Norrköping, 27 June 2017

To,
Prof. Thomas Wagner,
Editor, ACP

Sub: Revised submission of acp-2016-1091.

Dear editor,

We thank you and the two referees for the review comments. We believe the referee comments were very helpful and constructive. We have already uploaded response to the referee comments in the online forum. We are hereby submitting the revised manuscript that hopefully addresses referee concerns. The manuscript has undergone a major revision. The following major changes/investigations were done in response to the referee comments.

1) The entire analysis is revised and the selection of extreme events is now based on monthly thresholds instead of seasonal ones. This is to take into account the fact that NO_2 has strong monthly variability over the study region. Correspondingly, the figures are also revised.
2) The analysis was also repeated using only clear-sky retrievals to check the sensitivity of selected extreme events to cloud screening. A new section and three figures are added to discuss the results of this sensitivity study.
3) The parts of Introduction, Results and Conclusion sections are revised to strengthen and clarify the importance and value addition of the present study,

The changes made in the revised manuscript are highlighted by giving yellow background. We would be happy to work and improve the manuscript further, if needed. We look forward to receive feedback from you and the referees.

Thanks and regards,
Manu Thomas

Norrköping, 31 May 2017

Response to Referee #1

We thank the referee for constructive comments that lead to substantial improvements in the revised manuscript. Based on comments by both referees, a major revision of the manuscript is carried out. In particular we have tried to address the following three concerns.

1) The entire analysis is revised to investigate the sensitivity of our results to using OMI retrievals under cloud-cleared versus cloudy conditions. The analysis of AIRS and reanalysis datasets is also revised accordingly.
2) The revised analysis is now based on individual months (instead of seasons) to take into account even the monthly variability.
3) A stronger case is made to clarify precise contribution of the present work.

Please find below point-by-point reply to your general and specific comments.

In this manuscript, Thomas and Devasthale present an analysis of the meteorological conditions associated with high NO2 concentrations over Scandinavia (as inferred by satellite NO2 columns from the OMI instrument). They find that the highest NO2 in each season is predominantly associated with south-westerly winds, and identify a
clear 'transport pathway' using ERA reanalysis. The meteorological conditions tended to persist for either 1 day or 3-5 days, the latter potentially allowing for the longest range transport.

General Comments:

Overall, the paper is well-written with good consideration of related work. The methods are mostly well laid out, and the results are presented clearly. However, I have a couple important concerns.

We thank the referee for encouraging comments.

My main concern is that it is not clear what value has been added from this analysis. As the authors state, the dominance of south-westerly winds during "extreme" pollution is consistent with pollutant transport discussed elsewhere. What has been learned from this particular analysis of satellite NO2 columns in combination with ERA reanalysis data? What new concepts or ideas have been presented? In my opinion, in its current form the scientific significance of this paper is bordering on poor. For publication in ACP, the authors must make a stronger case for the novelty of their findings.

We agree that we had not made our contribution clear enough for readers. Below we would briefly like to point out our value addition.

1) First of all, we are not aware of such detailed, observationally constrained analysis of weather patterns associated with hundreds of extreme NO2 pollution events over Scandinavia, especially by making synergistic use of satellite and reanalysis datasets.
2) We have quantified relative importance of different wind directions during these events in various seasons.
3) We have also investigated the persistency of weather patterns during extreme events and which wind directions explain the observed modes in the persistency.

All of these results and statistics presented here will be helpful for chemistry and tracer transport models while studying the impact of norward long-range transport of pollutants.

Hopefully, the revised text will make it clear for the reader.

My secondary concern is that they tend to analyze the meteorological conditions during the "extreme" conditions only, without showing us that these conditions are unique to the "extremes" (and thus a defining factor in extreme pollutant concentrations for the region). In other words, the authors haven't clearly shown that the patterns associated with "extreme" NO2 are any different from patterns under more typical pollutant concentrations. What, then, is gained? The "clear transport pathway" could still exist under non-extreme conditions, in which case an explanation for extreme concentrations would need to look at other factors. I suggest the authors prove that these meteorological conditions tend to be specific to extreme NO2 by explicitly showing that the meteorological conditions for the other 90 percent of the time look different.

The aim of the study was to analyse meteorological conditions during extreme pollution events, ***irrespective*** of the fact whether these conditions turn out to be different or not compared to climatology. We respectfully disagree that nothing is gained if they don't turn out to be very different. It is in fact equally important to point out how challenging it will be for chemistry and tracer transport models to disentangle typical patterns associated with extreme events.

A more semantic concern: I am not a fan of their use of the term "extreme" throughout the manuscript. Given their definition of an "extreme" event (90th percentile concentrations in each individual season), an "extreme" event in July is nothing like an "extreme" event in January. The analysis is therefore a bit confusing in terms of potential relevance or impact. I wish they would use something a little more qualified throughout their manuscript. For example, could they say "the highest summertime concentrations" instead of "extreme NO2 concentrations".

Since the revised analysis is based on applying thresholds on individual months, the chosen extreme event is representative of that particular month. We appreciate the fact that an extreme event in Jan is different than in July and that is why we have chosen percentile matric instead of a fixed threshold on the absolute values of NO2. We believe such 90%ile threshold will allow the investigation of meteorological conditions in a fair manner during extreme events.

Specific Comments:
Lines 39-41: Why not include soils amongst the other sources of Nox?

Included.

Line 43: One rarely hears NO2 referred to as a strong oxidizing agent with respect to atmospheric chemistry. I suggest rephrasing.

This line is rephrased.

Line 52: I suggest the authors include a more recent projection, given the Lamarque et al. 2005 result is based on IPCC SRES A2 scenario which has very different Nox emission projects to 2100 from, say, the more recent RCP scenarios.

Included.

Line 100: Please provide a link to the direct source of the OMI data.

The OMI data were obtained from NASA's GES DISC. The following link is included in the revised version.
https://disc.gsfc.nasa.gov/Aura/data-holdings/OMI/omno2d_v003.shtml

Line 102: "Under cloudy conditions": Please clarify that this means you have not applied any cloud fraction threshold for quality assurance. More importantly, I understand the motivation for this was to include more data (and avoid bias), but these observations result in very high uncertainty. Zien et al. (2014) go through quite a bit of detail explaining their unique treatment of cloudy data, and propose a new computation for the air mass factor calculation in these cases. However, if the authors of the present study are simply using the standard product retrieval (OMNO2d V.3), I expect the cloudy observations to have little realistic meaning. In the best case, over polluted regions with very low cloud fraction, the NO2 tropospheric retrieval still has between 35-60% error (Boersma et al. 2004). This will be much higher for cloudy data. While the authors have made a good case for including the observations, they have not discussed how the poor quality of such observations could impact their results. I.e., what good is a lot of data, if most of it is bad? This must be addressed.

This is indeed a good point. However, purely from the user point of view, we find it difficult to see how we can improve our current analysis of satellite data. We have done our best to follow recommendations for the users. We appreciate that even the clear-sky retrievals could have large error bars, but based on the sensitivity analysis (please see response to another reviewer) and the consistency of spatial distribution of NO2 during all-sky and clear-sky conditions, we are confident that there aren't major artefacts affecting our analysis of OMI data.

Line 110: Please provide a link to the direct source of the AIRS data.

The AIRS daily gridded data (AIRX3STD) were also obtained from the NASA's GES DISC AIRS Holding. The following link is provided in the revised version.
https://disc.gsfc.nasa.gov/uui/search/%22AIRS%22

Line 113: Please provide a link to the direct source of the ERA reanalysis data.

ERA reanalysis data were directly downloaded from the ECMWF's data server.
http://apps.ecmwf.int/datasets/data/interim-full-daily/levtype=sfc/

Line 130: Please explicitly state here how the seasons were defined (DJF, MAM, JJA,SON).

This is now clarified. Please note that while the revised analysis is based on individual months, the results are presented for seasons to avoid too many smaller subplots.

Line 137: "It is interesting to see that...": I would argue that this is not at all interesting, but an obvious outcome of their design, given the definition of "extreme" and theseasonality of NO2.

Here we merely wanted to point out that, due to long-tailed distribution of NO2 in DJF, the chosen threshold for the winter months is higher than the one in the summer months.

Line 141: The authors note a "bimodal peak" in the distribution of NO2 extreme events. However, the definition of the seasons (DJF, MAM, JJA, SON) is extremely important here. As they have previously stated, NO2 has strong seasonality, peaking in the winter. So, if "spring" is defined as March-April-May, it is not surprising that most of the 90th percentile values for NO2 in this season will occur in March (the closest winter month). Likewise, if the fall is defined as Sept-Oct-Nov, it is obvious that most of the 90th percentile values for NO2 in this season would occur in November (the closest winter month). Thus, the "bimodal" shape of Figure 2b is almost certainly an artifact of their experimental design. I therefore don't understand what we learn from this figure, and suggest removing it, and any discussion of it.

The Fig. 2b is dropped in the revised manuscript.

Line 183: Are the specific humidity anomalies calculated with respect to a long-term mean? Or with respect to the other 90% of the NO2 concentration days? Please define how the "anomaly" has been calculated (this would be most appropriate in the Methods section).

Yes, the specific humidity anomalies are computed with respect to a long-term mean. This climatological mean is subtracted from the average humidity anomalies observed during extreme events.

Line 203: "higher wind speeds.. during extreme events compared to climatological conditions": Where are the climatological conditions shown? As I mention above, I suggest including figures of the climatological meteorology in all cases, for a more obvious comparison to the "extreme" conditions.

Please note that Figs. 7a and 7b also show wind distribution under climatological conditions (dotted lines). Furthermore, the numbers in brackets show average wind speeds under climatological conditions as a reference.

Line 209: If I wanted to reproduce this data, how was "persistency" actually determined? I.e. what computational steps were performed on the meteorological data to evaluate this.

The persistency is defined as follows. If an extreme event is observed on any particular day, the wind strength and wind direction is computed from the u and v vectors. We then check how many days back in time this particular wind direction is sustained over the centre of the study area.

Norrköping, 31 May 2017

Response to Referee #2

We thank the referee for constructive comments that lead to substantial improvements in the revised manuscript. Based on comments by both referees, a major revision of the manuscript is carried out. In particular we have tried to address the following three concerns.

4) The entire analysis is revised to investigate the sensitivity of our results to using OMI retrievals under cloud-cleared versus cloudy conditions. The analysis of AIRS and reanalysis datasets is also revised accordingly.
5) The revised analysis is now based on individual months (instead of seasons) to take into account even the monthly variability.
6) A stronger case is made to clarify precise contribution of the present work.

Please find below point-by-point reply to your general and specific comments.

In their manuscript "Typical meteorological conditions associated with extreme nitrogen dioxide (NO2) pollution events over Scandinavia", Thomas and Devasthale report on a study evaluating the meteorological conditions under which the highest tropospheric NO2 columns are observed in OMI data over Scandinavia. Their results show, that such events are linked to situations in which transport from the polluted regions in Europe towards Scandinavia occurs, that such events are mostly observed in winter and spring and that they persist for several days. The topic of the study (impact of meteorology and long-range transport on pollution) is interesting and fits well into ACP. The paper is also well written, clearly structured and to the point.

We thank the referee for encouraging comments.

I have however several concerns with respect to the relevance and also the methodology of the study which need to be addressed before the paper can be considered for publication.

