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***Interactive comment on “Novel application of satellite and in-situ measurements to map surface-level NO<sub>2</sub> in the Great Lakes region” by C. J. Lee et al.***

**C. J. Lee et al.**

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The manuscript by Lee et al. describes an analysis of measurement results from satellite and in situ-measurements in the Great Lakes region. This comparison study of spatial and temporal variations of NO<sub>2</sub> concentrations provides some interesting insights about the measurement techniques and could be published in the ACP special issue BAQS-Met after some major revisions as discussed below.

*We thank the reviewer for the comments and have carefully considered all the major revisions discussed below. We hope that the steps we have taken to address the reviewer's concerns have made the work more intelligible and that the revised version*

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*is worthy of being published in ACPD.*

**General comments:** The presented study provides a detailed analysis of the data sets involved and therefore could contribute to the BAQS-Met field study by providing an overview of the spatial and temporal variation of NO<sub>2</sub> concentrations. However, I'm missing the novel application which is promised in the title. The abstract doesn't mention a novel approach. In the introduction it is mentioned that "The objectives of the work in this paper are thus to develop an approach to use OMI tropospheric column data to estimate spatially resolved surface NO<sub>2</sub> concentrations at a regional-to-local scale" and to do this conversion "using publicly available data from a network of permanent surface monitoring stations in place of the CTM". That would be a novel approach, but it is not clear to me how this works. Eq. 1 uses ratios calculated from GEOS-Chem output, and then you write that you "obtain surface concentrations without the use of a CTM" by determining the average ratio of column to surface concentration (VCD to Surface) for each OMI overpass over the region using the high-time resolution surface monitors." Averaging over the whole region to determine the conversion factor would work if the profile shape is constant over the region, but as you mention later in the manuscript the surface concentrations vary significantly, so most likely profile shapes vary too. Furthermore, I don't see the value in comparing the OMI surface concentrations to ground measurements results (Sec. 3.1.1) if you use other ground measurements for the conversion, you might as well compare the different ground measurements directly. If this conversion is the focus of the paper, it should be described and analyzed in more detail, e.g. by comparing to other conversion methods, like using a CTM, and evaluating advantages and disadvantages. If this conversion is more a tool to filter out certain pattern in the data set and the focus of the paper is more the spatial and temporal analysis of different measurement data sets, then the title and the objectives should be changed accordingly.

*We agree that some sections are confusing. A number of paragraphs have been re-ordered or removed. To help point out the novelty of the work, we have added the*

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following paragraph to the introduction:

*To the best of our knowledge, this method for combining in situ data from permanent monitoring stations with high-resolution satellite data to obtain high-resolution ( $\sim 11$  km) estimates of long-term average surface NO<sub>2</sub> concentration maps is novel. In addition, the region covered and the 15 km spacing of the campaign-only passive monitors allowed for an unprecedented surface NO<sub>2</sub> dataset against which to compare the maps generated using this novel method. Finally, the use of integrated passive samplers provided a baseline which would be more representative of conditions which would be meaningful in a human-exposure context and therefore made this study more relevant to the health research community.*

*While we recognize that averaging over the region will not capture all the variation present in the shape profiles, the value in using these averaged surface-to-column ratios is apparent in the improvement in correlation between the OMI measurements and the intensive campaign surface passive monitoring network. Comparison with CTM-inferred surface concentrations was beyond the scope of this work, but we agree that more attention should have been paid to the method and have changed the manuscript to reflect this.*

*This is not simply a matter of comparing surface measurements to surface measurements. The permanent monitoring network had lower spatial resolution than the passive samplers, in that the campaign-only passive samplers, which were not used to develop the surface-to-column ratios, were spaced much closer to each other than the permanent monitoring instruments. Even single-overpass OMI data had higher resolution than the permanent monitoring network. Thus, combining multiple OMI overpasses with the permanent monitoring data allowed inference of high-spatial resolution ground based concentrations, at the level of the passive monitors, based on OMI data. Direct inference based on the permanent monitoring data alone would not have been possi-*

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ble. To help clarify this particular point, we have changed the statement of objectives in the introduction:

*In situ NO<sub>2</sub> data are publicly available. However, in situ monitors are sparse, point measurements. It is therefore difficult to provide an accurate representation of NO<sub>2</sub> concentrations in between monitoring stations. The objectives of the work in this paper are thus to develop an approach to use OMI tropospheric column data to estimate spatially resolved surface NO<sub>2</sub> concentrations at a regional-to-local scale and then to provide examples of the insights that can be gained regarding spatial and temporal patterns of surface NO<sub>2</sub> in the study region.*

