Supplement to "The influence of mid-latitude cyclones on European background surface ozone"

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S1 The long-term relationship between cyclone frequency and extreme/baseline surface O_3

To test the sensitivity of the results to the values chosen for high and low O_3, the analysis was repeated for the extreme high and low O_3 percentiles ($O_3 > 95^{th}$ pc and $O_3 < 5^{th}$ pc, respectively). A similar relationship between cyclone tracks and the extreme high and extreme low O_3 at Mace Head (Table S1) is found to the relationship between cyclone tracks and the high and low O_3 quartile percentiles (Table 2). The mean percentage of cyclone tracks associated with extreme high O_3 at Mace Head in the North (14 %) and Center (13 %) regions is greater than the mean percentage of cyclone tracks associated with extreme low O_3 through those two regions (8 and 11 %, respectively). In the South region, the mean percentage of cyclone tracks associated with extreme low O_3 at Mace Head (18 %) is greater than those associated with extreme high O_3 (14 %). The number of years where the percentage of cyclone tracks passing through a region are associated with extreme high O_3 is still greatest in the North region (14 years) and least in the South region (5 years; Table S1). Similar to the quartile percentile results, the West and East regions are evenly split, with slightly higher mean percentage of tracks associated with extreme low O_3 (17 %) than with extreme high O_3 (16 %) in the West region and a higher mean percentage of tracks associated with extreme high O_3 (13 %) than with extreme low O_3 (12 %) in the East region.

When considering the extreme high and low O_3 percentiles for Monte Vehlo, a different result emerges. In this case, all five regions show more tracks are associated with extreme low O_3 than with extreme high O_3 (Table S1), with 15 of the years in most regions identified by more tracks passing through a region being associated with extreme low O_3 than with extreme high O_3, although these differences are typically not significant (Table S1).

Mace Head					
Region	Mean percent of tracks per season associated with $O_3 > 95^{th}$ pc	Number of years with more tracks associated with $O_3 > 95^{th}$ pc (*SGF*)	Mean percent of tracks per season associated with $O_3 < 5^{th}$ pc	Number of years with more tracks associated with $O_3 < 5^{th}$ pc (*SGF*)	Mean number of tracks per season (MAM)
North	14 %	14 (*7*)	8 %	9 (*0*)	32
Center	13 %	12 (*3*)	11 %	11 (*1*)	30
South	14 %	5 (*1*)	18 %	18 (*3*)	25
West	16 %	9 (*1*)	17 %	14 (*3*)	50
East	13 %	10 (*1*)	12 %	13 (*1*)	52

Monte Velho					
Region	Mean percent of tracks per season associated with $O_3 > 95^{th}$ pc	Number of years with more tracks associated with $O_3 > 95^{th}$ pc (*SGF*)	Mean percent of tracks per season associated with $O_3 < 5^{th}$ pc	Number of years with more tracks associated with $O_3 < 5^{th}$ pc (*SGF*)	Mean number of tracks per season (MAM)
North	11 %	4 (*0*)	18 %	15 (*3*)	29
Center	14 %	4 (*0*)	19 %	15 (*1*)	24
South	22 %	4 (*0*)	26 %	15 (*0*)	17
West	22 %	4 (*1*)	26 %	15 (*0*)	39
East	18 %	5 (*0*)	22 %	14 (*2*)	45

Table S1. (top) The mean percent of cyclone tracks associated with extreme O_3 ($> 95^{th}$ pc) or baseline O_3 ($< 5^{th}$ pc) measured at the surface at Mace Head are given based on each MAM season in the period 1988–2010. The total number of years in the 23-year period (1988–2010) which have a greater number of tracks associated with $O_3 > 95^{th}$ pc and $O_3 < 5^{th}$ pc. **(bottom)** The total number of those years which have a significant χ^2 difference at $p < 0.05$ confidence interval are given in the brackets (*SGF*). See Fig. **??** for definition of the regions. The mean percent of cyclone tracks associated with extreme O_3 ($> 95^{th}$ pc) or baseline O_3 ($< 5^{th}$ pc) measured at the surface at Monte Velho are given based on each MAM season in the period 1989-2009 (excluding 1999 and 2005 which had insufficient data). The total number of years in the 19-year period (1989-2009, excluding 1999 and 2005) which have a greater number of tracks associated with $O_3 > 95^{th}$ pc and $O_3 < 5^{th}$ pc. The total number of those years which have a significant χ^2 difference at $p < 0.05$ confidence interval are given in the brackets (*SGF*).