General points

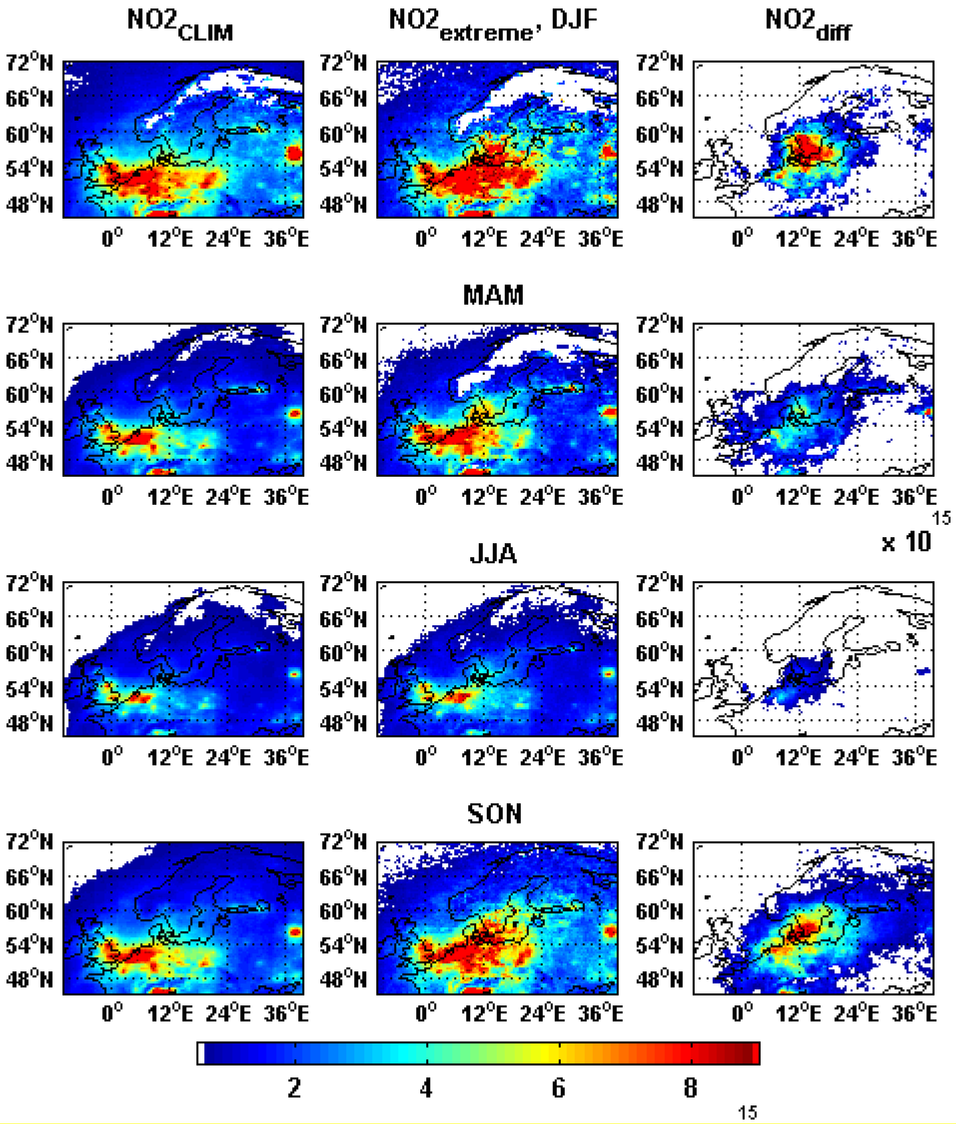
• Probably the most important point is that I'm not sure what the relevance of the results presented in this manuscript really is. It is not surprising that pollution transport from Germany and the Benelux countries impacts on Southern Scandinavian air quality. As there is no attempt made to quantify the impact in absolute
or relative terms, the study does little more than confirming what one would have guessed anyways. I think it would be good to try to become more quantitative in the sense of how many days are affected, what are the mean and maximum anomalies, and what is the relation to pollution from local sources.

Please note that the main aim of the study is to quantify typical meteorological conditions during the extreme pollution events and not to show that the transport from Germany and the Benelux countries impacts on Southern Scandinavian air quality. The latter conclusion, while still interesting since it is based on observationally constrained datasets, is the bi-product from the inferral of the combined analysis of circulation patterns and changes in humidity and cloudiness. The emphasis is rather given on describing meteorological conditions and whether and how they differ from the climatological conditions.

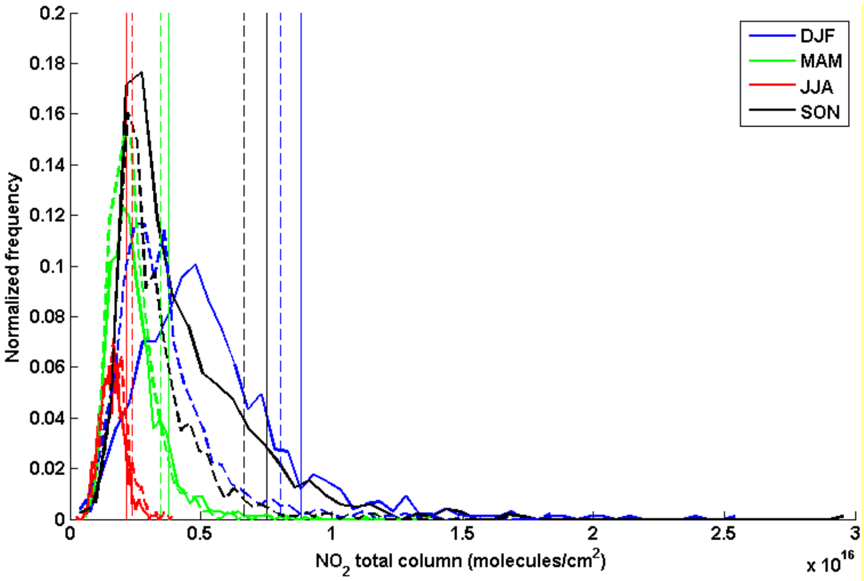
• A second very important point is the use of OMI satellite data without separating cloud free and cloudy situations. While the argument for this approach is clear as many transport events are associated to clouds, such data cannot be easily interpreted as for cloudy conditions, the assumptions made in the retrieval become very important for the results. In the current manuscript, this point is not addressed at all and I think the authors need to investigate differences between cloudy and clear sky averages in order to better understand the impact of clouds on the satellite data. They also need to discuss uncertainties linked to the satellite retrievals.

This is indeed a good point, which is also raised by another referee. We have now carried out sensitivity analysis to check to what extent our selection of extreme events, the spatial distribution of NO2 and corresponding humidity anomalies are affected by restricting to only clear-sky conditions.

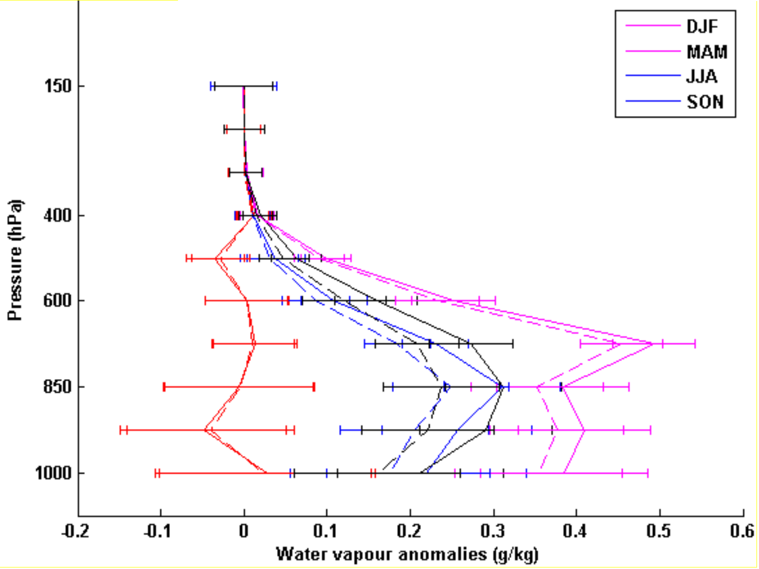
- A figure below shows the spatial distribution of NO2 during only clear-sky conditions for climatological conditions, during extreme events and the differences between the two. This figure can be compared to Fig. 3 in the manuscript. It can be seen that the general distribution of NO2 and its anomalies in the figure below are quite similar to Fig. 3.



- A figure below shows a comparison of the NO2 histograms over the center of the study area. The solid lines show NO2 distribution under all-sky conditions and dotted lines under clear-sky. The differences in the summer half year are minimal. The differences in the winter half year are not large enough to change either the spatial distribution of NO2 as shown above or to affect the tendencies of specific humidity anomalies as shown below.



- We further investigated the vertical structure of specific humidity anomalies under clear-sky conditions. A figure below shows the comparison thereof. Once again it is seen that the general tendency that humidity is increased during the extreme events does not change even under the clear-sky conditions.



These results provide us confidence that our analysis is most likely not affected by the contamination from clouds.

• The tacit assumption made in this study is that NO2 observed from satellite (partly above clouds) is indicative of enhanced NO2 levels on the ground. I'm not convinced that this is always the case during transport events and it would be good to support the timing and location of their extreme NO2 events by at least some surface observations showing that in deed air quality on the ground was also poor during the satellite observed pollution events.

This is also a good point, but we have to admit that addressing this would be out of the scope of the present study. As mere users of satellite data sets we have to put faith in these data sets (and the work done by the respective Science Teams) and hope that the tropospheric columns would capture the variability and coupling between the concentrations in the free troposphere and near-ground. If the referee insists and the extra time is granted by the editor, we would be happy to cross-check with surface observations. We also kindly request the referee if he/she would point out to us relevant references that discuss such discrepancy.

Definition of extreme cases is another critical aspect and I think that given the large seasonality of NO2, monthly thresholds would be better than seasonal thresholds. I'm also a bit confused by the relevance of Figure 2b) showing the number of extreme events per month – isn't the number of extreme events per season constant the ways the authors define their thresholds, and therefore the distribution over months just reflecting the seasonality of NO2?

In the updated version of the manuscript, Fig. 2b is dropped as this was also demanded by the other referee. We have further revised the entire analysis and based it on the monthly instead of seasonal thresholds. Please note that the major conclusions do not change even when we select extreme events based on the monthly thresholds.

- Considering Figure 3, I'm wondering why the situations with higher NO2 over Southern Scandinavia appear to also have higher than normal NO2 over the supposed source regions. Does this mean that under these conditions, pollution is accumulating in general? If this would be simple transport from Central Europe to Scandinavia, I would expect to see less NO2 in the source region or what am I missing here?

Please note that to qualify for being an extreme event over Southern Scandinavia, the pollutant levels in the source regions also need to be higher than usual together with favourable circulation patterns.

Minor points

- Line 40: Add soil emissions

Added.

- Line 47: Does NO2 really affect psychological health?

It has been claimed that it does, but indirectly by affecting physical health and thereby psychological. In the revised version, the reference to psychological health is removed.

- Figure 3: Are these total or tropospheric columns?

Only tropospheric columns are analysed. It is now clarified in the revised manuscript.

- Figure 6: Not sure what is "the same as in Fig. 5" here

This is clearly a mistake. Thanks for pointing it out. The Figure 6 shows the specific humidity anomalies (averages during extreme events minus climatological conditions).

- Figure 7 a / b are difficult to read (too many lines)

We agree that there are too many lines in those figures, but it was necessary to do so to show climatological conditions. For a quick and easy reference to the reader, the average values of wind speeds are also shown in those subplots.

- While the article is overall well written, there are many places in which I would add / remove articles. I therefore recommend another round of proof reading to fix these and other small English problems.

We have tried to fix misplacing of articles in the revised version.

