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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Abiotic and biogeochemical signals derived from the seasonal cycles of tropospheric nitrous oxide" *by* C. D. Nevison et al.

Anonymous Referee #2

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GENERAL COMMENTS

This study examines the different influences (abiotic vs. biogeochemical) on the seasonal cycles of atmospheric N2O derived from surface observations from four separate measurement networks. A focus of the study is on identifying the influence of stratospheric low N2O air at the measurement sites, and on evaluating its contribution to observed inter-annual variability in the seasonal cycle. This analysis follows up on previously published work, by the lead author and others, suggesting a significant role for stratospheric influence on tropospheric N2O.

The study makes a valuable contribution in compiling the N2O measurements from the

different networks, in comparing the seasonal cycles at the surface sites sites, and in discussing potential factors underlying observed variability. It is generally well-written, and is within the scope of ACP.

However, as presented here, the analysis of and conclusions on stratospheric influence are not fully convincing. Correlations between lower stratospheric temperatures and N2O seasonal minimum anomalies are presented as the primary evidence, however there is insufficient exploration of other possible causes of variability (e.g., in biogeochemical land-based processes). In addition, the inter-comparison of measurements from the different networks (while a useful exercise) also presents obstacles to clear identification of factors governing the N2O seasonal cycle. Observed seasonal cycles derived from the different networks demonstrate significant differences at certain common sites; this results in inconclusive findings at these sites, and the attempts to account for these differences among networks obscures the main focus of the study.

I suggest the authors revise the manuscript to focus on the primary aim of their study, and clearly identify their robust findings. They should also present a more convincing case for their conclusions on stratospheric influence, in particular, by evaluating in more detail other potential causes of inter-annual variability (e.g., in land-based biogeochemical fluxes). More detailed comments are given below.

1)Focus of study : The authors should focus clearly on their stated aim; i.e., an investigation of causes of variability in the observed N2O seasonal cycle. The intercomparison of measurements from different networks at the same sites (in sections 3.2 and 3.3) highlights differing behavior (e.g., between NOAA/CCGG and AGAGE) leading to inconclusive findings on governing processes. The accompanying discussion of these observational differences often covers a range of possible causes with no clear conclusion on which set of measurements are more representative. This is often confusing, and obscures the focus on the variability in the seasonal cycle. The authors should identify early on in the manuscript which set of measurements will be relied on in reaching their final conclusions.

2)Evaluation of stratospheric influence : The inference of stratospheric influence on lower tropospheric N2O is based primarily on the correlation analysis of high-latitude lower stratospheric temperatures with N2O seasonal minimum anomalies. This analysis is not always convincing, particularly when the authors note : (1) that they 'cannot rule out the possibility that these correlations could arise through a common driving factor such as tropospheric weather anomalies' (section 3.3.1, p. 25813, lines 10-15); (2) that late summer minima in the seasonal cycle at some Northern Hemisphere sites can also be obtained in model simulations representing primarily tropospheric processes, with no stratospheric N2O sink. The authors need to present a clearer case indicating which of their findings are robust indications of stratospheric influence at tropospheric sites.

3)Biogeochemical fluxes : There is relatively little focus on the 'biogeochemical' processes that may lead to variability in the seasonal cycle. There is some discussion of ocean fluxes influencing measurements at Trinidad Head and at some Southern Hemisphere sites. However, since land-based fluxes constitute well-over over 50% of surface emissions (Denman et al. 2007), the potential influence of variations in these fluxes should also be discussed more fully to fulfill the stated aim of this study.

4)Uncertainties associated with the analysis: There is little discussion of the uncertainties associated with the analysis and their implications for the conclusions. This is relevant for the derivation of the seasonal cycles (section 2.2, also see comment on detrending below), and in the use of the various proxies (especially those derived from ocean and atmospheric model analyses) employed in the correlation analysis (sections 2.3, 3.3). The authors should present estimates of uncertainties associated with the variables used in the correlation analysis, and discuss the implications for the robustness of their conclusions.

5)Detrending of atmospheric measurements : It would be useful to have some indication of the sensitivity of the derived seasonal and interannual anomalies to the detrending procedure used. There is little detail presented on the algorithms used, and how

they were applied (e.g., universally across all networks and sites ? or were specific polynomials derived for each site and network ?). It is therefore difficult to assess what exactly has been subtracted from the original time-series. Much of the study's analysis relies on the derived seasonal and inter-annual residuals following the detrending procedure; the study would be more convincing with some demonstration that these derived quantities and the resulting conclusions are not sensitive to the detrending procedure used. (The authors themselves allude to this possibility in section 3.3.1, p. 25813, lines 25 -28)

SPECIFIC COMMENTS

1)Abstract : p. 25805 : Lines 25-30 : '…..surface mixing ratio data by themselves are unlikely to provide information about seasonality in surface sources (e.g., for atmospheric inversions)…..' This is unclear and should be clarified. An atmospheric inversion, through its use of a transport model and 'best estimate' prior fluxes, should represent several of the relevant abiotic processes, and therefore enable estimation of these separate influences. The authors should indicate more precisely the limitations of the surface mixing ratio data in estimation of governing processes (e.g., inability to resolve correlated fluxes, sparsity of sampling, etc.)

2)Section 2.2, p. 25808 to 25809 : The discussion of the detrending procedure needs more quantitative detail on methods. What is the form of the 3rd order polynomial fit used ? Was the same form used for all sites and networks ? How sensitive were the derived anomalies to variations in the assumed polynomial ?

3)Section 2.4 Thermal signals : This section would benefit from more discussion of the uncertainties associated with estimating the thermal signal, and the implications for the study's conclusions.

4)Section 3.2 : Are there no pollution event filtering procedures used for the NOAA/CCGG measurements at MHD ?

5)Section 3.3.1, bottom p. 25813 to top p. 25814 : 'Atmospheric growth rate. . .resulting in a "flatter" curve being subtracted. . ..'. This is unclear. Please clarify.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 25803, 2010.