S2 Additional cyclone figures

S2.1 N1 cyclone using MERRA-2 reanalysis and STFR

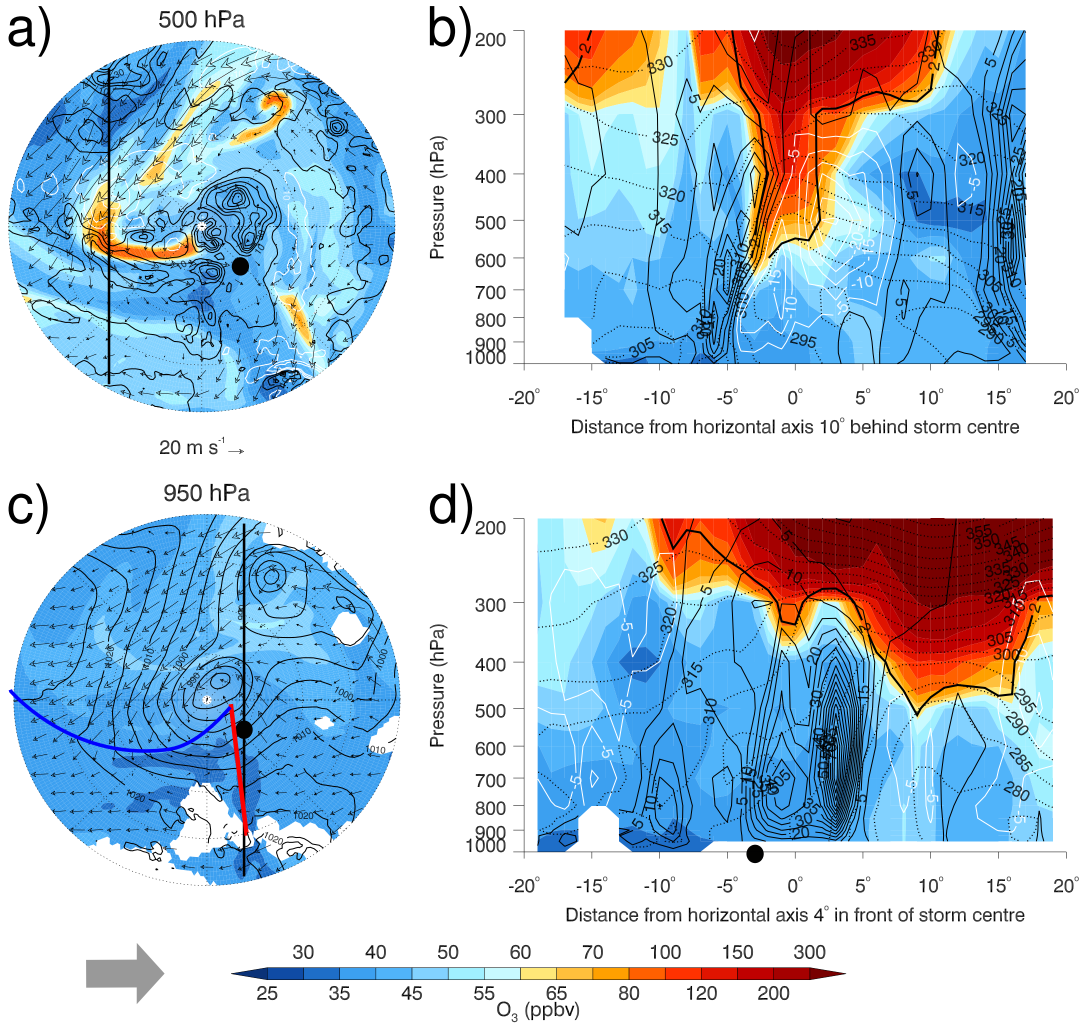

Figure S1. N1 cyclone on 4 March 2007 00UTC; similar to Fig. 6 except using MERRA-2 reanalysis data. Note, panel **(c)** is 950 hPa instead of 1000 hPa to minimize the missing data plotted. This occurs since the GEOS-5 model masks orography.

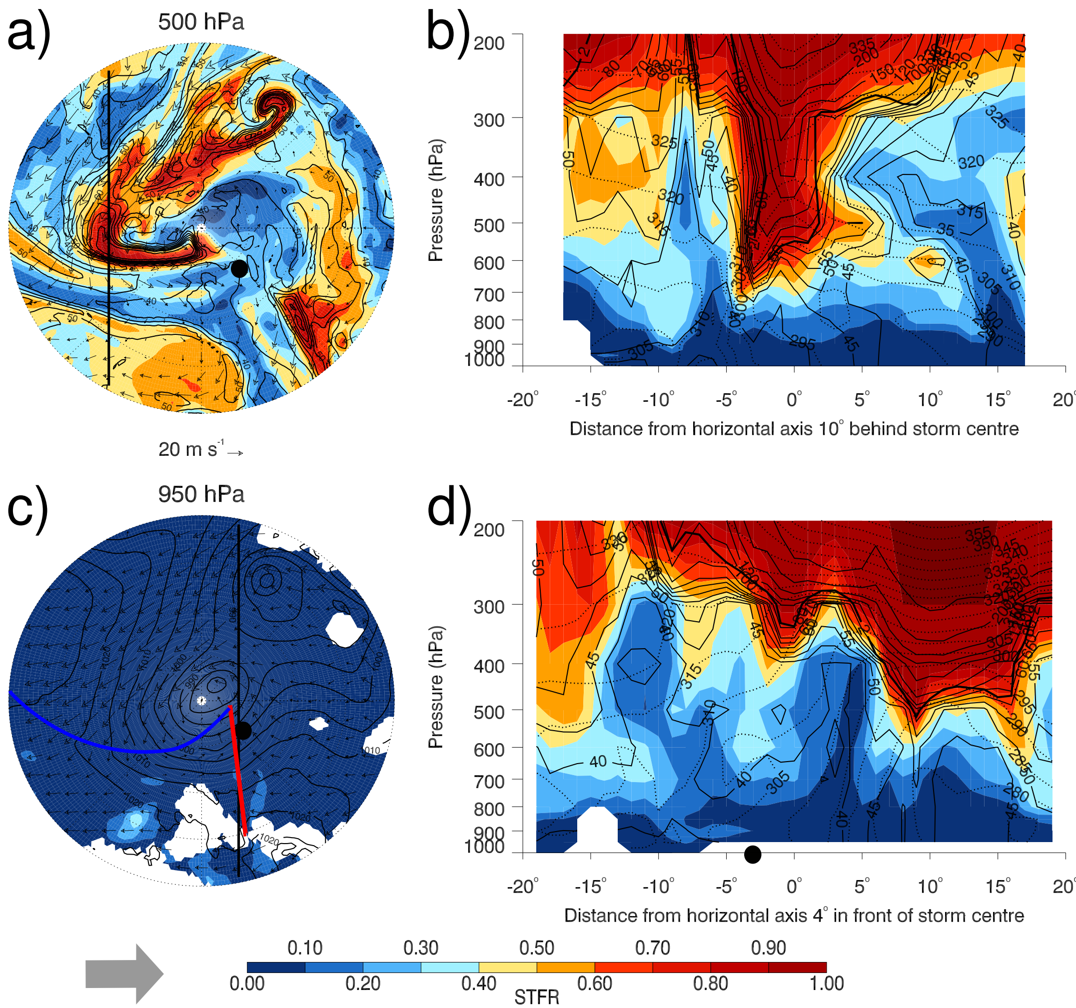

Figure S2. N1 cyclone on 4 March 2007 00UTC; similar to Fig. 6 except cyclone-centered STFR (color) and MERRA-2 O_3 (**a**,**b**,**d**, thin black contours; ppbv, increments matching Fig. 3 color bar) and SLP (**c**, black contours; 5 hPa increments). Figure also shows MERRA-2 θ_e (**b**,**d**), PV (**b**,**d**) and system relative winds (**a**,**c**). Note, panel **(c)** is 950 hPa instead of 1000 hPa to minimize the missing data plotted. This occurs since the GEOS-5 model masks orography.