Typical meteorological conditions associated with extreme nitrogen dioxide (NO_2) pollution events over Scandinavia

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Abstract

Characterizing typical meteorological conditions associated with extreme pollution events helps in the better understanding of the role of local meteorology in governing the transport and distribution of pollutants in the atmosphere. The knowledge of their co-variability could further help to evaluate and constrain chemistry transport models (CTMs). Hence, in this study, we investigate the statistical linkages between extreme nitrogen dioxide (NO_2) pollution events and meteorology over Scandinavia using observational and reanalysis data. It is observed that the south-westerly winds dominated during extreme events, accounting for 50-65% of the total events depending on the season, while the second largest annual occurrence was from south-easterly winds, accounting for 17% of total events. The specific humidity anomalies showed an influx of warmer and moisture-laden air masses over Scandinavia in the free troposphere. Two distinct modes in the persistency of circulation patterns are observed. The first mode lasts for 1-2 days, dominated by south-easterly winds that prevailed during 78% of total extreme events in that mode, while the second mode lasted for 3-5 days, dominated by south-westerly winds that prevailed during 86% of the events. The combined analysis of circulation patterns, their persistency, and associated changes in humidity and clouds suggests that NO_2 extreme events over Scandinavia occur mainly due to the long-range transport from the southern latitudes.

1. Introduction

Nitrogen dioxide (NO_2) is one of the highly reactive gases of the nitrogen oxides (NOx) family. The major sources of NO_2 are fuel combustion in motor vehicles, industrial boilers, emissions from soil and agricultural biomass burning. The natural source of NO_2 is lightning and forest fires. Recent studies indicate increasing trends in NO_2 in developing countries and decreasing trends in developed countries as a result of environmental regulation policies (Richter et al. 2005; Zhang et al. 2007; van der A et al. 2008; Schneider et al. 2015; Geddes et al. 2016). NO_2 is an oxidizing agent resulting in the corrosive nitric acid and plays an important role aiding the formation of ozone. It can also contribute to the formation of particulate matter (PM) and secondary organic particles through photochemical reactions. Increased NOx concentrations not only severely affect human physical health through reduced lung function, but also affect aquatic ecosystems through acid deposition and eutrophication of soil and water (Sjöberg et al. 2004; Klingberg et al. 2009; Bellandar et al. 2012; Gustafsson et al. 2014; Nilsson Sommar et al. 2014; Oudin et al. 2016; Taj et al. 2016). According to the fifth IPCC assessment report, the total global NOx emissions have increased by 20% since pre-industrial periods to 324 ppb in 2011. (Lamarque et al., 2013, based on the model-intercomparison project,ACCMIP, assessed increases in regional nitrogen deposition by up to 30-50% from RCP 2.6 to RCP 8.5). In heavily polluted areas NO_2 can also have noticeable impact on the local radiation budget (Vasilkov et al. 2009).

Compared to other pollutants such as carbon monoxide (CO) that has a life span of weeks to few months, NO_2 has a relatively shorter life time in the atmosphere and ranges typically from a couple of hours in the boundary layer to up to few days in the upper troposphere (Beirle et al., 2011). Therefore, NO_2 can be typically associated with short-range transport events. For long range transport (LRT) or intercontinental transport of pollutants and in particular of NO_2 to occur, the associated weather systems need to be linked with stronger winds and rapid convective-advective events such as cyclones or warm conveyor belts (WCBs) that can lift air masses from their source regions up into the free troposphere and be transported across the oceans (Eckhardt et al. 2003; Stohl et al. 2003). Due to lower concentrations of radical species in the free troposphere, the reaction with NO_2 is limited. Zien et al. (2014) identified about 3800 LRT events of NO_2 during a 5 year period from the major pollution hotspots such as the east coast of North America, central Europe, China and South America, predominantly during autumn and winter months.

There have been several studies reporting individual LRT events of NO_2. To mention a few, Stohl et al. (2003) in a study explained "intercontinental express highways" being responsible for almost 60% of the total intercontinental transport of pollutants from across the Atlantic to Europe, resulting in an increment of average European winter NOx mixing ratios by about 2-3 pptv. In yet another study, Schaub et al. (2005) demonstrated that at least 50 % of the NO_2 recorded at the Alpine region was advected via a frontal system from the Ruhr area in central Germany in February 2001. Donnelly et al. (2015) reported that easterly air masses during winter resulted in increased NO_2 concentrations in the urban and rural sites in Ireland. LRT of NOx across the Indian Ocean from South Africa to Australia in May 1998 was reported by Wenig et al. (2003).

The Nordic countries often lie at the receiving end of short-range pollutant transport from northern Europe or they are a part of a much larger transit pathway of eventual long-range transport to the Arctic, originating from either Europe or North America. To what extent such a transport from the southerly latitudes affects the characteristics of extreme pollution events (such as magnitude, frequency and persistence) over Scandinavia depends largely on prevailing circulation patterns and meteorological conditions. The local meteorology can enhance or dampen the concentration of the pollutants depending on the degree of persistency; the knowledge of which would help to better constrain the chemistry transport models (CTMs). Therefore, identifying the dominant weather patterns over Scandinavia especially during extreme pollution is important. However, there has not been a systematic study linking the transport events of NO_2 to different meteorological conditions, solely from observational data over the Scandinavian region. Therefore, the main aim of the present study is to characterize circulation regimes and meteorological conditions extreme pollution events, to understand to what extent they differ from climatological conditions. There are two different ways to study this co-variability solely using observational data: 1) the "top- down approach" wherein the atmospheric state is first identified and then the variability of the tracers is evaluated. This approach gives a general perspective of the distribution of tracers based on a particular weather state and 2) the "bottom-up approach" wherein the pollution episode is first identified and the weather state associated with it is studied. In this study we make use of the bottom-up approach as explained in the next section.

2. Data sets and methodology

The NO_2 tropospheric column densities from OMI (Ozone Monitoring Instrument) on board the

EOS Aura satellite are used in this study to define and identify extreme events (Boersma et al., 2001, 2008, 2011; Bucsela et al., 2006, 2008, 2013; Lamsal et al. 2008, 2010, 2014). 11 years (2004 – 2015) of daily Level 3 gridded standard product, available at 0.25x0.25 degrees resolution is analysed (OMNO2d, Version 3, available at: https://disc.gsfc.nasa.gov/Aura/data-holdings/OMI/omno2d_v003.shtml). This particular product is used as it provides good quality OMI retrievals, already screened based on recommendations by the OMI Algorithm Team. We allowed retrievals under partially cloudy conditions to be analysed, not only to have robust number of samples, but also to avoid clear-sky biases since the NO_2 transport is often associated with cyclonic systems that lead to increased cloudiness (Zien et al. 2014). We further tested the sensitivity of our results to using only cloud screened retrievals, to evaluate if the selection of extreme events and associated meteorological conditions are different from those cases when retrievals under partially cloudy conditions are used.

Humidity and cloud fraction retrievals from the AIRS (Atmospheric Infrared sounder) instrument on board Aqua satellite are used (Chahine et al. 2006; Susskind et al. 2014; Devasthale et al. 2016). Both Aqua and Aura satellites are a part of NASA’s A-Train convoy, providing added advantage of simultaneous observations of trace gases from OMI-Aura and thermodynamical information from AIRS-Aqua. AIRS Version 6 Standard Level 3 Daily Product (AIRX3STD) for the same period (2004-2015) is used (data available at: https://disc.gsfc.nasa.gov/uui/datasets?keywords=%22AIRS%22).

To investigate circulation patterns, u and v wind components at 850 hPa from ECMWF’s ERA-Interim Reanalysis are used (Dee et al., 2011; http://apps.ecmwf.int/datasets/data/interim-full-daily/levtype=sfc/).

In order to investigate co-variability of meteorological conditions and pollutants using observations, two different approaches can be taken (Fig. 1). In a “top-down” approach, a weather state classification can be done to identify most prevailing weather states that occur over the study area and then the relative distribution of pollutants can be investigated under those states to rank them. This approach was adapted by Thomas and Devasthale (2014) and Devasthale and Thomas (2012). In a “bottom-up” approach on the other hand, a set of pollution events can be identified first and then the corresponding meteorological conditions can be investigated. This bottom-up approach is the focus of the present study. It should be mentioned that both of these approaches have their advantages and limitations. For example, the dominant weather pattern identified in the top-down approach may not have the largest impact on pollutant variability and the pollution events identified in the bottom-up approach may not be associated with the dominant weather pattern or may not have the largest impact on an average in the weather state they occur. Therefore, only the combination of these two approaches will provide a complete picture of the co-variability between meteorological conditions and pollutants.

In the present study, an “extreme” pollution event is defined as follows. First, the histograms of NO_2 tropospheric column densities using OMI data for each month are computed over the centre of the study area (55N-60N, 11E-20E). This area is chosen because it accommodates top ten polluted and populated cities/regions in Sweden (Sjöberg et al. 2004; Klingberg et al. 2009; Bellandar et al. 2012; Gustafsson et al. 2014; Nilsson Sommar et al. 2014; Oudin et al. 2016; Taj et al. 2016). All events that surpass the 90-percentile (90%ile) value are considered as extreme events. The monthly histograms of NO_2 over the study region are shown in Fig. 2 along with 90 percentile thresholds for each month (vertical lines). Since NO_2 distributions over the study area show strong monthly variability, the monthly thresholds were chosen to define extreme events. The distributions of NO_2 have longer tails during winter half year and the concentrations are also higher. Therefore, the resulting 90%ile thresholds are also higher in winter compared to summer months. However, using thresholds based on percentiles (rather than having a fixed value throughout the season or year), makes the criteria for the selection of extreme events fair and equally applicable for each month.

3. Meteorological conditions observed during extreme events

The spatial distribution of tropospheric NO_2 column during climatological conditions, extreme events and anomalies thereof is presented in Fig. 3. Note that although the thresholds for defining extreme events are different for each month, the results are compiled over four distinct seasons for the sake of brevity. By definition, NO_2 anomalies during extreme events are similar in magnitude to climatological values over Scandinavia. The spatial extent of the severity of the extreme pollutant episodes over southern Sweden is noticeable. Under climatological conditions, highest concentrations are observed over northern Germany and France, the Netherlands and Belgium (the Benelux region). There is a good spatial coherence between NO_2 distributions under climatological conditions and extreme events, in the sense that the high concentrations of NO_2 seemed to have spread over southern Scandinavia during extreme events from the regions where climatological values are usually higher. It is to be noted that during extreme events the pollution levels over northern European regions are also enhanced. For an event to qualify as an extreme event over southern Scandinavia, the pollutant levels in the source regions also need to be higher than usual in order to allow strong transport under favourable atmospheric circulation patterns. This provides confidence in the selection process of extreme events. The NO_2 concentrations are relatively higher in winter and autumn compared to the summer months. This is mainly because atmospheric removal by radical species and deposition are much more efficient in the summer months.

