observation of LRT over several days!?

30967/26: which is difficult anyhow due to changing NO/NOx, formation and decomposition of PAN etc.

30969/1-3: There are many possible explanations for increasing NO2, e.g. LNOx, conversion of NO into NO2, or decomposition of PAN.

30970/23: “deceleration”

30970/26: Please reformulate this sentence.

30972/19: Values for m’ are derived for each season, but these numbers, their meaning, and potential impacts are not discussed at all.

30975/10-11: add “… by creating a similar map (Fig. 16)”, and skip the last sentence of the paragraph (line 14).

30975/20: Given the uncertainties of the back-trajectories as discussed in the following paragraph, I see the discussion of “bush fires” as significant sources as rather speculative. According to long-time means, the Highveld area and Johannesburg are by far the dominating source regions over South Africa.

30979/9: Isn’t that negative anomaly caused automatically by the algorithm? On day+1, the mean is calculated from the days before and after, including day0.

30979/25: “Fig. 23 shows no NAO characteristics”: I do understand this statement; Fig. 23 shows a very clear dipolar pattern!?

30980/1-11: Please add a figure of the discussed cloud fraction anomaly.

Fig. 6: “indicated by purple circles”: → add “in the center and right columns”.

Fig. 19: The observed NO2 flux does not have to correspond 1:1 to the emissions at ground, thus I recommend to change the y-axis label.