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# *Interactive comment on* "The Meteorology and Chemistry of High Nitrogen Oxide Concentrations in the Stable Boundary Layer at the South Pole" *by* William Neff et al.

**Anonymous Referee #1**

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Neff et al. SP NOx

This manuscript focuses on the factors (mainly dynamical, with some attention to chemical processes) that contribute to large, but variable, mixing ratios of NO at South Pole (SP) during the spring to summer transition. Previously unpublished measurements of NO and NO2 during the 2006-2007 field season are presented and discussed in context of observations made in 3 earlier campaigns (1998/99, 2000/01, and 2003/04 seasons). Several previous papers by this team have established that the surprisingly high levels of NO sometimes seen at SP are sustained by snow to air flux of NO produced by photolysis of nitrate in the snow, building up when low boundary layer height

(BLH) limits vertical mixing. Low BLH resulted from low wind speeds and clear skies, with the latter also enhancing the rate of nitrate photolysis. Intervals with low BLH generally coincided with easterly winds, leading to a suggestion that some of the NO during extreme events was transported down slope to SP, with extensive recycling of redeposited nitric and pernitric acids combined with non-linear NOx chemistry resulting in mixing ratios approaching and even exceeding 1 ppbv.

The addition of the 2006 results confirms earlier findings but does not produce any fundamental new understanding. However, this manuscript does extend prior papers by exploring the links between BL dynamics at SP and synoptic and mesoscale dynamics in the upper troposphere and even into the stratosphere over Antarctica. In my opinion the key findings are that high NO at SP are most likely to occur when the flow at 300 hPa is from SE, which is found to correlate with relatively weak surface winds from the E-S quadrant and generally clear skies, factors leading to low BLH. It is also pointed out that the south-easterly flow at the surface is due to large scale pressure gradients, and not katabatic flow as had earlier been suggested.

These findings are significant enough to merit publication, but I find the manuscript overly long, repetitive at times, and hence not as accessible as it could be. In particular, I find that a lot (too much) of the discussion relies on figures in the supplemental material, and that graphical or tabular comparisons of NO in the 4 different seasons are never provided in the main body of the manuscript. Figure S10 does provide such an overview, but I suggest it would work better as a 4 panel plot (one panel for each season) and should be moved out of the supplement to someplace early in the main text. Current version of sections 2.1 and 2.2 go into great detail about meteorological similarities and differences across the 4 sampling seasons, but rarely relate these to observed NO concentrations at SP (which seems like it should be the main point of paper). Referring (probably often) to a revised figure S10 put into section 2 would help. Similarly, Fig 2 would be improved by adding some kind of NO metric (e.g., season mean, or fraction of sampling hours > than some threshold (250 pptv is used





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elsewhere)), and the heavy black line showing average NO as function of surface wind direction in Fig 3 now averages all 4 seasons, unnecessarily obscuring interannual variations which are important and interesting. In the following list of detailed comments I mix purely editorial suggestions (typos, etc.) with comments related to streamlining the narrative by reducing redundancy and interesting side stories. Comments are linked to line numbers in the on-line pdf, hope these are conserved across different platforms.

69-73 Would help to indicate the length of these weather cycles. Seems the timescales are a few days of clouds, then a few with clear skies and strong inversion based on case studies presented much later, but here the reader could assume you mean seasonal changes.

73 I would argue that high albedo and low SZA are characteristic of most of Antarctic plateau. What is "unique" about SP is that the SZA barely changes within 24 hour day. Later on it seems the SZA is felt to be significantly lower at SP than at 75 or 80 S, but is that true averaged over a full day? Might help just to be quantitative about meaning of "low" SZA.

78 "omnipresent" seems an odd description of the inversion, given you just said the really stable boundary layer reforms every weather cycle.

87-88 words missing here, like "Antarctica" or the Antarctic ice sheet. Does not make sense to describe storm track as pole centric, or covering so many

95-96 Does not seem the semi-annual oscillation or the circumpolar trough are ever mentioned again, so why introduce the terms here? Are they controlling NO at SP?

98-100 Another interesting tidbit about large scale flow in winter that seems to have nothing to do with NO at SP in Spring/Summer

103-104 Is it fair to compare one 135 degree sector to another that is only 90 degrees? I also note that if wind direction was completely uniformly distributed the larger sector would account for 37.5% and the W to N quadrant 25%, making the quoted 43 and 31





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% fractions observed only slightly enhanced. Figure 1 makes the point, so consider deleting this sentence.

109-113 These last 2 sentences repeat ideas introduced earlier (in this paragraph, also in first paragraph of section 2, and in earlier papers). It is also odd to be making predictive statements about how NO should respond, rather than point to data that confirms the points discussed. Granted, the 2006 results have not yet been show, but the other 3 campaigns are published.

114-121 Perhaps unnecessary detail, the correlation is interesting and useful for chemists on its own.

121 can, however, vary—-→ varies

124 mention 1999 and 2002 and say they coincide with a single year with sudden strat warming. Which one had the warming?

125 Any decadal periodicity is not at all obvious in Fig 2. Even if it is there, is it important to understanding any differences between the 4 years (spread over 9 years) with observations at SP.