In order to characterize typical meteorological conditions that can result in such high concentrations over Scandinavia, we first investigated the dominant wind direction at 850 hPa associated with those extreme events using ERA-Interim reanalysis data. The normalized frequency of occurrence of different wind directions during four seasons is shown in Fig. 4. It can be seen that, irrespective of the season, the south-westerly winds are dominant during extreme events accounting for 50-65% of total events. This is consistent with south-westerly extension of pollution plume mentioned earlier. The second largest annual occurrence is from south-easterly winds, accounting for 17% of total events followed equally similar contribution from north-westerly winds. Compared to climatological conditions, south-westerly winds have 30-40% more likelihood of being dominant during extreme events depending on the season. However, such clear tendency compared to climatological conditions is not observed in the case of other wind directions. The spatial pattern of the 850 hPa winds based on ERA-Interim reanalysis and corresponding humidity anomalies at 850 hPa based on AIRS data during extreme events are shown in Figs. 5 and 6 respectively. A clear transport pathway from the northern continental Europe to Scandinavia is visible. The strongest winds are observed during the DJF months followed by the SON months with average wind speeds reaching over 10 m/s. The weakest winds are observed during the JJA months. The circulation pattern is characterized by the presence of low pressure systems in the Norwegian Sea that create favourable conditions for the transport of pollutants from continental Europe into Scandinavia. The location of the center of these cyclonic systems can slightly vary over the Norwegian Sea, affecting the direction and strength of the northward flow, as evident in Fig. 5. For example, in the DJF months, the center is located far away in the open Norwegian Sea allowing stronger south-westerly winds over southern Scandinavia. In the JJA months, the center of cyclonic systems is close to western Norwegian coast. While this pattern also leads to south-westerly winds, air masses are mixed with colder and drier air from the northern Norwegian Sea.

The specific humidity anomalies show an influx of warmer and moister air masses over Scandinavia (Fig. 6), except in summer as mentioned above. The seasonality in the vertical structure of the specific humidity anomalies over Scandinavia is shown in Fig. 7c. While there are large deviations in humidity anomalies, influenced by the strength of the wind flow, they are positive regardless of the season during extreme events and peak at 2-3 km above the surface. Such increase in the free tropospheric moisture, especially during winter half year in the absence of local moisture sources, can only be explained by the transport from southern latitudes. The vertical water vapour anomalies are higher in winter half year (DJF and SON), consistent with high NO_2 anomalies during those months. Fig. 8 further shows cloud fraction anomalies. Average cloudiness is increased in all seasons during extreme events, in particular during winter half year. During this time of year, the large-scale frontal systems originating from the southwesterly regions can bring moister airmasses over Scandinavia, as can be seen in the circulation patterns and humidity anomalies, creating favourable conditions for cloud formation. Therefore, these positive cloud fraction anomalies, in combination with positive humidity anomalies and circulation patterns, are indicative of the long-range transport of airmasses associated with increased NO_2 concentrations.

For an extreme pollution event to be linked with the transport the wind flow should be stronger allowing rapid advection and associated circulation pattern also needs to be persistent. Fig. 7a and 7b show the histograms of wind speed at 850 hPa over the study areas during extreme events when data are partitioned by wind direction and by season respectively. The average values of wind speeds are also shown for extreme events and climatological conditions (in brackets). Although the distributions are shifted to higher wind speeds in nearly all cases during extreme events compared to climatological conditions, the average wind speeds are not significantly different. The south-westerly winds are strongest and show largest difference in average wind speeds, while the northeasterly winds are weakest. Average wind speeds during the winter half year (DJF and SON) are higher than the summer half year, consistent with observed positive anomalies of humidity and clouds.

The persistency of the different circulation patterns during these extreme events is further evaluated as shown in Fig. 7d. The persistency is defined as follows. If an extreme event is observed, the wind speed and wind direction are computed for the last 10 days. It is then checked how many days back in time that particular wind direction was *continuously* sustained. Two distinct modes in the persistency of circulation patterns are observed, one in which a particular wind direction persists for a day or two and a second mode in which winds persists for 3 to 5 continuous days. This is clearly different from the degree of persistency observed under climatological conditions when winds persisted in one particular direction predominantly for few days. It was identified that during extreme events south-easterly winds dominated the first mode explaining 78% of the total occurrence in that mode and the westerly winds dominated the second mode explaining 86% of the total occurrence. In the latter case, when the winds persist for few days (3-5 days), the conditions are favourable for the long-range transport from the southern latitudes since circulation patterns (Fig. 5) are associated with typical frontal systems and baroclinic disturbances that make their way over Scandinavia.

4. Sensitivity of chosen events to cloud clearing procedure

As mentioned in Section 2, we allowed retrievals under partially cloudy conditions to be analysed, not only to have a robust number of samples, but also to avoid potential clear-sky biases. However, clouds can contaminate the NO_2 retrievals by modulating scattering in the atmosphere. Moreover, clouds are highly variable not only in space and time but also in their nature, thus making it challenging to assess their overall impact on the quality of retrievals. In the case of our study, potential cloud contamination can affect the selection of extreme events and thereby associated weather patterns that are being studied. Therefore, we carried out a sensitivity study wherein the entire analysis was repeated using only cloud screened NO_2 retrievals to investigate to what extent cloud clearing would affect the chosen events and subsequent analysis. Fig. 9 shows the histograms of NO_2 total columns under partially cloudy (solid lines) and cloud screened conditions (dotted lines). The histograms are accumulated over four seasons instead of months for clarity (to avoid too many lines). The chosen 90%ile thresholds are certainly different under partially cloudy and cloud screened conditions, but only slightly. We also found that, depending on the month, the selected extreme events match under partially cloudy and cloud screened conditions between 76% and 88% of the time. Fig. 10 further shows the spatial climatological distribution of NO_2 and during extreme events using only cloud screened retrievals. When compared to Fig. 3, the spatial distributions look patchy as a result of selected screening, but the magnitude and spatial features do not change significantly, providing confidence in our earlier analysis based on partially cloudy retrievals. Finally we evaluated if the events based on cloud screened data impact the analysis of meteorological conditions investigated here. Fig. 11 shows the vertical structure of specific humidity anomalies over the study region under partially cloudy (solid lines) and cloud screened conditions (dotted lines). While the slight differences in the vertical structure do exist, their sign and magnitudes are not large enough to change any previous argumentation.

5. Conclusions

The main aim of the present study was to characterize typical meteorological conditions associated with extreme NO_2 pollution events over Scandinavia. To that end, the study employs the bottom-up approach, in contrast to top-down approach taken by Thomas and Devasthale (2014) to study statistical co-variability of weather states and pollutant distribution. Such detailed analysis characterizing circulation patterns and meteorological conditions involving more than 300 extreme pollution events identified using satellite data has not been done before over the Scandinavian region. It is observed that the south-westerly winds dominated during extreme events accounting for 50-65% of total events, while the second largest annual occurrence was from south-easterly winds, accounting for 17% of total events followed by an equally similar contribution from north-westerly winds. Wind speeds are generally higher during extreme events, but only slightly, making it challenging to delineate distinct circulation regimes under these events. For the first time, we investigated the degree of persistency of wind direction during extreme events. In contrast to climatological conditions, two distinct modes of persistency were found; first one lasting a day or so and dominated by winds from south-easterly direction and the other mode lasting 3 to 5 days dominated by south-westerly and north-westerly winds. This information on the degree of persistency in conjunction with circulation patterns could be useful to identify extreme transport events. Further analysis of circulation patterns in combination with spatial distribution of humidity and its vertical structure suggest that these events occur as a result of long-range transport from southern latitudes, most likely from the northern parts of Germany and France, the Netherlands and Belgium. The analysis presented here provides information that can be used in the process oriented evaluation of chemistry transport models over Scandinavia.

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References

Beirle, S., Boersma, K. F., Platt, U., Lawrence, M. G., and Wagner, T.: Megacity Emissions and Lifetimes of Nitrogen Oxides Probed from Space, Science, 333, 1737–1739, doi:10.1126/science.1207824, 2011.

Bellander T, Wichmann J, and Lind T., Individual Exposure to NO2 in Relation to Spatial and

Temporal Exposure Indices in Stockholm, Sweden: The INDEX Study. PLoS ONE 7(6): e39536. doi:10.1371/journal.pone.0039536, 2009.

Boersma, K. F., E. J. Bucsela, E. J. Brinksma, J. F. Gleason, NO2, OMI-EOS Algorithm Theoretical Basis Document: Trace Gas Algorithms: NO2, 4, 12-35, 2001. http://eospso.gsfc.nasa.gov/eos_homepage/for_scientists/atbd/docs/OMI/ATBD-OMI-04.pdf

Boersma, K. F., Jacob, D. J., Bucsela, E. J., Perring, A. E., Dirksen, R., van der A, R. J., Yantosca, R. M., Park, R. J., Wenig, M. O., and Bertram, T. H.: Validation of OMI tropospheric NO2 observations during INTEX-B and application to constrain NOx emissions over the eastern United States and Mexico, Atmos. Environ., 42, 4480–4497,doi:10.1016/j.atmosenv.2008.02.004, 2008.

Boersma, K. F., Eskes, H. J., Dirksen, R. J., van der A, R. J., Veefkind, J. P., Stammes, P., Huijnen, V., Kleipool, Q. L., Sneep, M., Claas, J., Leitão, J., Richter, A., Zhou, Y., and Brunner, D.: An improved tropospheric NO2 column retrieval algorithm for the Ozone Monitoring Instrument, Atmos. Meas. Tech., 4, 1905–1928, doi: 10.5194/amt-4-1905-2011, 2011.

Bucsela, E. J., Celarier, E. A., Wenig, M. O., Gleason, J. F., Veefkind, J. P., Boersma, K. F., and Brinksma, E. J.: Algorithm for NO2 vertical column retrieval from the ozone monitoring instrument, IEEE T. Geosci. Remote, 44, 1245–1258, doi:10.1109/TGRS.2005.863715, 2006.

Bucsela, E. J., Perring, A. E., Cohen, R. C., Boersma, K. F., Celarier, E. A., Gleason, J. F., Wenig, M. O., Bertram, T. H., Wooldridge, P. J., Dirksen, R., and Veefkind, J. P.: Comparison of tropospheric NO2 from in situ aircraft measurements with near-real-time and standard product data from OMI, J. Geophys. Res.,113, D16S31, doi:10.1029/2007JD008838, 2008.

Bucsela, E. J., Krotkov, N. A., Celarier, E. A., Lamsal, L. N., Swartz, W. H., Bhartia, P. K., Boersma, K. F., Veefkind, J. P., Gleason, J. F., and Pickering, K. E.: A new stratospheric and tropospheric NO_2 retrieval algorithm for nadir-viewing satellite instruments: applications to OMI, Atmos. Meas. Tech., 6, 2607-2626, doi:10.5194/amt-6-2607-2013, 2013.

Chahine, M. T and co-authors, AIRS: Improving Weather Forecasting and Providing New Data on Greenhouse Gases, Bull. Am. Meteorol. Soc., 87, 911–926, 2006.

Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N. and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system. Q.J.R. Meteorol. Soc., 137: 553–597, doi:10.1002/qj.828, 2011.

Devasthale, A. and M. A. Thomas, An investigation of statistical link between inversion strength and carbon monoxide over Scandinavia in winter using AIRS data, Atmospheric Environment, Vol. 56, 109-114 s, DOI: 10.1016/j.atmosenv.2012.03.042, 2012.

Devasthale et al. A decade of space borne observations of the Arctic atmosphere: novel insights from NASA's Atmospheric Infrared Sounder (AIRS) instrument. Bull. Amer. Meteor. Soc. doi:10.1175/BAMS-D-14-00202.1, in press, 2016.

Donnelly, A. A., Broderick, B. M. and Misstear, B. D.: The effect of long-range air mass transport pathways on PM10 and NO2 concentrations at urban and rural background sites in Ireland: Quantification using clustering techniques, J Environ Sci Health A Tox Hazard Subst Environ Eng., doi: 10.1080/10934529.2015.1011955, 2015.

Eckhardt, S., A. Stohl, S. Beierle, N. Spichtinger, P. James, C. Forster, C. Junker, T. Wagner, U. Platt, and S. G. Jennings, 2003: The North Atlantic Oscillation controls air pollution transport to the Arctic, Atmos. Chem. Phys., 3, 1769-1778, 2003.

