



Boundary Layer and Synoptic Effects on NO Concentrations at the South Pole: A Multiyear Perspective

William Neff (1) and Douglas Davis (2)

(1) University of Colorado, CIRES, NOAA/ESRL/PSD, Boulder, United States (william.neff@noaa.gov), (2) Earth & Atmospheric Sciences, Georgia Institute of Technology, Atlanta GA

A series of experiments have explored the behavior of NO concentrations at the South Pole as part of the ISCAT (1998, 2000) and ANTCI (2003, 2005, 2006-7) field programs [Davis et al., 2008]. The relationship between NO and boundary layer depth (BLD) proposed by [Davis et al., 2004] was verified by [Neff et al., 2008] using direct sodar measurements of BLD during the period November-December 2003. A longer time series of NO_x was generated in the ANTCI program from sunrise in 2006 into summer 2007. However, no direct BLD measurements were available. To address this deficiency, we used multiple linear regression on data from 2003 where both directly observed BLD and meteorological variables were recorded. This analysis showed that the three most important variables were wind speed ($r^2=0.56$), Delta T_{2-22m} ($r^2=0.32$), and wind direction ($r^2=0.10$). The strong dependence on wind speed is consistent with the results of [Neff et al., 2008] showing the dependence of BLD on surface stress (representing turbulent mixing of momentum to the surface). The dependence on wind direction may be unique to the South Pole because of the constancy of surface winds from the northeast that are weakly perturbed by synoptic weather systems: winds from grid east tend to be light, colder, and with shallower BLD whereas those from grid north are stronger, warmer, and have greater BLD. To further test these regression results, we used lower resolution sodar data from the austral spring of 1993 (e.g., October/ November). From these data we found that applying the 2003 regression analysis results to 1993 data, ~32% of the variance could be accounted for, despite the coarseness of the 1993 observations. The latter result provided the justification for applying the 2003 BLD regression analysis to our estimating BLDs on the 2006-7 NO_x data set.

As found in the 2003 data set, the general trend in the 2006-7 data showed that predicted shallow BLDs consistently correlated with higher concentrations of NO_x. One of the unique features of the 2006-7 data was the capture of the full transition from winter to summer in terms of insolation, temperature, and a wide variety of weather regimes. As a result, an important new finding from this analysis was the apparent dependence of the level of observed atmospheric NO_x on local temperature as proposed by Davis et al., 2010. These authors have further suggested two possible mechanisms that might explain this trend. The first of these involves the local availability of high concentrations of HO₂NO₂ at South Pole (SP) as reported by [Davis et al., 2008]. This reflects the fact that HO₂NO₂ is stable only at very low temperatures. As a result it represents a significant loss process for atmospheric NO_x at this site. However, since HO₂NO₂ also has a much larger photochemical absorption cross-section in ice versus HNO₃ (a molecule also involved in the removal of NO_x), it defines a much more efficient recycling mechanism for returning nitrate to the atmosphere. A second hypothesis put forward by Davis et al. 2010, involves the possible role of ice-surface adsorbed HNO₃. In this scenario the density of isolated HNO₃ molecules on ice crystals is strongly influenced by the levels of atmospheric water vapor. The latter, of course, is a strong function of temperature. Higher temperatures would lead to elevated H₂O thus leading to enhancements in the burial rate of surface adsorbed HNO₃. The latter mechanism could be of critical importance since surface adsorbed HNO₃ would have a much higher quantum yield for photochemical production of NO₂ than in-situ bulk ice HNO₃ which when photolyzed experiences significant cage effects that greatly reduced its recycling efficiency.

Using our BLD estimation technique, we stratified the NO_x-Temperature relationship reported by [Davis et al., 2010] by boundary layer depth. What was found was that NO_x concentrations systematically decreased with both increasing BLD as well as temperature. As noted by Davis et al. (2010), virtually no other global polar site has shown the high levels of NO_x that are routinely observed on the plateau during the Austral spring/summer time period, particularly as recorded at SP. Of particular note are the recent spring/summer observations at Summit Station Greenland [Van Dam et al., 2013]. These data revealed very low levels of NO (typically less than 20 pptv) and no BLD height dependence. Thus, they would appear to be consistent with the hypotheses of Davis et al. 2010, since the temperatures there were typically greater than -25C.

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