*In addition to this, we have clarified section 2.4 to separate the original GEOS-Chem-based formulation for inferring surface NO<sub>2</sub> from OMI from our new, somewhat similar method. Equation 1 now reflects the GEOS-Chem-based procedure and a second equation was added to explain the new method presented in this paper:*

*Whereas in situ measurements are true surface measurements, OMI tropospheric NO<sub>2</sub> vertical column density measurements include both the surface-level NO<sub>2</sub> and its vertical distribution through the tropospheric column. This distribution depends on the chemical lifetime of NO<sub>x</sub>, the partitioning of NO<sub>x</sub> into NO and NO<sub>2</sub>, layering of the atmosphere and the dispersion of NO<sub>x</sub> during vertical mixing. Lamsal et al. (2008) inferred surface-level NO<sub>2</sub> concentrations (S) from OMI tropospheric vertical column densities (Ω) by applying the ratio of surface-level NO<sub>2</sub> concentrations (S<sub>G</sub>) to vertical column densities (Ω<sub>G</sub>) calculated using the GEOS-Chem global CTM:*

$$S = \frac{S_G}{\Omega_G} \times \Omega \quad (1)$$

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*In order to obtain surface concentrations without the use of a CTM, we determined the average ratio of in situ surface concentration ( $\bar{S}$ ) to OMI columns ( $\bar{\Omega}$ ), coincident with surface monitoring stations, over the region (equation 2),*

$$S = \frac{\bar{S}}{(\bar{\Omega} - \Omega_{BG})} \times (\Omega - \Omega_{BG}) \quad (2)$$

*where  $\Omega_{BG}$  is the background OMI  $\text{NO}_2$  column as described in the subsequent paragraph.*

Several sections are incoherent in style and information content with a confusing repetition of descriptions and details, e.g. for special time periods or different measurements of the campaign. In some cases, it is hard to track whether certain details are actually relevant and used in the study.

*In addition to the above listed changes a number of significant changes have been made throughout the paper to improve flow and clarity and reduce repetition. We have tried to de-emphasize details which were provided mainly in the name of scientific completeness, and, as necessary, refer back to the stated objectives throughout the paper to ensure continuity of thought.*

*We would also like to point out that the 2nd reviewer felt the paper was well-written. We have, therefore, attempted to find a balance between altering the manuscript enough to ensure that it is coherent, while still respecting the other reviewer's opinion.*

The introduction of this extensive and elaborate campaign is understandably lengthy but would benefit to great extent from either postponing details to subsequent sections or introducing overview tables (and maps) for the entire campaign and periods of special interest. This holds also for other parts of the paper.

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*We hope that the description of the campaign details has been improved by the additional context provided by the careful changes we have described above. We were wary of changing this section for a number of reasons. Most of the detail was provided in the interest of ensuring the paper was a complete representation of the methods used and the reasoning behind these methods. Because this paper is intended to be part of a special issue of ACP, readers with a broader interest in the campaign can get broader campaign information from the overview paper or better details from other papers in the issue. As before, we wished to respect the other reviewer's opinion that the paper was generally well-written.*

**Detailed comments:** It's not clear to me why the "use of in situ data to determine the surface-to-column NO<sub>2</sub> relationship makes this analysis insensitive to potential bias in the OMI NO<sub>2</sub> data." You take out the bias by dividing the VCDs by the average VCDs (Eq. 1), but that has nothing to do with using in situ data. Please explain.

*Please see below for our response to the next point.*

Page 17250, line 16: "This approach has the advantage of simplicity since it is not dependent upon CTM runs, which are relatively costly and not available to all OMI data users. -> how about "time consuming" instead of "costly"? CTM runs are not expensive compared to buying and operating measurement instruments, assuming that you don't have to buy a computer cluster to run them. I believe the argument in the following sentence, the elimination of a potential model bias is the more important advantage and could be emphasized more (e.g. take out the word "potential" in this sentence). Why is your method "less sensitive to the OMI data product" and why is that an advantage?