Ehhalt, D. H., Rohrer, F., and Wahner, A.: Sources and Distribution of NO x in the Upper Troposphere at Northern Mid-Latitudes, J. Geophys. Res., 97, 3725–3738, http://www.agu.org/journals/jd/v097/iD04/91JD03081/, 1992.

Geddes JA, et al. Long-term trends worldwide in ambient NO_2 concentrations inferred from satellite observations. Environ Health Perspect 1243281–289.2892016, doi:10.1289/ehp.1409567, 2016.

Gustafsson, M., H. Orru, B. Forsberg, S. Åström, H. Tekie, K. Sjöberg, Quantification of population exposure to NO2, PM2.5 and PM10 in Sweden 2010, Swedish Environmental Research Institute (IVL), IVL Report B2197, pp. 74, December 2014.

Klingberg, J., M. P. Björkman, G. P. Karlsson, and H. Pleijel, Observations of Ground-level Ozone and NO2 in Northernmost Sweden, Including the Scandian Mountain Range, AMBIO, 38(8):448-451. doi: http://dx.doi.org/10.1579/0044-7447-38.8.448, 2009.

Lamarque, J.-F. and Dentener, F. and McConnell, J. and Ro, C.-U. and Shaw, M. and Vet, R. and Bergmann, D. and Cameron-Smith, P. and Dalsoren, S. and Doherty, R. and Faluvegi, G. and Ghan, S. J. and Josse, B. and Lee, Y. H. and MacKenzie, I. A. and Plummer, D. and Shindell, D. T. and Skeie, R. B. and Stevenson, D. S. and Strode, S. and Zeng, G. and Curran, M. and Dahl-Jensen, D. and Das, S. and Fritzsche, D. and Nolan, M., Multi-model mean nitrogen and sulfur deposition from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): evaluation of historical and projected future changes, Atmos. Chem. Phys., 13, 7997-8018,2013.

Lamsal LN, Martin RV, van Donkelaar A, Celarier EA, Bucsela EJ, Boersma KF, et al. Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: insight into the seasonal variation of nitrogen oxides at northern midlatitudes. J Geophys Res 115D05302; doi:10.1029/2009JD013351 , 2010.

Lamsal LN, Martin RV, van Donkelaar A, Steinbacher M, Celarier EA, Bucsela E, et al. Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument. J Geophys Res 113D16308; .10.1029/2007JD009235, 2008.

Lamsal, L. N., Krotkov, N. A., Celarier, E. A., Swartz, W. H., Pickering, K. E., Bucsela, E. J., Gleason, J. F., Martin, R. V., Philip, S., Irie, H., Cede, A., Herman, J., Weinheimer, A., Szykman, J. J., and Knepp, T. N.: Evaluation of OMI operational standard NO_2 column retrievals using in situ and surface-based NO_2 observations, Atmos. Chem. Phys., 14, 11587-11609, doi:10.5194/acp-14-11587-2014, 2014.

Nilsson Sommar, J., A. Ek, R. Middelveld, A. Bjerg, S-E. Dahlén, C. Janson, B. Forsberg, Quality of life in relation to the traffic pollution indicators NO2 and NOx: results from the Swedish GA2LEN survey, BMJ Open Resp Res 2014;1:1 e000039 doi:10.1136/bmjresp-2014-000039.

Oudin, A., L. Bråbäck, D. Oudin Åström, M. Strömgren, and B. Forsberg, Association between neighbourhood air pollution concentrations and dispensed medication for psychiatric disorders in a large longitudinal cohort of Swedish children and adolescents, BMJ Open 2016;6:6 e010004 doi:10.1136/bmjopen-2015-010004, 2016.

Richter A, Burrows JP, Nüss H, Granier C, Niemeier U. Increase in tropospheric nitrogen dioxide over China observed from space. Nature 437129–132.132; doi:10.1038/nature04092, 2005.

Schneider, P., Lahoz, W. A., and van der A, R.: Recent satellite-based trends of tropospheric nitrogen dioxide over large urban agglomerations worldwide, Atmos. Chem. Phys., 15, 1205-1220, doi:10.5194/acp-15-1205-2015, 2015.

Sjöberg K., M. Haeger-Eugensson, M. Lijeberg, H. Blomgren, and B. Forsberg, Quantification of population exposure to nitrogen dioxide in Sweden, Swedish Environmental Research Institute (IVL), IVL Report B1579, pp. 31, September 2004.

Stohl, A., Huntrieser, H., Richter, A., Beirle, S., Cooper, O. R., Eckhardt, S., Forster, C., James, P., Spichtinger, N., and Wenig, M.: Rapid intercontinental air pollution transport associated with a meteorological bomb, Atmos. Chem. Phys., 3, 969–985, 2003, http://www.atmos-chem-phys.net/3/969/2003/.

Schaub, D., Weiss, A. K., Kaiser, J. W., Petritoli, A., Richter, A., Buchmann, B., and Burrows, J. P.: A transboundary transport episode of nitrogen dioxide as observed from GOME and its impact in the Alpine region, Atmos. Chem. Phys., 5, 23–37, doi:10.5194/acp-5-23-2005, 2005.

Susskind, J., J. M. Blaisdell and L. Iredell, Improved methodology for surface and atmospheric soundings, error estimates and quality control procedures: the atmospheric infrared sounder science team version-6 retrieval algorithm, J. Appl. Remote Sens., 8(1), 084994, doi:10.1117/1.JRS.8.084994, 2014.

Taj T, Stroh E, Åström DO, Jakobsson K, Oudin A., Short-Term Fluctuations in Air Pollution and Asthma in Scania, Sweden. Is the Association Modified by Long-Term Concentrations? PLoS ONE 11(11): e0166614. doi:10.1371/journal.pone.0166614, 2016.

Thomas, M. A. and Devasthale, A.: Sensitivity of free tropospheric carbon monoxide to atmospheric weather states and their persistency: an observational assessment over the Nordic countries, Atmos. Chem. Phys., 14, 11545-11555, doi:10.5194/acp-14-11545-2014, 2014.

van der A, R. J., H. J. Eskes, K. F. Boersma, T. P. C. van Noije, M. Van Roozendael, I. De Smedt, D. H. M. U. Peters, and E. W. Meijer, Trends, seasonal variability and dominant NOx source derived from a ten year record of NO2 measured from space, J. Geophys. Res., 113, D04302, doi:10.1029/2007JD009021, 2008.

Vasilkov, A. P., Joiner, J., Oreopoulos, L., Gleason, J. F., Veefkind, P., Bucsela, E., Celarier, E. A., Spurr, R. J. D., and Platnick, S.: Impact of tropospheric nitrogen dioxide on the regional radiation budget, Atmos. Chem. Phys., 9, 6389-6400, doi:10.5194/acp-9-6389-2009, 2009.

Wenig, M., Spichtinger, N., Stohl, A., Held, G., Beirle, S., Wagner, T., Jähne, B., and Platt, U.: Intercontinental transport of nitrogen oxide pollution plumes, Atmos. Chem. Phys., 3, 387–393, doi:10.5194/acp-3-387-2003, 2003.

Zhang, Q., et al., NOx emission trends for China, 1995–2004: The view from the ground and the view from space, J. Geophys. Res., 112, D22306, doi:10.1029/2007JD008684, 2007.

Zien, A. W., Richter, A., Hilboll, A., Blechschmidt, A.-M., and Burrows, J. P.: Systematic analysis of tropospheric NO2 long-range transport events detected in GOME-2 satellite data, Atmos. Chem. Phys., 14, 7367–7396, doi: 10.5194/acp-14-7367-2014, 2014.

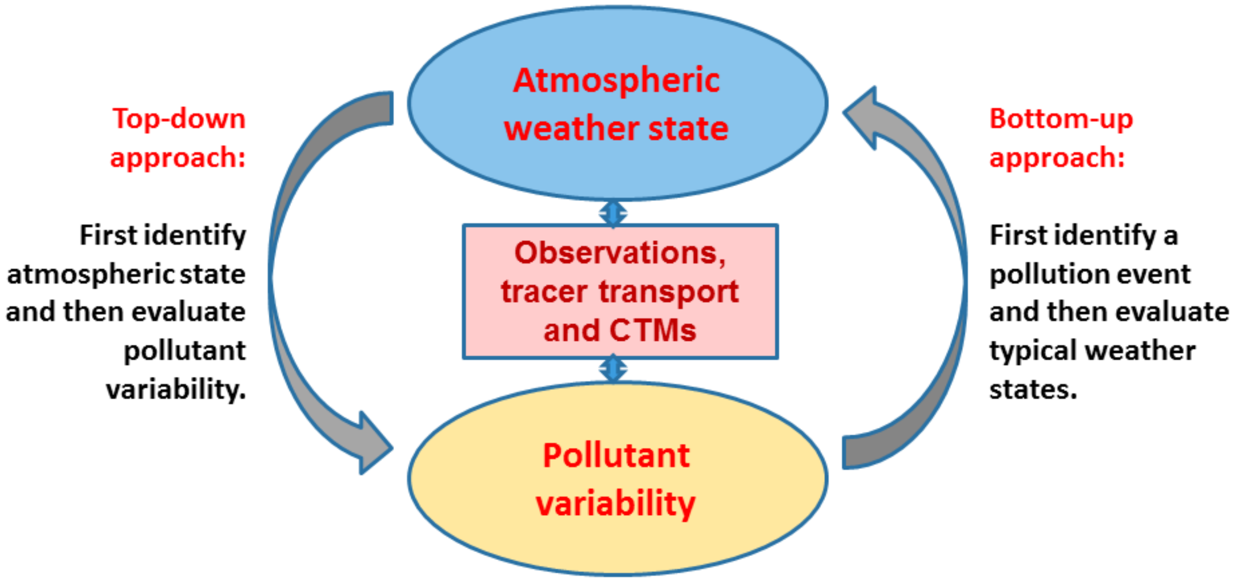


Fig. 1: Schematic showing two different approaches to study statistical co-variability of atmospheric weather states and pollutant concentrations.