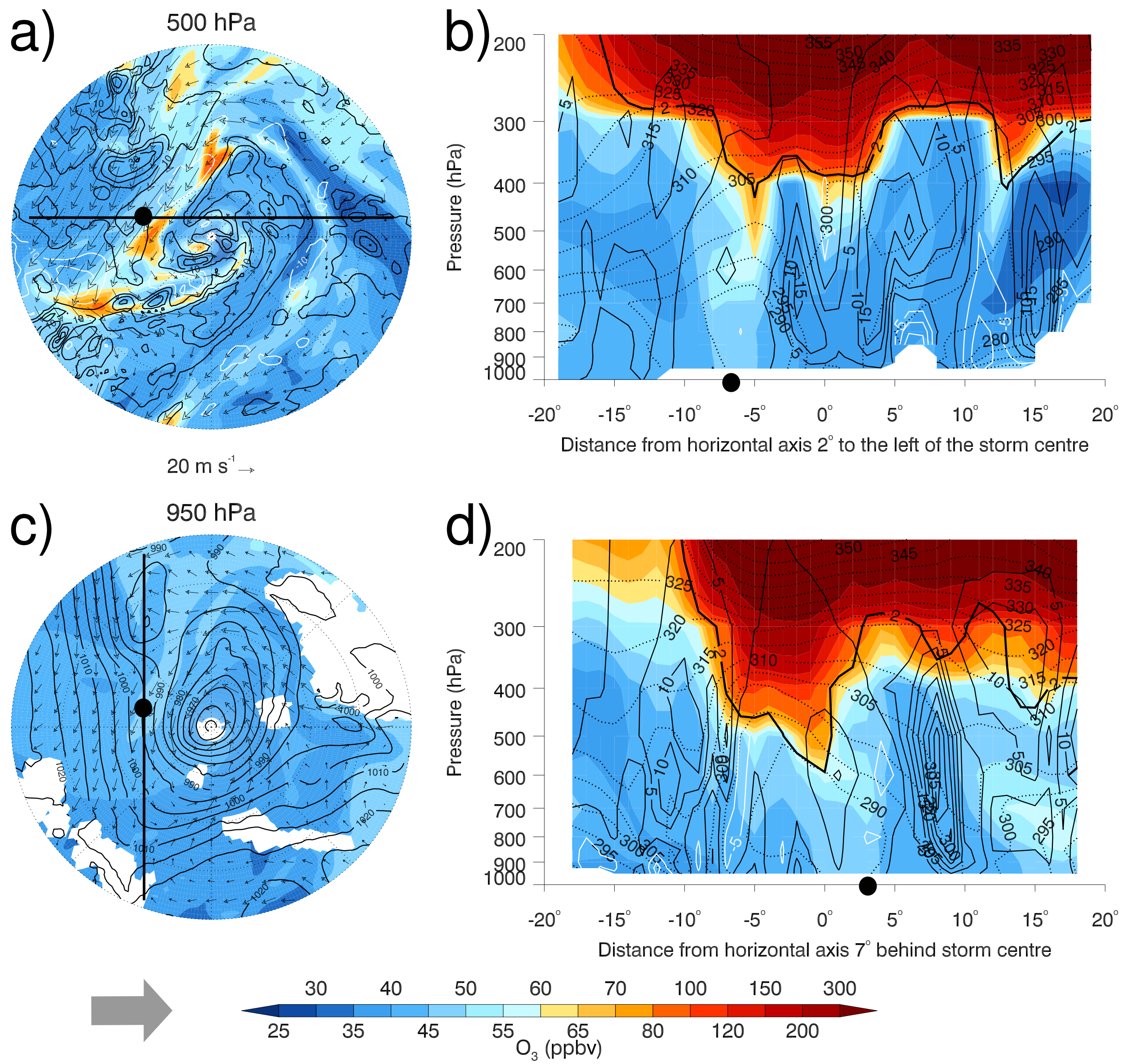

Figure S3. N1 cyclone on 5 March 2007 00UTC; similar to Fig. 7 except using MERRA-2 reanalysis data. Note, panel **(c)** is 950 hPa instead of 1000 hPa to minimize the missing data plotted. This occurs since the GEOS-5 model masks orography.

