

Interactive comment on “Impact of mixing and chemical change on ozone-tracer relations in the polar vortex” by R. Müller et al.

Anonymous Referee #2

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This paper presents a rather complete analysis of the issues and limitations related to the use of tracer-tracer (specifically, ozone-tracer) correlations as a quantitative diagnostic of polar chemical ozone loss. While this technique has been widely used for decades in the analysis of polar ozone depletion, several authors have criticized their usefulness in quantifying ozone chemical loss in the presence of transport. The spatial distributions of trace gas mixing ratios are dependent on both chemical and dynamical processes. However, compact correlations between the mixing ratios of long-lived species are quite well established in the lower to middle stratosphere provided that rapid isentropic mixing is dominant. In favorable cases, these correlations can be used as indicators of mixing processes and, ultimately, distinguish between air masses

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of different physical origins.

For the polar vortex, in particular, compact correlations between the mixing ratios of trace gases are established such that relationships inside the vortex are significantly different from the canonical midlatitude relationships. The key word here is “tracer”, such that mixing ratio variations are dominated by transport rather than chemical lifetime. This point is indeed addressed carefully in the paper with regard to the establishment of the early vortex ozone-tracer relationship; in the early vortex, the absence of sunlight makes ozone a good “long-lived” dynamical tracer. However, the conditions for generating the compactness of this early winter vortex reference are closely tied to the evolution, and indeed the weak extra-vortex mixing processes competing with the rather rapid diabatic descent and rapid intra-vortex mixing. The main criticism concerning the use of ozone-tracer correlations occurs when the photochemistry is activated by the return of sunlight. Ozone changes on isentropic surfaces are now affected by both relatively rapid transport and chemistry. Transport here reflects both horizontal and vertical transport. Vertical transport is caused by diabatic effects only and hence can be followed by the gradient in tracer mixing ratio with potential temperature and the cooling rates. As a direct consequence of the rather weak correlation “curvature”, differential descent with rapid mixing will have little effect on the magnitude of the ozone tracer correlations for even unrealistically large cooling rates, however, extra-vortex entrainment can have a significant effect on the correlation. Several studies suggest that owing to the neglect of this extra-vortex mixing on the tracer correlations, ozone loss may be overestimated. The authors present a rather compelling and complete analysis that show, on the contrary, including such effects will actually lead to an underestimate. The discrepancy here can be traced to the conditions imposed on the model studies where it was implicitly assumed that the same ozone tracer correlations hold both inside and outside the polar vortex. This is clearly not the case for ozone N_2O (or CH_4) correlations where extra vortex ozone mixing ratio is significantly larger for a given tracer isopleth. There are, however, some issues related to the unique usefulness of the techniques when the vortex is weaker. Under such conditions, periodic warming

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events are more prominent and hence ozone loss is smaller, owing to the lack of extensive PSC coverage. In such cases, the vortex can be viewed as much more porous with regard to extra-vortex air and the combination of this with the intra-vortex mixing and differential descent can lead to an ambiguous evolution of the ozone tracer correlations from that of the early vortex reference. Here, it may require further insight from true “tracer-tracer” correlations to completely segregate the chemical changes from the dynamical changes. For example, CFC/N₂O or CFC/CH₄ correlation may be the only choice to independently follow the dynamics. Using N₂O/CH₄ will not necessarily work since the correlations throughout the lower stratosphere polar vortex are nearly linear and thus provide no useful information (contrast) concerning end-point mixing processes.

Overall, I agree with the major conclusion of the paper, namely, if a reliable early vortex reference can be obtained and if vortex measurements are well separated from the extra vortex, then (and only then) can ozone-tracer correlations provide a reliable tool for the quantitative determination of chemical ozone loss in the polar vortex. It is appropriate that in this context the authors preface their conclusion with the phrase “if applied with care...”, I couldn’t agree more and only wished other publications had followed a more “careful” approach.

Overall, I find the paper to be both clear and well written. It builds upon theirs (and other authors) previous studies over the last decade concerning value ozone-tracer correlations as a tool to deduce chemical ozone loss in the polar vortex. While there is no real new science here, it does a rather complete job of addressing a few of the key concerns related to the criticisms brought forth over the last several years regarding the usefulness of the approach in separating the effects of extra-vortex transport. Additionally, the authors do an excellent job in combining the observational arguments with a detailed Lagrangian Chemical Transport Model and in the end, providing a complete picture.

In summary, I find that the subject matter and scientific quality are appropriate for

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