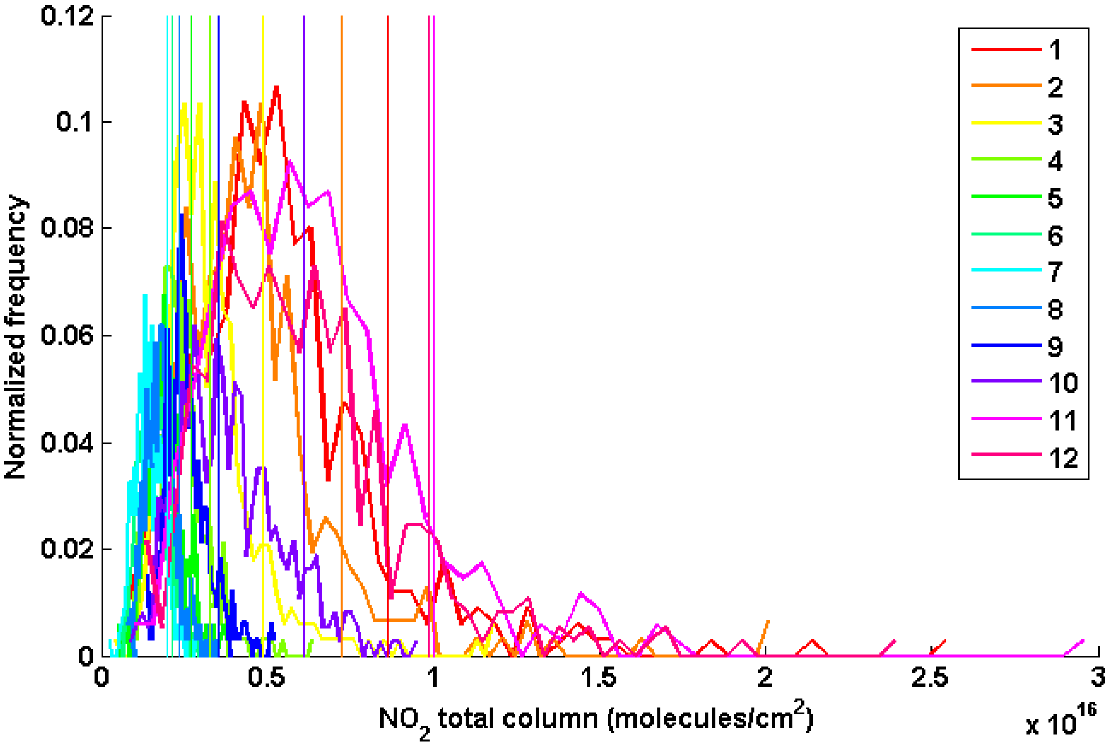
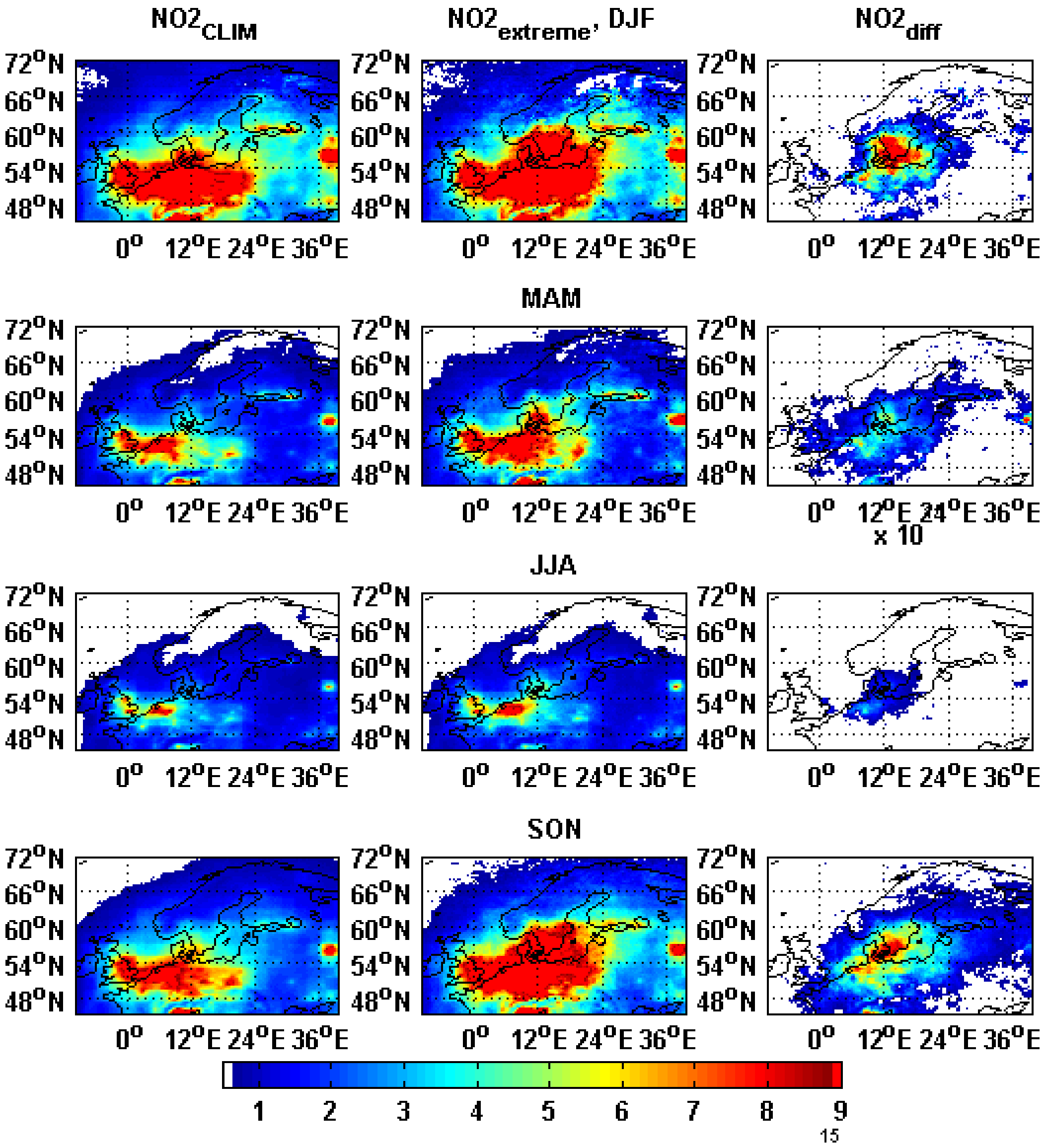


Fig. 2: Monthly histograms of tropospheric total column NO_2 over the centre of the study area (55N-60N, 11E-20E) and corresponding 90%ile thresholds (shown by vertical lines). The legend shows the month number from starting January to December.



Fig, 3: Seasonal, climatological average tropospheric NO_2 total column (first column) based on nearly 11-yr OMI data (2004-2015), NO_2 distribution during extreme events (second column) and the difference between the two (third column). The units are in molecules/cm^2.

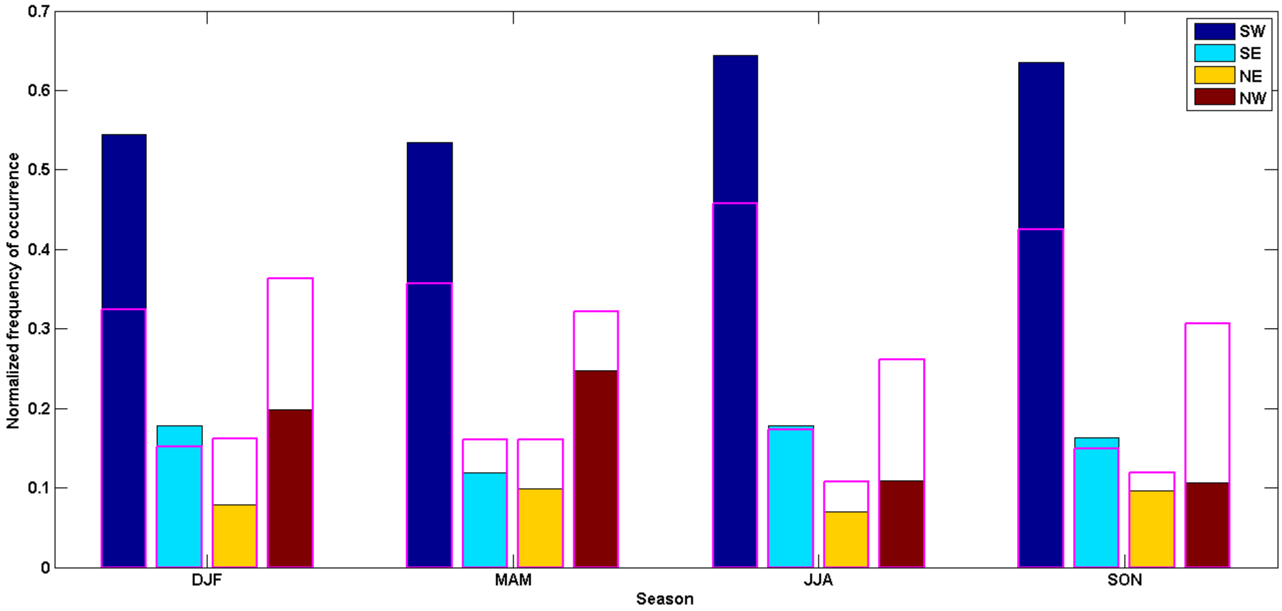


Fig. 4: Seasonal normalized frequency of occurrence of a particular wind direction at 850 hPa when NO_2 extreme pollution events were observed. The hollow magenta bars show normalized frequency under climatological conditions.

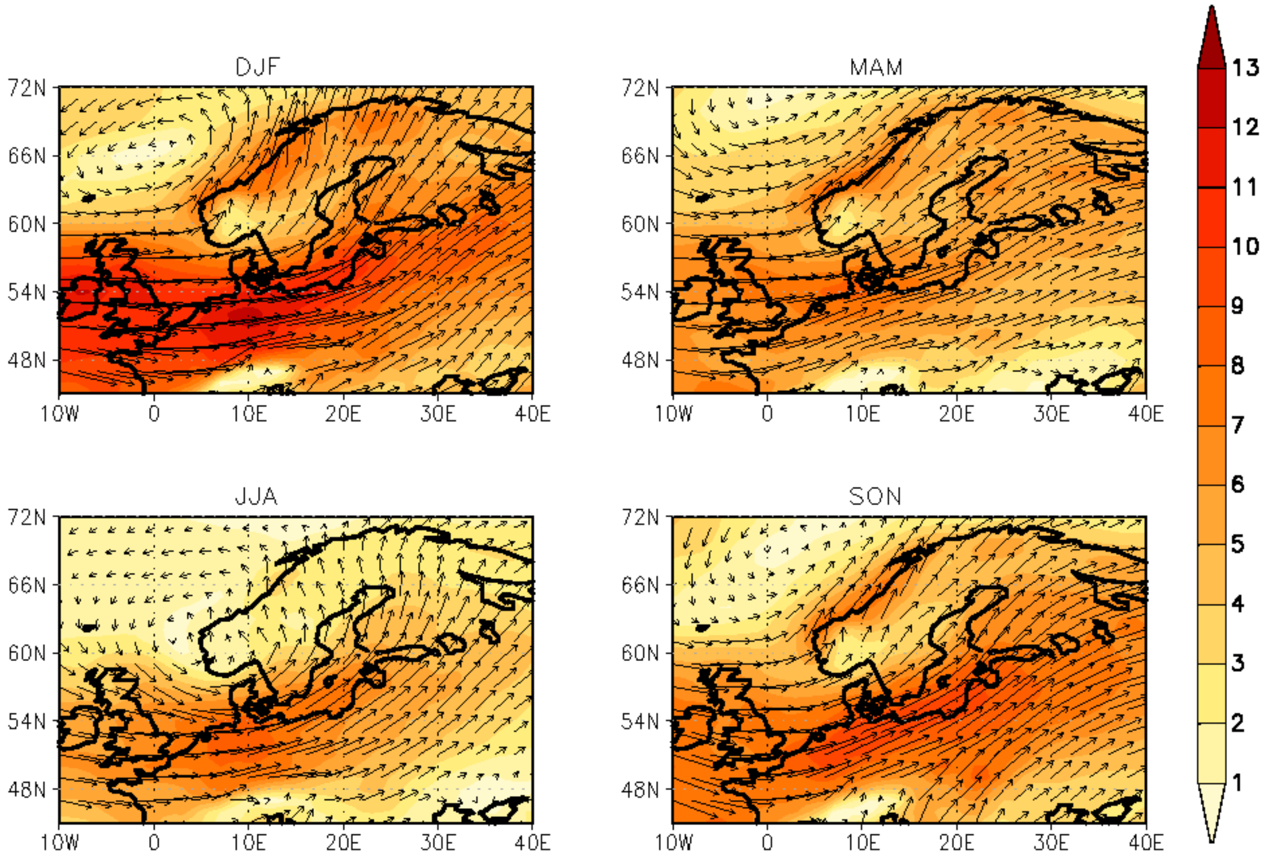


Fig. 5: Seasonal average wind strengths and direction at 850 hPa showing dominant circulation pattern observed when NO_2 extreme pollution events occur.