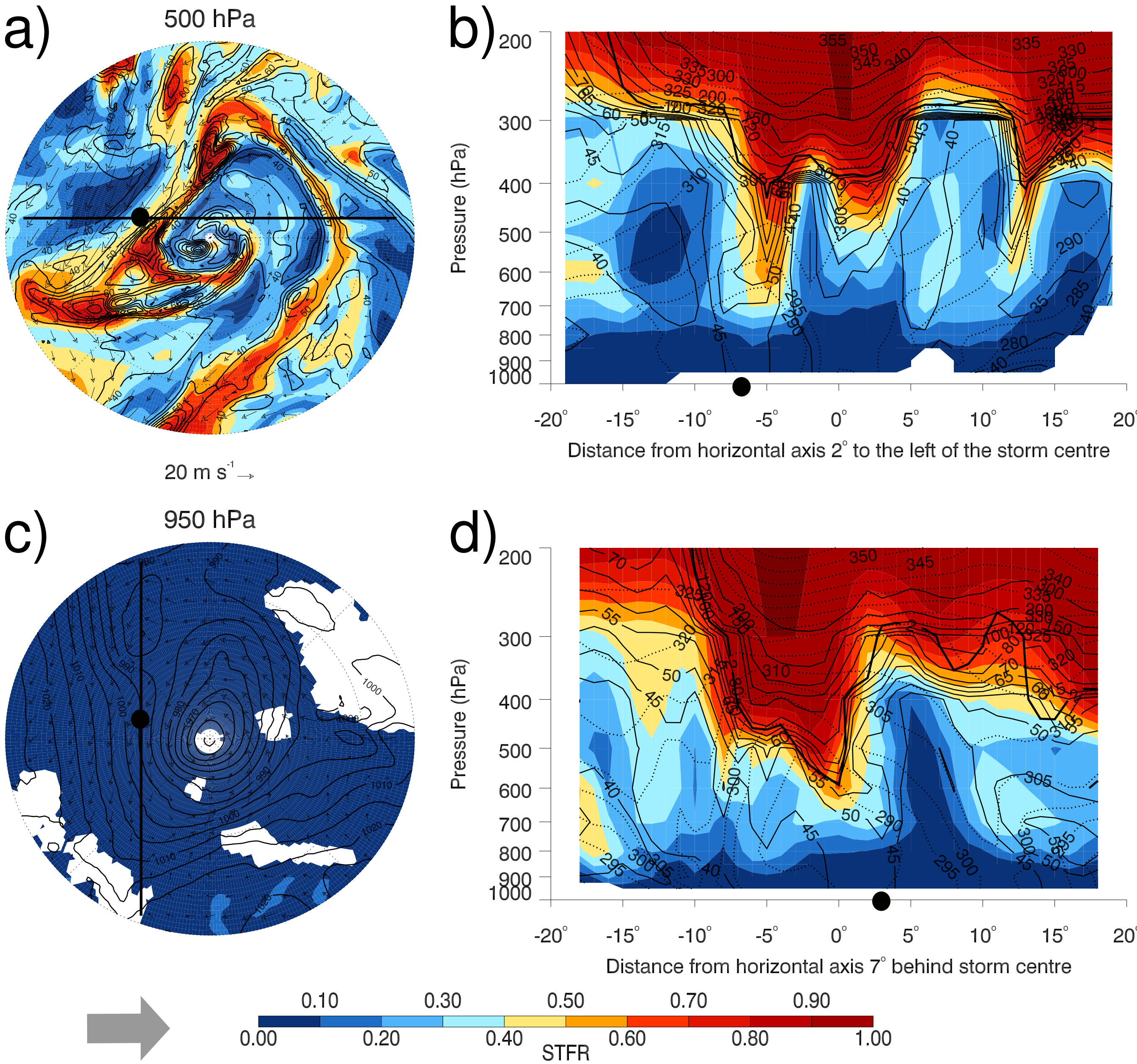

Figure S4. N1 cyclone on 5 March 2007 00UTC; similar to Fig. 7 except cyclone-centered STFR (color) and MERRA-2 O_3 (**a**,**b**,**d**, thin black contours; ppbv, increments matching Fig. 4 color bar) and SLP (**c**, black contours ; 5 hPa increments). Figure also shows MERRA-2 θ_e (**b**,**d**), PV (**b**,**d**) and system relative winds (**a**,**c**). Note, panel **(c)** is 950 hPa instead of 1000 hPa to minimize the missing data plotted. This occurs since the GEOS-5 model masks orography.

S2.2 S1 cyclone TFP and STFR

The frontal zones associated with the S1 cyclone on 25 April 2012 00UTC and 18UTC are shown using the TFP function in Fig. S5. Similar to the N1 cyclone, the front trailing from the center of the S1 cyclone toward the west is diagnosed as a cold front (Fig. S5a) and denoted as such in Fig. 10c. At this time the S1 cyclone has become occluded and the location of the warm front is not clear in the TFP values (Fig. S5a), however it is estimated in Fig. 10c in conjunction with the synoptic analysis chart (not shown). The decaying parent low can be identified by the elongated 1000 hPa MSLP contour to the east of S1 cyclone center in Fig. S5a, with no clear frontal zones. In Fig. S5b, 18 hours later, the frontal zones close to the S1 cyclone are not found at the cyclone center in the TFP values. A portion of the trailing cold front can be identified over 10° to the west of the cyclone center.

