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.

<u>General Comments:</u>

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.

- 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.
- 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 these asonality 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.