129-130 Is it really a new finding that NO at SP is not tightly controlled by any single parameter? Seems this has been noted in earlier papers.

134-170 A little confusing here, with first sentence saying SE winds aloft are associated with E winds at the surface, but figures S2 and S3 show dominant surface wind mode to be 70-210 or 60-180 which are also SE. Also note that last sentence in block mentions SE surface winds.

151 Does Fig S4 need to show full annual cycle?

153-154 Statement that Fig S4 “shows” that geostrophic winds dominate katabatic forcing of surface winds in peak summer kind of has to be taken on faith (no details about calculations supporting this until discussion of Figs 7 and 8 around line 390).

Can this idea be presented clearly just once in the manuscript (it does seem to be a finding authors want to stress).

167-170 It is usually risky to assume transport path based on wind direction observations at a receptor site. Do trajectories follow the contours as suggested? Strictly speaking, Fig 1b suggests that air traveling from Concordia to SP along 123 would cross at least 1 contour and flow uphill for almost the last half of the trip.

170-172 These closing sentences are problematic for at least 2 reasons. First is that heading out of SP along 110 is more “uphill” toward the central dome than just discussed, and one could imagine weak katabatic flow being influenced by the subtle topography east of SP shown in Fig 5 (and discussed later). Second is that very high NO at the same time winds and BLH are both high sort of explodes the entire conceptual model developed up to this point, so it hardly seems fair to ask the reader to take these observations as being in support of suggestion that summer time katabatic winds are over emphasized in the literature, and then dismiss them as being unusual.

184-191 Is it important to know about evolution of vortex dynamics since 1960, or are the key points the different date of breakup in the 4 study years, and the fact that the O3 hole results in higher actinic flux while the vortex is still intact?

192-199 this paragraph mainly restates comments made earlier

200-213 Of mild interest perhaps, but what is the quantitative link to NO at SP?

215-222 Nice verbal summary of Fig 8, but no link to NO at SP

223-229 Another paragraph linking large scale and BL dynamics that almost makes predictions about impact on NO at SP, but does not test them with the data at hand (note that cloudiness in the 4 years mentioned in line 224 does not directly correlate with frequency and length of high NO episodes, 2003 does have high NO and few clouds, but 1998 has most clouds and second highest NO).

230-247 Nice case study, and reassuring that it shows features observed in other





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events in the 3 earlier seasons. Just seems kind of late in the manuscript to finally be getting to NO observations at SP in any detail.

290-291 Pretty sure that Berhanu et al. and Frey et al. feel these sequences are not coincidental. Beside that, they must have physical/chemical explanation, even if not all the details have been nailed down.

295-420 Please make it more clear what new insights are gained from the 2006 data set. Mostly seems to just reinforce things that were seen before.

300-302 Can't see any hash marks plotted at -20 in Fig 6.

324-338 A little unclear how the comparisons in the early part of this block lead to suggestion that late Dec emissions in 2006 were low due to photobleaching of the snow. In the day 305-340 block NO was 2 x higher in 2003 with similar wind/cloud statistics, but then in Dec the mean concentrations were similar despite 2003 being cloudier with deeper BL. Seems that maybe 2003 started with more nitrate in the snow for some reason, but in 2006 the lower amount available to make NOx kept doing so longer due to favorable BL dynamics (e.g., average decreased by factor of 2.2 in 2003 in response to seasonal change, while in 2006 the early season to late season difference was only factor of 1.4)

351 as noted earlier, seems surface winds tend to be from SE, not E, when 300 hPa winds are SE

352-353 Not very likely that much NOx is transported in FT to SP

354-356 Just mentioned these earlier studies and suggested that maybe the link was just random.

356-372 What motivates this rehash of 2003 results in a section on "Case studies and insights from new 2006 data"?

385-400 Nice development of the argument against significant katabatic forcing, but



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it feels redundant since the finding was earlier declared (with little support first time around)

403-410 Here is another example of using case study to make a solid point, that was earlier just boldly declared (lines 70-74). If this is so well established that it is in introductory remarks, does it need support here? Or should the earlier section be pulled back a little.

420-446 Could/should these details be moved to supplement? Key point is to use the estimated BLH to estimate NO fluxes.

455 measurements at SP

461 associated lifetime

513-522 Think another important aspect of these dynamical findings is that none of them, alone, had strong direct correlation with NO concentrations or fluxes at SP. This point is raised several times in the rest of manuscript, why not here?

574 because of potential

647 and 649 is the “0.000 level” correct?

817 the red line in 4a is not that easy to discern from black, try lighter shade

Figure S3, red diamonds in a and b seem same maroon as in Fig 4a, very close to black. Try lighter hue

Figure S5 caption, what do you mean by “year-to-year scatter is not unreasonable”?

Figure S6, Not sure this figure is central to the NO story, but if it stays may need to say something about what is so significant about the Breakup date. In both intervals it certainly looks like hole is filling weeks earlier. Also, why is the early interval averaged 1964-1980 (dots) but break up date averaged 1961-1980?

Figure S9 caption. Figure 3 does not show any basin. Better callout would be Fig 5





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(possibly Fig 1, but suspect 5 is better).

Figure S10, as noted in text, would probably be better as 4 panels. And it should be in the main paper, not supplement.

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