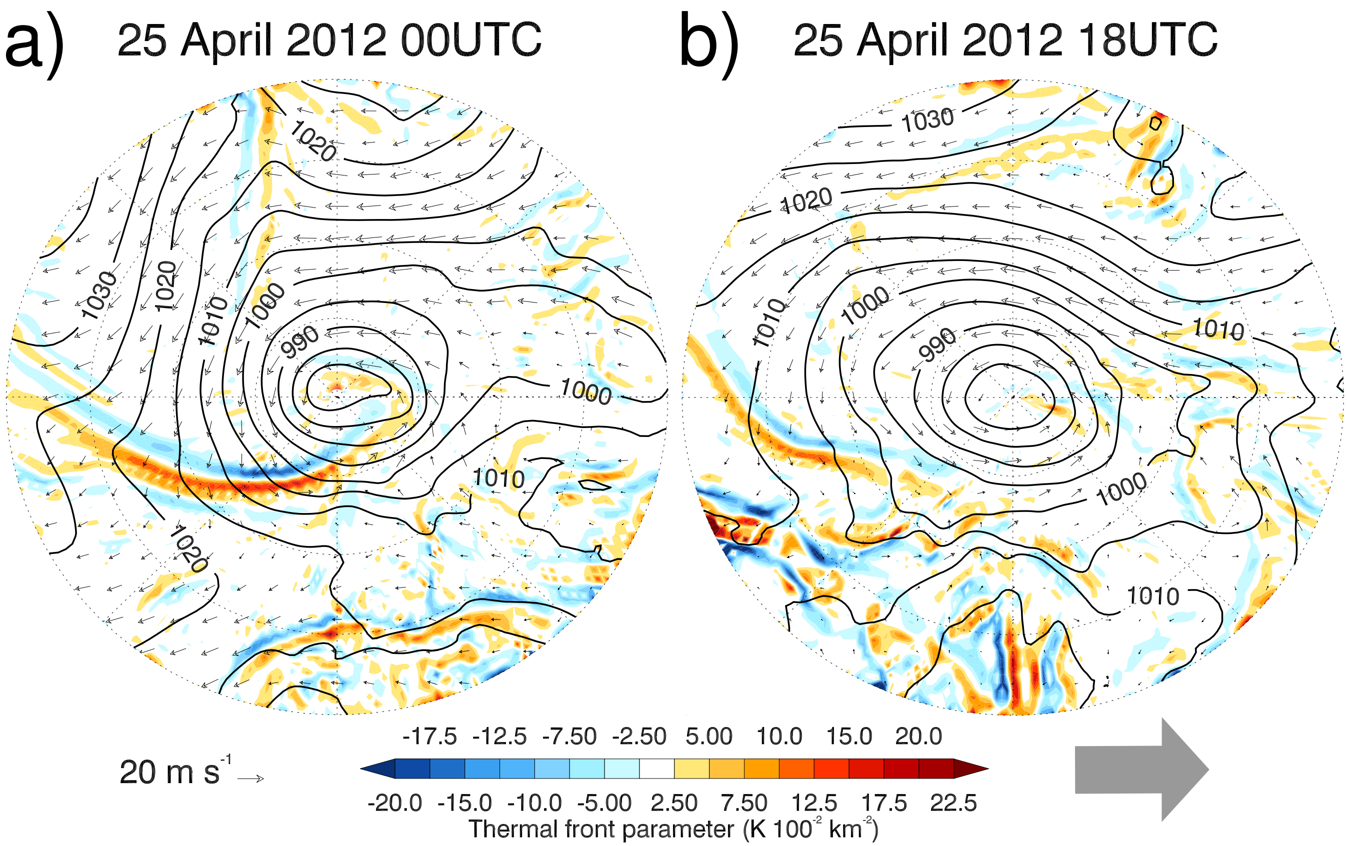

Figure S5. S1 cyclone-centered MSLP (solid black contours, 5 hPa contour intervals), 925 hPa system relative horizontal winds (20 m s^{-1}, reference arrow), and 925 hPa thermal front parameter (color, 2.5 K 100^{-2} km^{-2} intervals) on **a)** 25 April 2012 00UTC and **b)** 25 April 2012 18UTC. Radial dotted lines are plotted every 45° and dotted circles represent 5°, 10°, 15°, and 20° radii from cyclone center. The cyclone has been rotated in order that the direction of cyclone propagation is toward the right, indicated by the large grey arrow. N.b., geographical north for **a)** and **b)** is toward the top of the page.

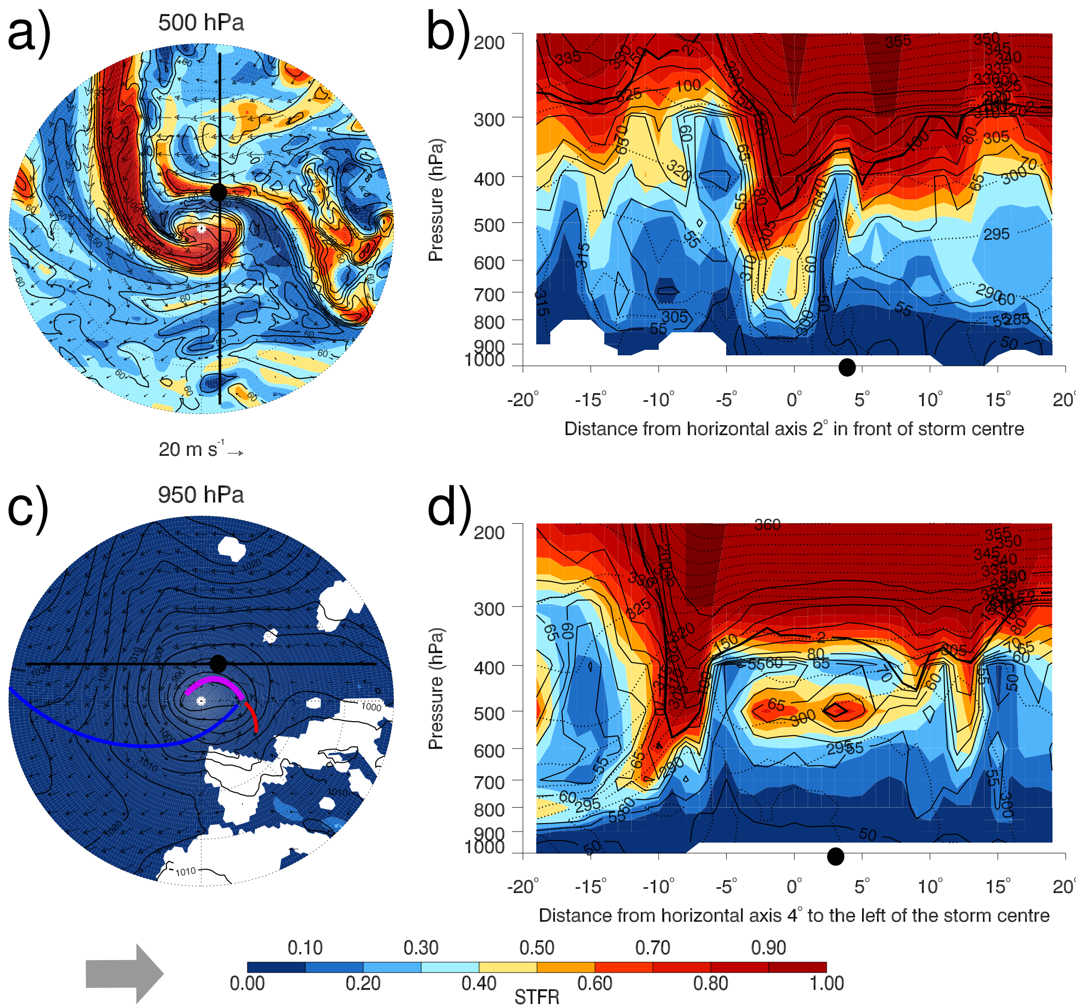

Figure S6. S1 cyclone on 25 April 2012 00UTC; similar to Fig. 10 except cyclone-centered STFR (color) and MERRA-2 O_3 (**a**,**b**,**d**, thin black contours; ppbv, increments matching Fig. 10 color bar) and SLP (**c**, black contours; 5 hPa increments). Figure also shows MERRA-2 θ_e (**b**,**d**), PV (**b**,**d**) and system relative winds (**a**,**c**). Note, panel **(c)** is 950 hPa instead of 1000 hPa to minimize the missing data plotted. This occurs since the GEOS-5 model masks orography.

