

***Interactive comment on* “Detection of pollution transport events southeast of Mexico City using ground-based visible spectroscopy measurements of nitrogen dioxide” by M. L. Melamed et al.**

Anonymous Referee #3

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Major Comments

The present study presents ground-based vertical DOAS measurements of nitrogen dioxide and comparisons with in situ NO_y, NO₂, and NO measurements during the March 2006 MILAGRO campaign. Employing ceilometer measurements of the vertical mixing height in conjunction with the above measurements the authors infer vertical mixing of NO₂ for a number of case studies at their sampling site south of Mexico City. Although the authors present a nice discussion for the utility of their approach using the combined measurements, this paper does not contain enough new information regarding overall pollution transport, chemical transformations, and/or photochemistry

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to and from Mexico City other than a few isolated case studies of transport. Thus, the present study really does not add to our knowledge base about pollution from a megacity, despite the tremendous potential of their combined measurement approach. It would be much more effective to use their combined measurements to support other measurements at their site to tease out some new aspect of pollution photochemistry, perhaps related to transport and/or vertical mixing. Alternatively, the authors could embark on a several month-long study to characterize some new aspect of overall pollution transport. For these reasons, this reviewer does not recommend publication in ACP in its present form. The authors should instead consider re-submitting their paper after they provide a more substantive story on the atmosphere over the Mexico City basin. Alternatively, the authors may wish to consider submitting their paper to another journal that focuses more on techniques development.

Minor Comments After addressing the above major issues the authors should then consider the following minor comments. This paper should have been written for a more general atmospheric audience rather than focusing on the DOAS community. Specifically, a number of terms and concepts particular to DOAS practitioners were not defined and/or further explained, and these are pointed out below. 1. Page 4772, 2nd sentence in Section 2: change “nitrous oxide” to “nitric oxide” 2. Page 4773, 3rd line down: the authors should indicate that a discussion of the acquisition of the background spectrum in a non-polluted atmosphere will be discussed in a later section. 3. Same page further in paragraph: reword the expression “extra-atmosphere spectrum” 4. Page 4774: the authors should explain the units for the O₂-O₂ complex for those not familiar with DOAS fundamentals. 5. Same page, the authors need to indicate that the use of the O₄ complex in qualitatively restraining the interpretation of the enhancements of the NO₂ VCDs will be discussed in a later section. The reader is left hanging what this means in the present text. 6. Page 4775: 1st paragraph: Further explain and discuss DOAS jargon “Ring cross section” and the “shift” and “stretching” of the foreground spectrum to align it with laboratory spectra. Why is this needed? Are pressure shifts and/or pressure broadening important here or are the laboratory spectra not

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correct? How are the DOAS retrieval errors determined? Using the residuals in Fig. 2 this reviewer estimates an order of magnitude larger error than the value stated. What is the estimated limit of detection? 7. Page 4776, 3rd paragraph, 3rd sentence: correct the spelling of “artifact” 8. Page 4777 in Section 2.3: The authors should indicate the type of NO₂ to NO converter employed and whether the lower limit of detection of 50 pptv pertains to all 3 species. This is needed because certain NO₂ converters may yield artifacts. 9. Page 4780 in the discussion of plume 2 in Fig. 6 in 1st paragraph, this reviewer estimates that the NO₂ VCD reaches a maximum of only about 1.3×10^{16} and not 2.0×10^{16} . Please explain. This is important, since the author’s argue that the increased surface NO₂ mixing ratio between plumes 2 and 3 is countered by the increased mixing layer height. In Fig. 6 the mixing layer height of plumes 2 and 3 look to be approximately 0.4 and 0.8 km, respectively, and thus from their argument one would expect a VCD increase of a factor of ~ 2 from about 1.3×10^{16} molecules cm⁻² to about 2.6×10^{16} for plumes 2 and 3. This is not consistent with the factor of ~ 3.7 increase observed in VCD from Fig. 6. Please explain this inconsistency here and in Fig. 8. Also this simple analysis assumes a uniformly mixed mixing layer and that the elevated NO₂ observed by the VCD measurements are in this mixed layer. One could also explain plumes 2 and 3 by an inhomogeneous mixed layer coupled with elevated NO₂ VCD aloft from another air mass outside the mixed layer. Another troubling aspect of this analysis is the fact that the DOAS measurements did not observe anything for the 1st plume, which is nearly identical in mixing ratio as the 2nd and 3rd plumes and appears to have the same mixing height as the 2nd plume. Please explain. 10. Vertical DOAS measurements at different slant heights could provide more information regarding vertical distributions, and the authors should consider adopting this approach to their suite of measurements. 11. Page 4783, 9th line from bottom regarding uptake of NO₂ into convective clouds: the author should consider another word other than uptake (perhaps “ingestion”) because uptake could also be confused with uptake of NO₂ into the liquid phase, which of course would result in diminished VCD measurements of NO₂

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