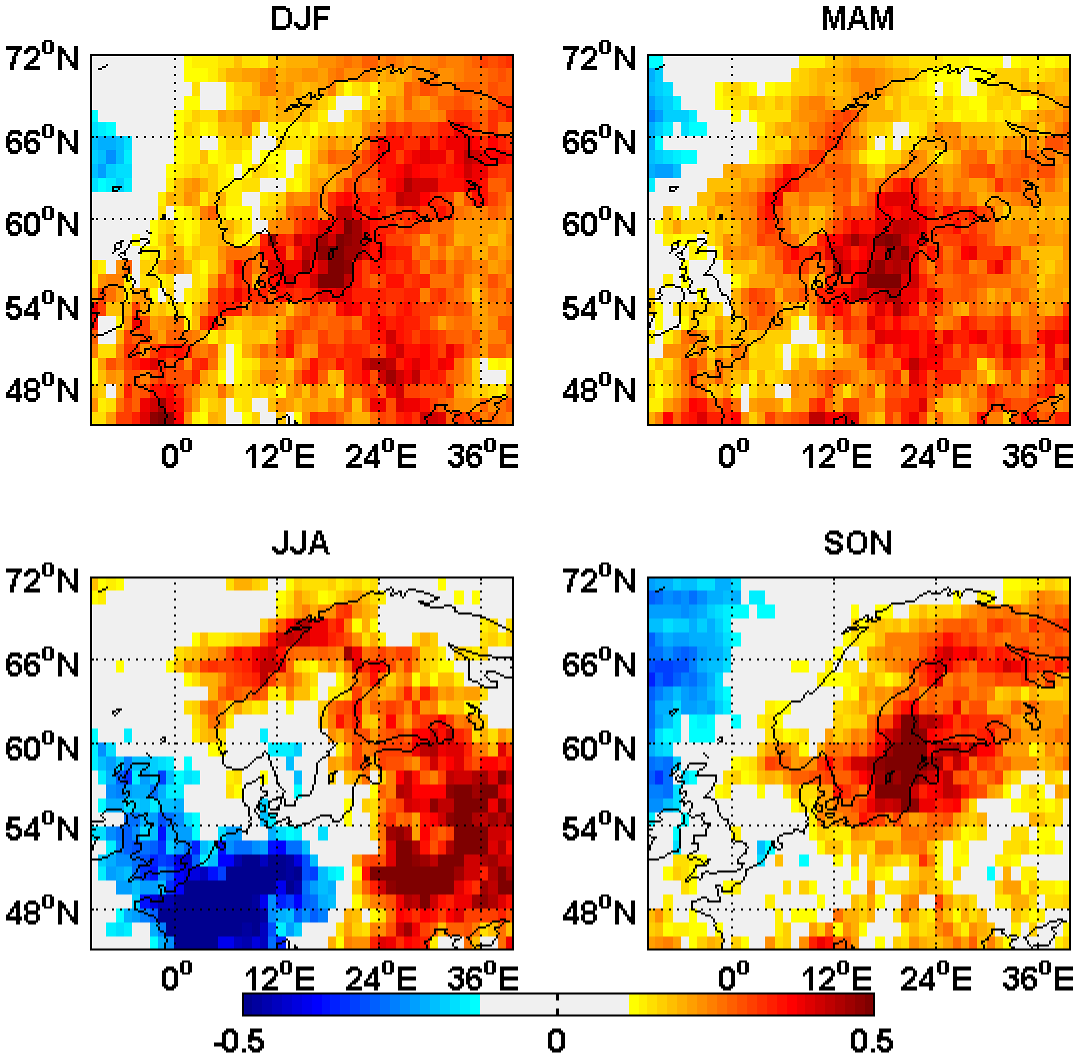


Fig. 6: The seasonal spatial patterns of specific humidity anomalies (g/kg) during extreme NO_2 pollution events.

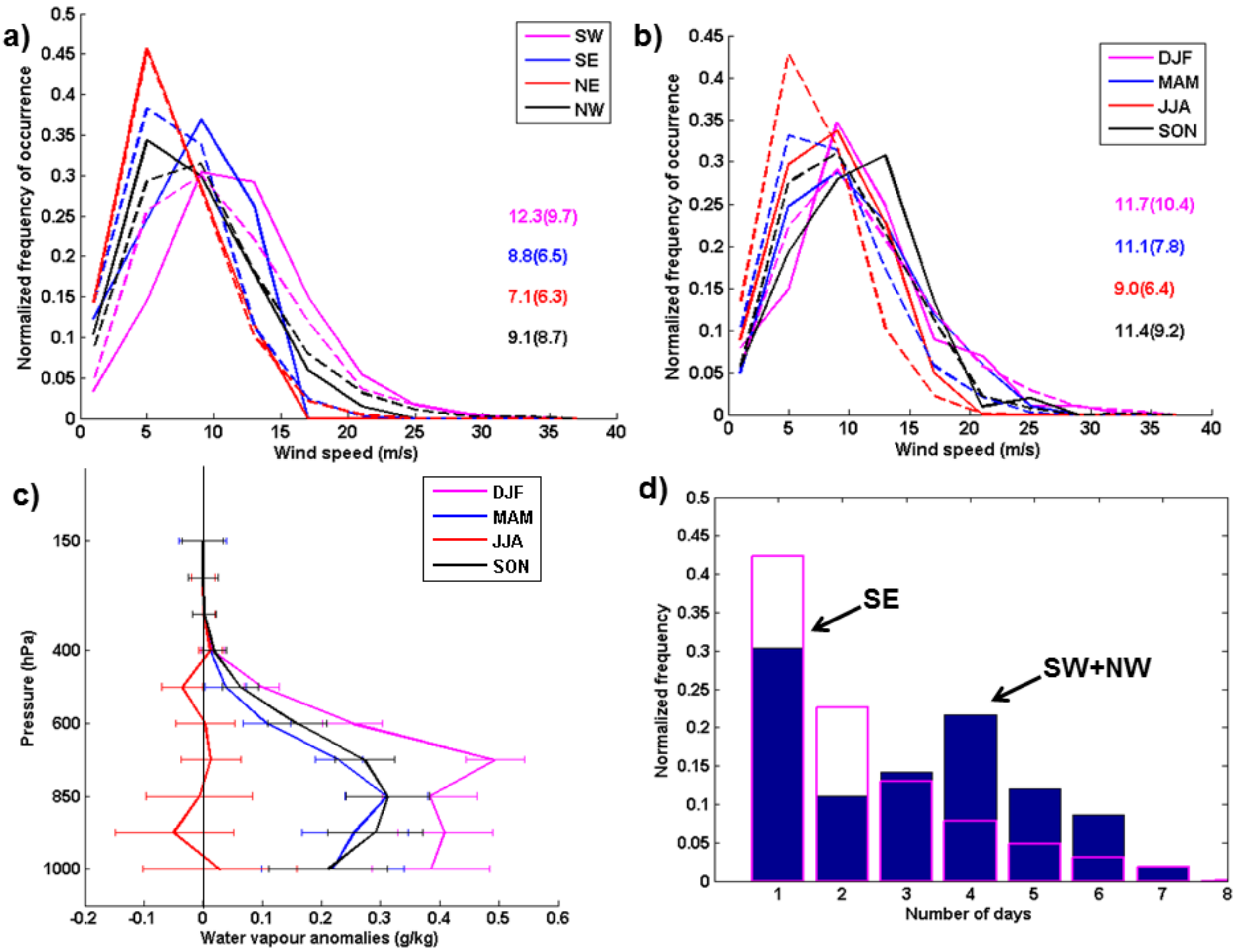


Fig. 7: a) Histograms of wind speeds (m/s) at 850 hPa over the center of the study area (55N-60N, 11E-20E) during extreme events (solid lines) and climatological conditions (dotted lines, 2004-2015) when data are partitioned for different wind directions. The numbers show average wind speeds (m/s) during extreme events and in brackets under climatological conditions. b) Same as in (a), but when wind data are partitioned for different seasons. c) Vertical anomalies of specific humidity (g/kg) during extreme events with horizontal bars showing standard deviations. d) Persistency of wind directions as a function of number of continuous days. The magenta bars show persistency under climatological conditions.

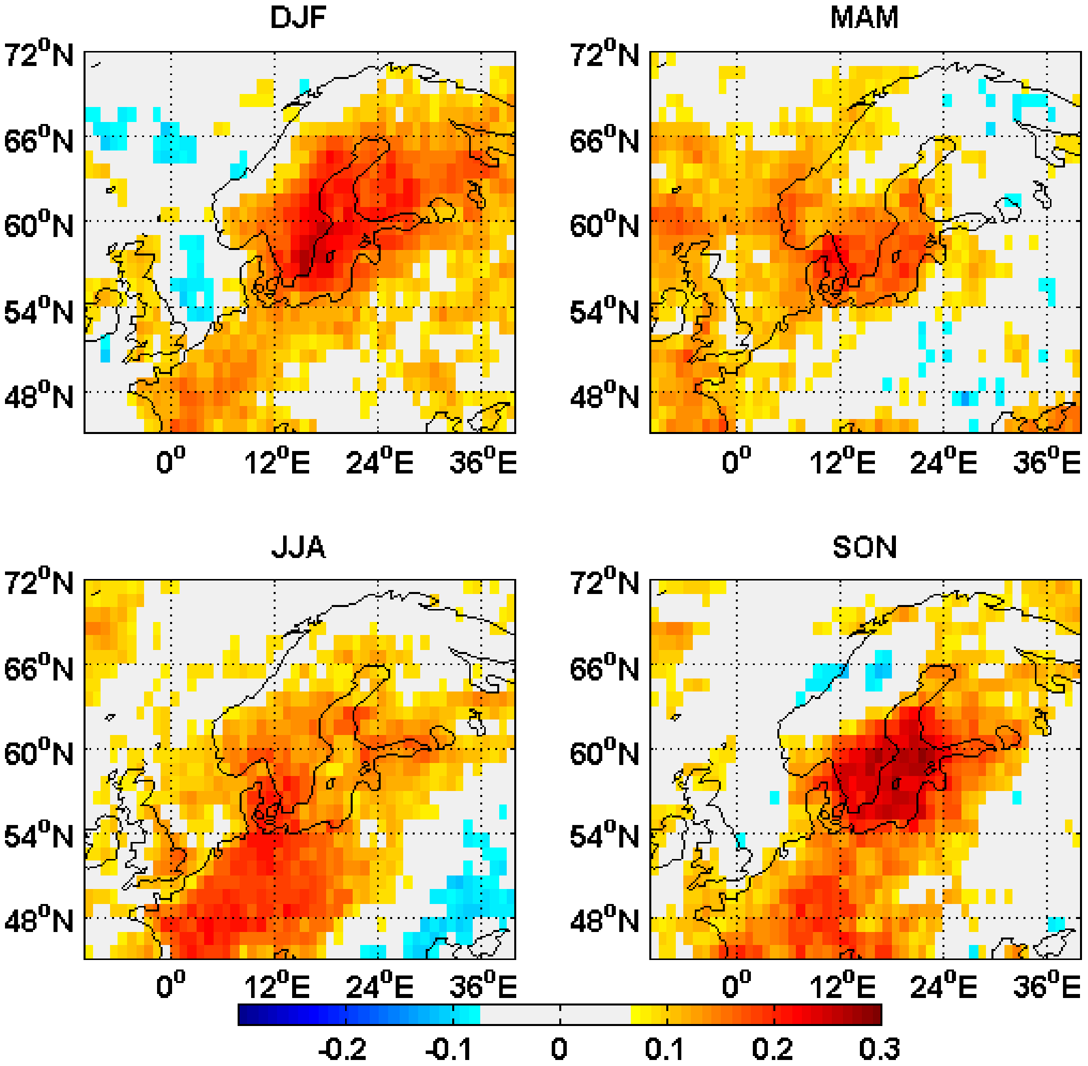


Fig. 8: Total cloud fraction anomalies observed during extreme events based on AIRS data.