S2.3 N2 cyclone: synoptic conditions associated with high O_3 at Monte Velho

The N2 cyclone is a strong cyclone which passes north of Monte Velho and advects high O_3 to the monitoring station. It is also an example of a cyclone associated with low O_3 before high O_3 is observed at Monte Velho. Cyclogenesis of the N2 cyclone occurs off the east coast of Florida on 17 May 2006 at 12UTC (not shown) and then tracks northeastward. Downwind of the N2 cyclone are a series of low pressure systems across the NA region as well as a strong Azores High stretching over most of the NA (Fig. S7c, about a day after cyclogenesis, the N2 cyclone is located just south of Newfoundland). The large pressure gradient results in strong westerly winds at $\sim$45°N throughout the troposphere (Fig. S7a–c). At 300 and 500 hPa, there are strong southerly winds on the western side of two upper-level ridges between 20° to 45°N. Between the two ridges there is an O_3-rich (> 80 ppbv) streamer of air reaching south of 30°N (Fig. S7a and b), resulting in high O_3 at lower latitudes that can descend within the DI of the low pressure systems or subside in the Azores high pressure system.

Over the next two and half days, the N2 cyclone continues to move northeastward towards the UK following the upper-level jet stream, the narrow region of relatively strong winds at 300 hPa (Fig. S7a and S8a). During this time, a high pressure system from over the Labrador Sea moves in behind the N2 cyclone, blocking further cyclones and pollutants from being transported from North America across the NA (Fig. S8c). The high pressure block in the western NA is similar to the S1 synoptic conditions presented in Sect. 4.2.2.

In Fig. S8c, the cold front (blue line) associated with the N2 cyclone is approaching Spain from the northeast. The cold front can be identified at 1000 hPa in the wind field, where the wind at the surface near the Iberian Peninsula diverges in different directions, and in the MSLP field where the gradient in the isobars decreases (Fig. S8c). On 21 May 2006 at 06UTC, low O_3 value is observed at Monte Velho. The low O_3 arrives at Monte Velho from the southwest and flows anticyclonically around the Azores High over the NA and northward parallel to the N2 cyclone's cold front as part of the WCB (Fig. S8c). This low O_3 wraps around the cyclone center as the N2 cyclone reaches maximum vorticity (Fig. S8c). It can be noted that the composition near the cyclone center impacts the O_3 observations at Mace Head, located $\sim$15°N north of Monte Velho. The low O_3 within the cyclone center results in persistent low O_3 at Mace Head ($O_3 < 40$ ppbv) as the N2 cyclone tracks over the UK.

High O_3 (> 45 ppbv) first occurred at Monte Velho on 22 May 2006 06UTC (Fig. S9, about five days after the N2 cyclogenesis (Fig. S9d). Prior to the high O_3 events at Monte Velho, there is an increase of O_3 over time in the mid- to low troposphere due to the low pressure systems in the NA bringing O_3-rich air toward lower levels within their respective DI airstreams (Figs. S7b, S8b, and S9b and c). At the time high O_3 is reported at Monte Velho, the N2 cyclone has tracked over the UK (N2 located at 52°N, 2°W, Fig. S9d) and the trailing cold front (blue line) has passed over Monte Velho. The high O_3 at the surface is likely associated with stratospheric O_3 descending within the DI of the N2 cyclone as well as high O_3 subsiding on the eastern side of the Azores High (Fig. S9a-c). High O_3 persisted for most of the period until the end of the month as N2 continued to track north, associated with subsequent descent of O_3-rich air over the Iberian Peninsula behind the front and subsidence within the high pressure (not shown).

To summarize, during the life cycle of the N2 cyclone, successive low pressure systems in the region interacting with the O_3-rich stratospheric air lead to a build up of high levels of O_3 in the mid- to lower troposphere. There is descent of both