*These comments are well taken. The second clause in the first sentence you mention here has been removed: "This approach has the advantage of simplicity since it is not dependent upon CTM runs." We have added a reference to the methodology section where we have added the following paragraph:*

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*From equation 2, it can be seen that the use of in situ data to determine the surface-to-column NO<sub>2</sub> relationship makes this analysis insensitive to potential bias in the OMI NO<sub>2</sub> data. Because OMI column densities appear in both the numerator and the denominator, any bias present in the particular retrieval used would be expected to cancel out. As an example, it has been suggested that the SP is biased low in the winter (Lamsal et al., 2008). If the surface-to-column ratio is calculated using the (assumed, in this case, unbiased) modeled surface concentration and modeled column density, the inferred surface values from these low OMI columns will also be biased low. However, by calculating the ratio using the observed OMI columns, assuming the in-situ measured surface concentration is the same as the modeled surface concentration, the surface-to-column ratio will be greater than the modeled surface-to-column ratio which will increase the final inferred surface concentrations.*

Page 17251, line 9: refer to figure 8.

*This sentence has been removed.*

Page 17252, line 11: the section title is High-time resolution measurement by chemiluminescence, but then you reduce it to 1h averages. Why is that?

*The high-time resolution is in comparison with the 2-week integrated averages. The sentence now reads: “Measurements were averaged to 1 hour to match the data available from the permanent MoE network.” In addition to this, the result section previously titled “High-time resolution measurments” has been changed to “Spatial and temporal patterns inferred from chemiluminescence monitor data” to further reduce confusion.*

Page 17252, line 14: Considering the details provided in the preceding paragraph, the location of the Environment Canada CL monitors should be mentioned.

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*We have added “One of these monitors was located at Bear Creek, collocated with a MoO-CL monitor; one was located at Harrow; and one was onboard the Environment Canada mobile platform, CRUISER.”*

Page 17253, line 19: “the first 6 periods...”-> Specify periods.

*The clause now reads “the first 6 periods (May 31 to August 22) had 17 simultaneous sampling sites and the final period (August 23 to September 6) had 13 sampling sites.”*

Page 17253, line 28: You mention “additional quality assurance checks”, what are those?

*The additional checks involved examining all of the ion chromatography runs related to samples which differed by more than 20% for problems, re-analyzing suspect samples, re-integrating peaks and rejecting the outliers and then producing the best result possible for that two week period and that site. The sentence now reads: “Collocated filters were checked for consistency and if values differed by more than 10% the two measured concentrations were subjected to additional quality assurance checks including examination of the chromatograms and possible reanalysis to improve the IC results and, if acceptable after these changes (within 20%), the two values were averaged. However, if one of the two was still found to be an outlier or contaminated then it was excluded from the averaging.*

Section 2.4: Aura is crossing the equator in south-north direction, not north-south. "OMI was designed to provide daily global coverage", instead of “...intended to provide...”

*Fixed.*

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Page 17254, line 16 “removing as many as 18 pixels” instead of “removing as many at 18 pixels”

*Fixed.*

Page 17254, line 19 “provided by NASA” instead of “provided NASA”

*Fixed.*

Page 17254, line 21: You might want to refer to the web pages of the two data products instead of Lamsal et al. 2010.

*URLs added.*

In the following sentences: The first step of the retrieval algorithm is clearly a DOAS retrieval, but writing that the data product begins by using the DOAS algorithm sounds a little weird. The 365-500 nm you are referring to is the spectral range of the VIS channel, but for the NO<sub>2</sub> DOAS retrieval the wavelength range from 405.0 nm to 465.0 nm is used (see Bucsela et al. 2006). Aerosol particles are not considered for the calculation of the AMFs. If this feature had been added (e.g. by calculating your own AMFs) please provide a reference. It sounds a little misleading to write that the AMF “corrects” for the viewing geometry and light-scattering influences. The AMF concept converts the slant column densities (SCDs) which are the direct result of the DOAS retrieval and depend on the viewing geometry and influenced by scattering, into vertical column densities (VCDs) which are (supposedly) independent of those influences. The CDs are not incorrect, they just represent a quantity difficult to interpret and compare to other data sets. Therefore, I would rather call the AMF concept a conversion than a correction.

*Agreed. The wavelength range has been changed to 405 to 465 nm and the AMF section now reads “The air mass factor (AMF) accounts for viewing geometry and light-*

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*scattering interferences such as clouds; this AMF is applied to convert the measured slant columns into tropospheric vertical column densities. It is these conversions that introduce the most uncertainty in the reported vertical columns over polluted areas (Boersma et al., 2007, 2004; Martin et al., 2002)."*

For the weights of the gridding process you might want to consider adding the measurement error or a term accounting for the cloud influence (see for example Wenig et al 2008, "Validation of OMI tropospheric NO<sub>2</sub> column densities using directSun mode Brewer measurements at NASA Goddard Space Flight Center") in order to further reduce the errors. This reference can also be used to justify using the inverse of the area of the pixel (in contrast to for example  $1/\text{area}^2$  or something like that, which would give near-nadir pixels an even higher weight).