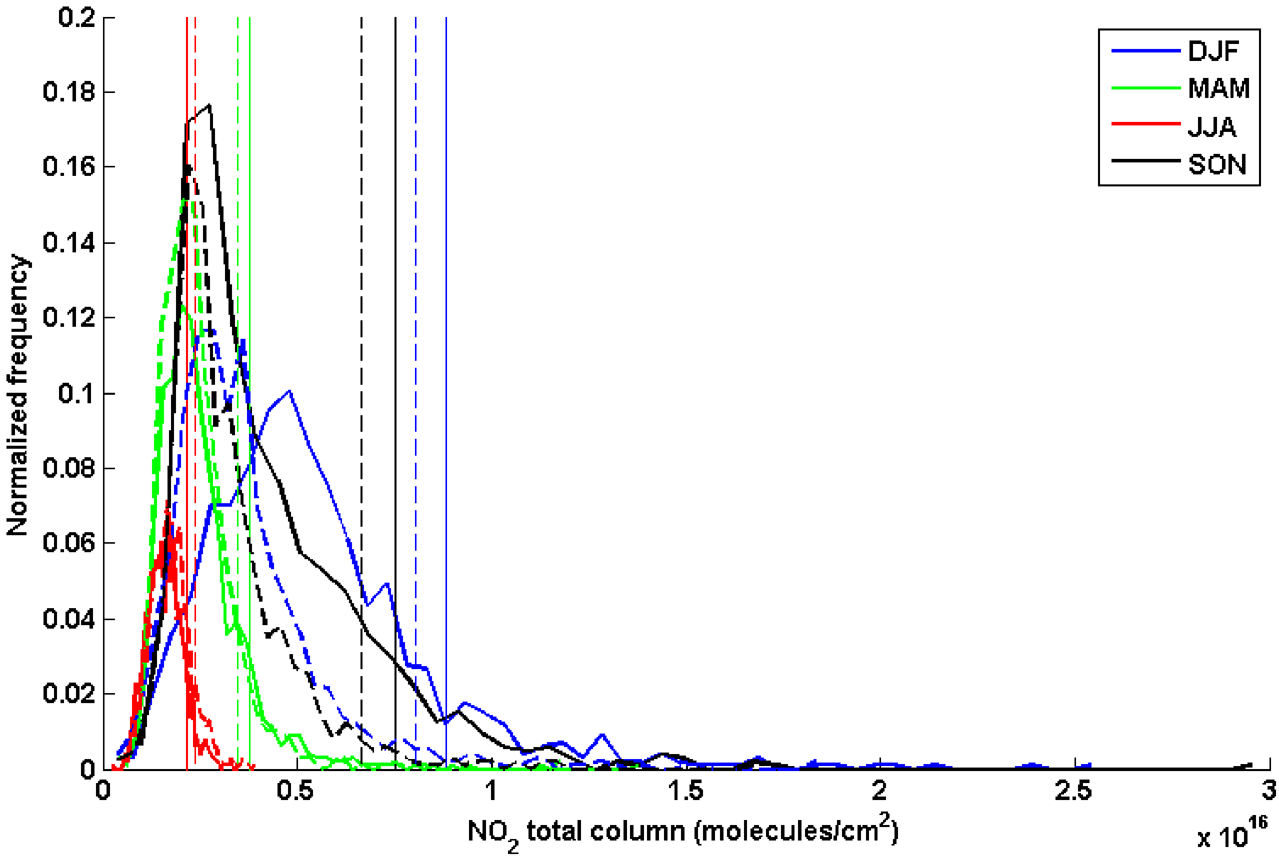


Fig. 9: Seasonal histograms of total column tropospheric NO_2 over the centre of the study area (55N-60N, 11E-20E) and corresponding 90%ile thresholds (shown by vertical lines). The solid lines show histograms based on retrievals under partially cloudy conditions, while the dotted lines show histograms based only on cloud cleared retrievals.

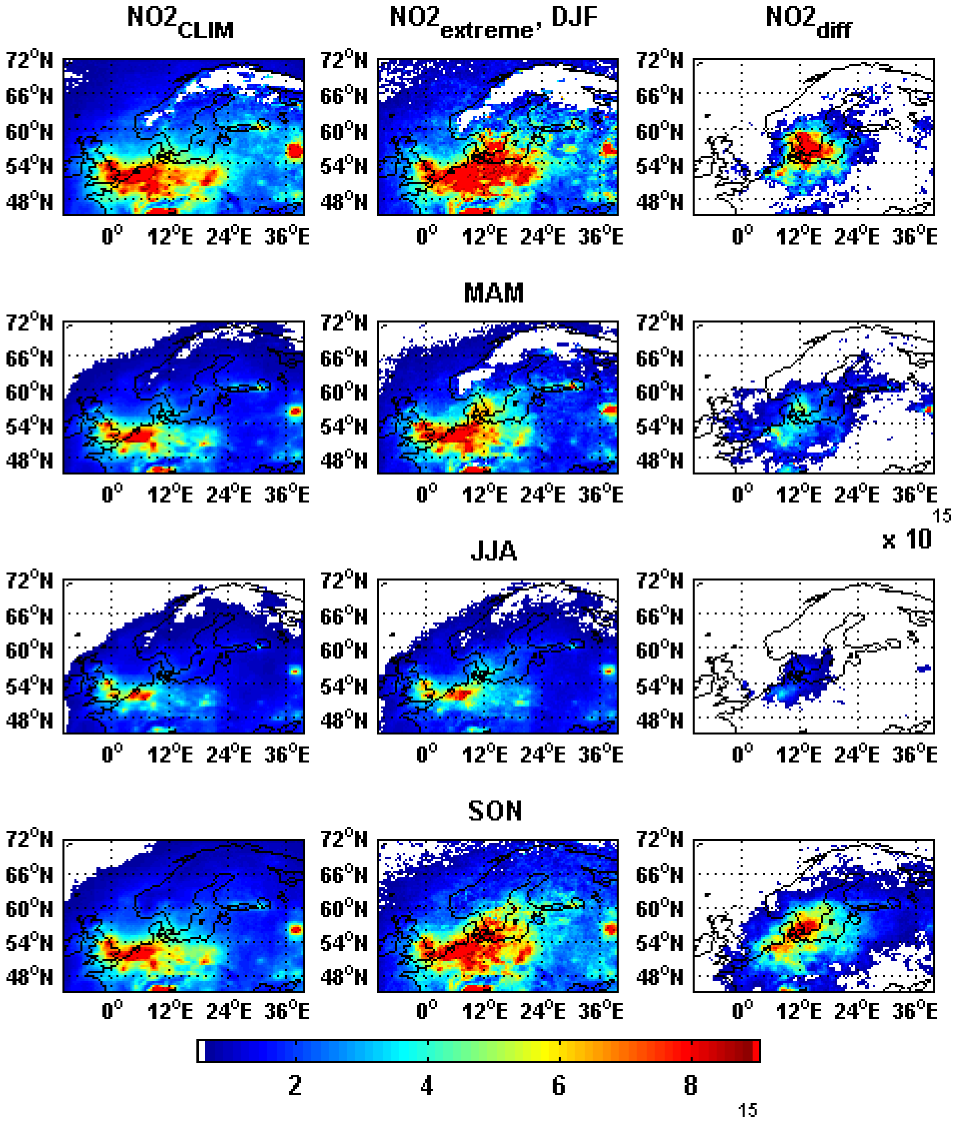


Fig. 10: Seasonal, climatological average tropospheric NO_2 total column (first column) based only on cloud screened OMI data (2004-2015), NO_2 distribution during extreme events (second column, also based on cloud screened data) and the difference between the two (third column). The units are in molecules/cm^2.

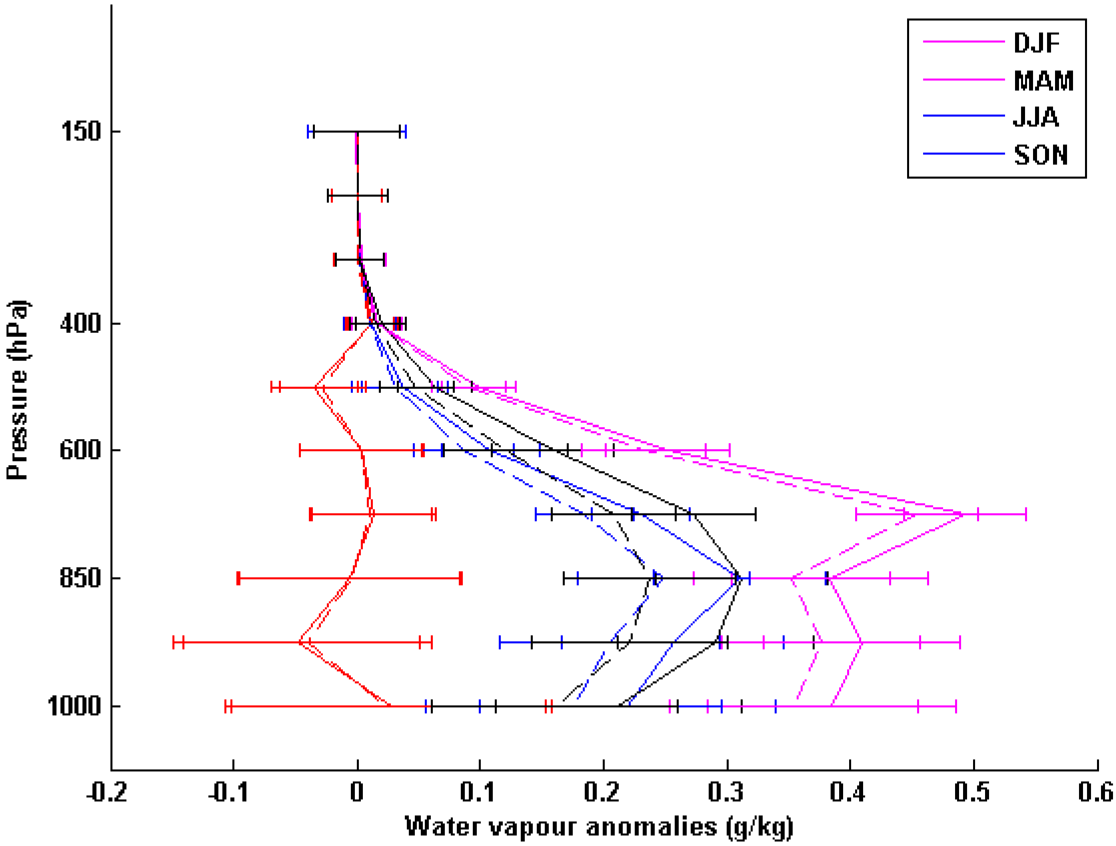


Fig. 11: Vertical anomalies of specific humidity (g/kg) during extreme events with horizontal bars showing standard deviations. The solid lines show anomalies under partially cloudy retrievals and dotted lines based on cloud screened retrievals.