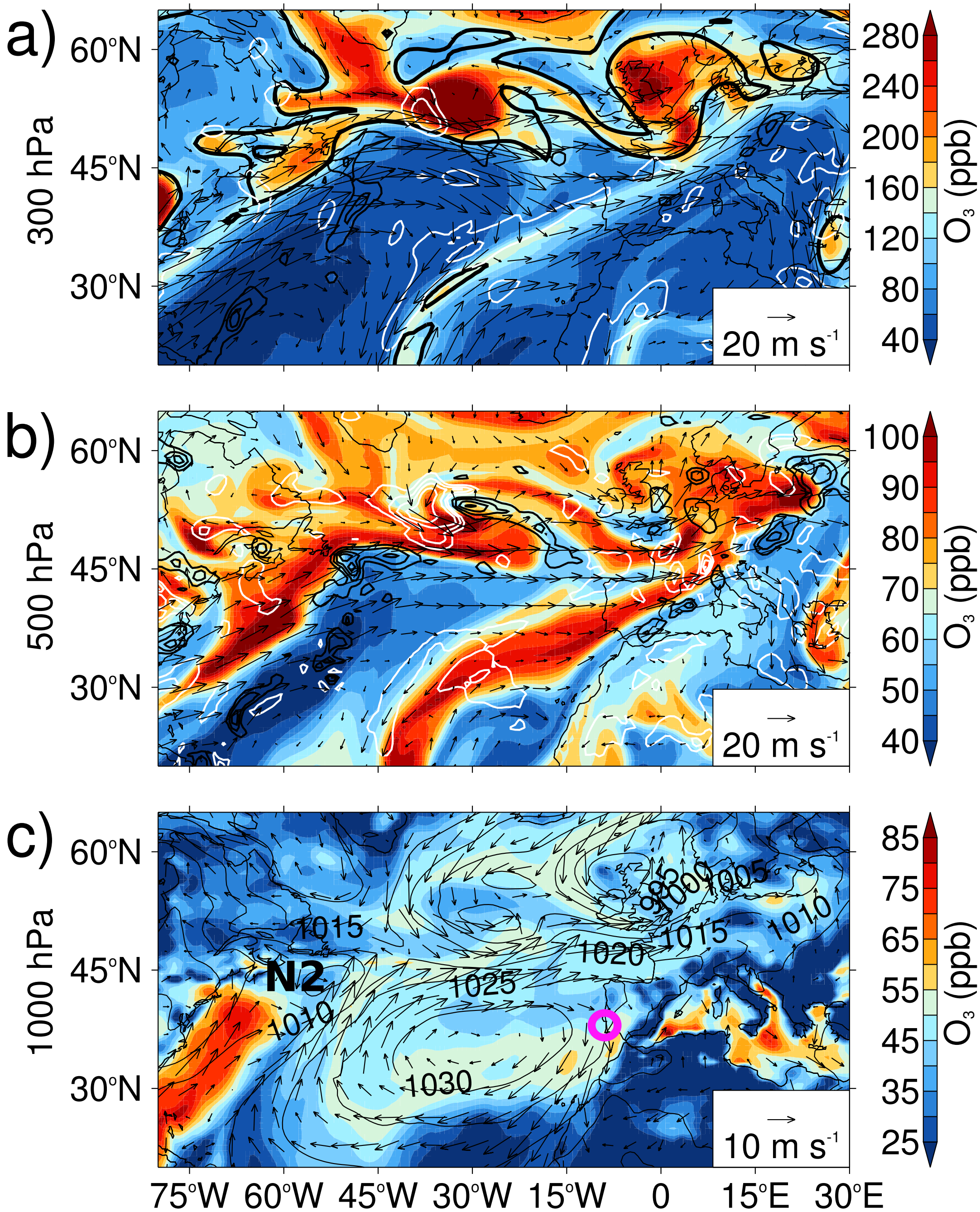

Figure S7. Synoptic conditions and O_3 distribution for the N2 cyclone (**c**; 58°W, 41°N) on the 18 May 2006 at 18 UTC, 2.5 days before maximum ζ_{850} was reached. O_3 (color; note different scales are used) is shown on three levels: **a)** 300 hPa, **b)** 500 hPa, and **c)** 1000 hPa. In addition, MSLP (**c**; solid contours, 5 hPa intervals), ω (**a** and **b**; black contours for positive values indicating ascent, 10 hPa h^{-1} contour intervals, and white contours for descent, -5 hPa h^{-1} contour intervals), and 2 PVU isosurface (**a**; thick contour) are shown. Monte Velho indicated by pink open circle (**c**).