*We have added the sentence "Although this weighting scheme has been used with a squared-uncertainty term (Wenig et al., 2008), it was found that inverse-area-alone weighted values used here differed from inverse-area, inverse-squared-uncertainty weighted values by less than 3% for the 2-week averages over the campaign period. "*

Page 17256, line 15: Please specify "below" for easier reference.

*Now reads "The error in this assumption is discussed in section 3.3.1. "*

Sec 3.1.1: Why did you calculate the correlation coefficients between the different stations? It would make reading this section easier, if key values for NO<sub>2</sub> concentrations and correlation coefficients would be given in the text.

*These correlations are important for helping to identify potential sources of pollution affecting different areas in the campaign region, which provides some insight regarding the level of spatial heterogeneity that the satellite derived observations will need*

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to resolve. This section, which has been modified to address these questions and suggestions, now reads:

*NO<sub>2</sub> concentrations were found to vary both spatially and temporally. The hourly and daily concentrations of NO<sub>2</sub> showed moderate to weak correlations between the sites over the duration of the campaign for both hourly and daily average measurements (Table 2). This comparison is important to help identify potential sources of pollution affecting different areas in the campaign region, which spans urban, rural and industrial areas. Ridgetown showed a relatively high correlation with the urban sites (~ 0.7), which was surprising because Ridgetown is over 100 km from the urban and industrial centres of Windsor. However, the Ridgetown measurement site was 5 km from highway 401, a major provincial highway in the region leading directly to the Ambassador Bridge border crossing in Windsor. This relatively strong correlation despite the spatial separation implied that 401 traffic was an important source at both sites.*

*NO<sub>2</sub> concentrations varied diurnally and day-to-day at both the urban (Windsor) and rural (Harrow, Bear Creek) sites (Figure 3). Peak concentrations for Windsor occurred between 6 and 10AM (local standard time) on most days, while these morning rush-hour peaks were sometimes delayed at the rural sites downwind. Variation between sites was greater than week-to-week variations in same-site NO<sub>2</sub> concentrations (Table 3). The largest between-site ratio in the median weekly concentration was over 4 (Windsor Downtown had a weekly median concentration of 13.6 ppb while Peelee had a weekly mean of only 2.9 ppb) whereas the largest ratio between maximum and minimum weekly concentrations at a single site was less than 2 (Peelee had the highest ratio with a maximum weekly concentration of 3.1 ppb and a minimum of 1.7 ppb). In contrast, daily averages varied about as much at a given site as they did between sites. This in-*

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*licated that spatial distributions remain fairly stable over the region, even though local events may briefly increase the spatial heterogeneity. This result is important for high spatial resolution interpretation of satellite remote sensing data because these methods take advantage of differences in the daily footprint of the instrument overpass to extract long-term patterns in spatial air pollution distributions.*

Page 17257, line 19: “When averaged over a longer period of time, such as a week, NO<sub>2</sub> concentrations varied much more spatially than temporally“ -> Isn't that obvious? Averaging (or smoothing) reduces variability, so please explain why this is worth mentioning.

*It is true that this section generally highlights observations of what would be expected, but we have chosen to leave it in the paper because it provides readers, who may not be familiar with this region of the globe, with some insight regarding the general conditions related to NO<sub>2</sub> and highlights the features that we are aiming to use the satellite observations to better understand. In particular, the intended point here was to compare the temporal and spatial variability, specifically to show that at the level of temporal averaging of interest here, the spatial variability predominated. This sentence was replaced with: “Variation between sites was greater than week-to-week variations in same-site NO<sub>2</sub> concentrations (Table 3).”*

Page 17260 , line17: What are the units of the numbers given in the brackets?

*Ppb added: “... mean NO<sub>z</sub> between noon and 1400 EDT were found to be low (2.62 ppb and 3.22 ppb for rural locations and 2.62 ppb for urban) ...”*

Figure 8: The term “false color” is typically used when showing an image in colors that differs from a color photograph, so I think you can use just “color” instead of “false color”.

*Fixed.*

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 17245, 2011.

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