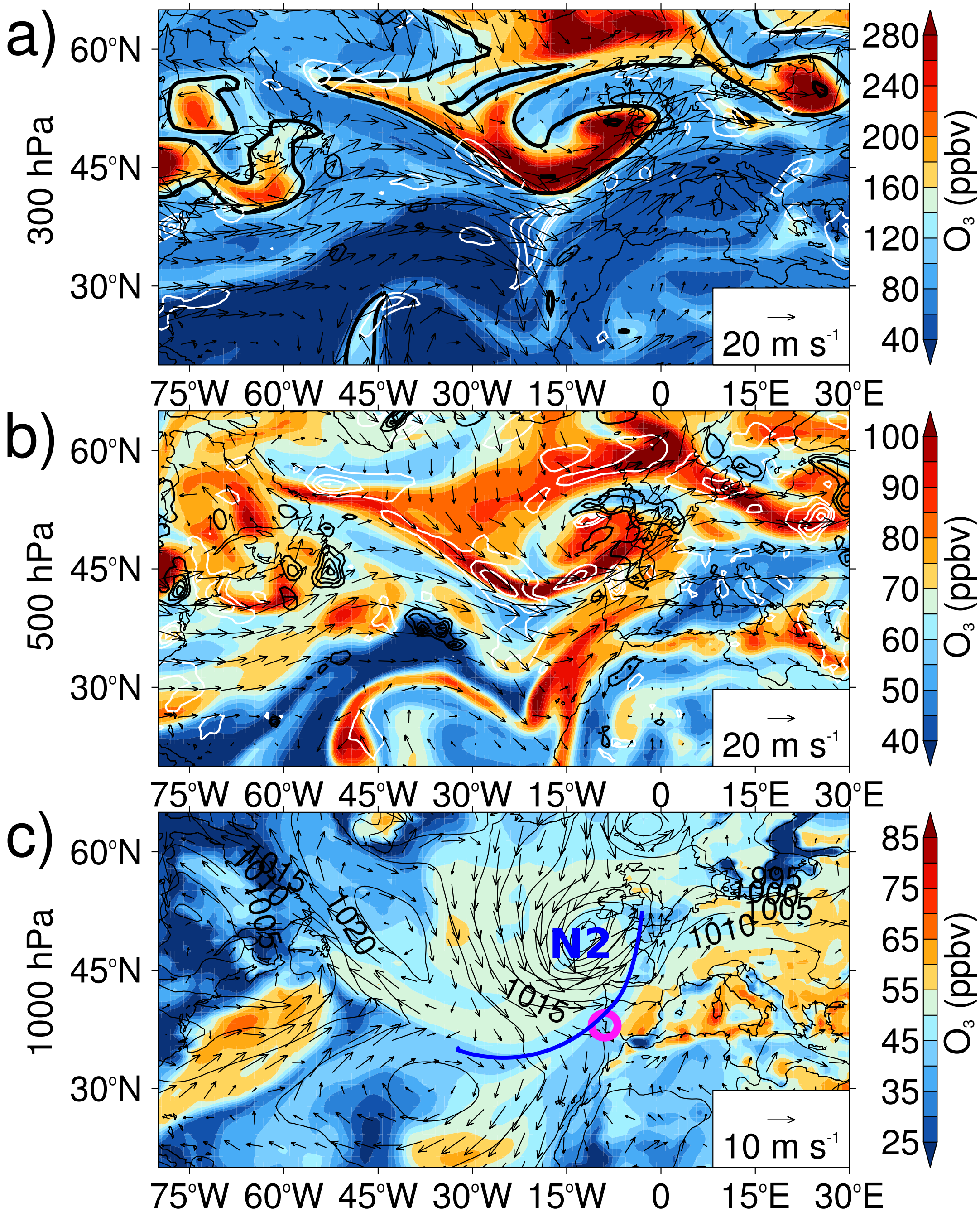

Figure S8. Similar to Fig. S7 for the N2 cyclone (**c**; 15°W, 48°N) except 2.5 days later on 21 May 2006 06UTC, at the time of maximum ζ_{850}. Monte Velho, indicated by pink open circle (**c**), is located to the southeast of an approaching cold front (approximate location indicated by blue line). Low O_3 was observed at Monte Velho.

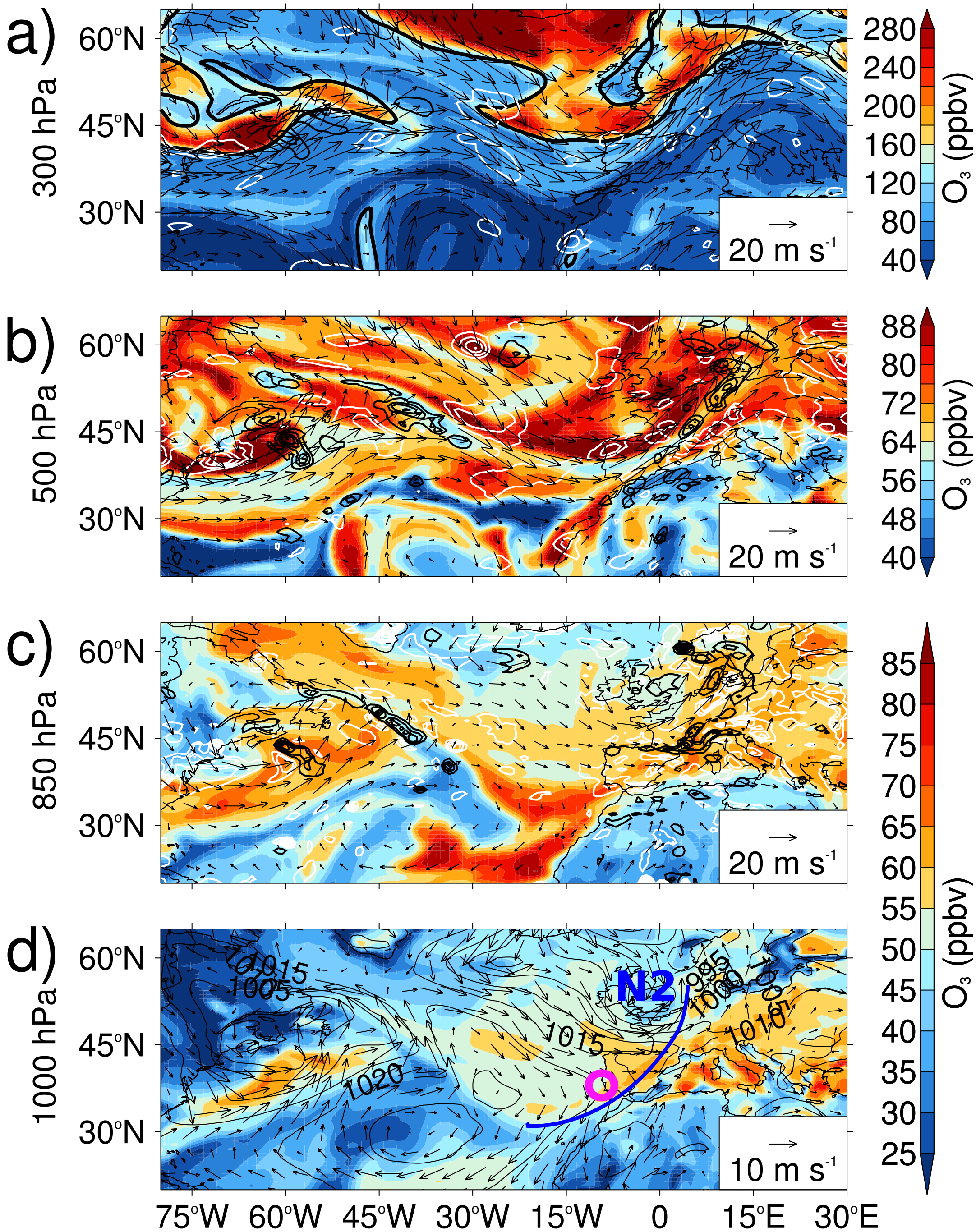

Figure S9. Similar to Fig. S8 for the N2 cyclone (**c**; 2°W, 52°N) except 24 hours later, on the 22 May 2006 06UTC, 18 hours after maximum ζ_{850}. Monte Velho, indicated by pink open circle (**c**), is located to the northwest of an passing cold front (approximate location indicated by blue line). High O_3 was reported at Monte Velho. Four levels are shown: **a)** 300 hPa, **b)** 500 hPa, **c)** 850 hPa and **d)** 1000 hPa.

stratospheric air as well as the residual high levels of O_3 within the DI airstream of the N2 cyclone, reaching the surface over the Iberian Peninsula. As was seen in the N1 cyclone, low O_3 was first reported at the observation station as clean subtropical air influenced the site, prior to the cold front passing over the station. Once the cold front passed over the station, there was a substantial increase in the O_3 recorded. At the time of maximum intensity of the N2 cyclone, the high pressure system was in a prominent position over the western NA, blocking potential transport from North America. Since the synoptic-scale circulation is similar to the life cycle of the S2 cyclone, the cyclone-centered analysis has not been repeated for the N2 cyclone.