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Interactive comment on "Effects of daily meteorology on the interpretation of space-based remote sensing of NO_2" *by* Joshua L. Laughner et al.

Anonymous Referee #3

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The study by Laughner et al. addresses the relevant issue of highly resolved a priori profiles for NO2 retrievals from OMI. The issue –a priori NO2 profile information is required at the spatial resolution of an OMI pixel (∼300 km2) for the actual day *and* time of the measurement– has been known for a long time (e.g. Boersma et al., 2007; Heckel et al., 2011). The merit of this study is that the authors quantify the effect of diurnal and highly resolved a priori NO2 profiles on the air mass factor calculations, relative to AMFs using either coarse profiles or monthly mean profiles. As expected, considerable differences are found in the retrieved NO2 columns, and these differences can be interpreted as systematic error contributions in operational retrievals making use of coarse spatial resolution or monthly mean averaged NO2 profiles in their AMFs.

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Discussion paper

Moreover, the authors show that this undersampling problem has serious consequences for the recently developed technique to simultaneously estimate NOx emissions and NO2 lifetime for isolated pollution sources from wind-sorted satellite measurements of tropospheric NO2 columns (Beirle et al., 2011). The authors address those consequences by comparing the effect of using NO2 retrievals with different profiles (high vs. low spatio-temporal resolution) on the final estimates of NOx emissions and lifetimes. In doing so, the authors use an interesting approach: that of a 'pseudo-retrieval'. It allows to more or less investigate the effect of profile changes on the retrievals without the usual perturbing influences from other retrieval parameters such as variable viewing geometries, clouds, albedo, etc. They find much higher NOx emissions from satellite retrievals using highly-resolved a priori profiles, and this is an intriguing result in view of previously published city emission estimates using retrievals with coarser a priori profiles, as the differences are easily a factor of 2.

Major issues:

* One limitation is the focus on only one area (Atlanta, GA) and one season (Summer). As pointed out by another reviewer, focusing on such a short period leads to a limited dataset, from which it is difficult to obtain estimates of emissions and lifetimes that are significantly different. Since the differences in the estimated emissions are at least a factor of 2, it should be feasible to achieve statistically significant differences by analysing a longer period. This would strengthen the paper considerably.

* Then there is a serious error in the theoretical framework for BEHR AMFs. According to Eq. (2) and the text in section 2.2, the cloudy AMF is calculated only between the cloud pressure level and the tropopause. In principle this can be done, but then the retrieval needs to account for a so-called "ghost column" [Burrows et al., 1999]. A ghost column correction however, is not being applied here. The better alternative is to calculate both the clear-sky and cloudy-sky AMFs by integrating the NO2 profile from the surface pressure to the tropopause. This formulation ensures that the AMF value returns a tropospheric column that is representative for all NO2 in the troposphere,

and not just for the above-cloud fraction (in the limiting case of cloud fraction 1.0). The authors should revisit their integration limits for Eq. (2), probably also for their current BEHR-product.

This error is also the reason for the incorrect statement on page 7, under point 3: "... a cloudy pixel will have a much greater AMF than a clear one". The statement would be correct only if the word 'AMF' would be replaced by 'above-cloud AMF'. And, in line with the above criticism, an above-cloud AMF can only return an above-cloud NO2 columns, which cannot be the purpose of a state-of-science retrieval.

* There are difficulties with the interpretation of uncertainties in the tropospheric column retrieval. On page 10, the choice for 1.0x10ˆ15 molec. cmˆ-2 as a typical number for the uncertainty in the tropospheric NO2 column is rather arbitrary. Detailed error analyses (e.g. Boersma et al. [2004]) have pointed out that the uncertainty in the tropospheric column is highly variable because of AMF (a multiplicative factor indeed) uncertainties, which depend strongly on variable clouds, surface albedo, and NO2 profile shapes. Moreover, the estimate used by the authors is rather optimistic. More realistic uncertainties are on the order of 1.0x10ˆ15 molec. cmˆ-2 +25% (of the individual column value).

Also, part of the AMF (25%) uncertainty is random in nature, and part is systematic. So by averaging over a large number of pixels, as is done on page 11, only the random part of the AMF-related uncertainty reduces, but not the systematic part. This makes the estimate of the 'nominal uncertainty' of ∼1.6x10ˆ14 molec.cmˆ-2 too optimistic. The given value may hold for the SCD-related uncertainty, but is not representative for the uncertainty in averaged tropospheric NO2 columns, where systematic (e.g. albedo-related or cloud-related) errors are likely still of concern.

Specific issues:

Abstract, line 6: this paper does not address variations of NO2 in power plant plumes, so this should be removed. The paper is about NO2 variations in urban plumes.

Introduction, lines 18 and 30: a reference to the work by Vinken et al., ACP, 2014 on ship emissions estimates using improved-resolution a priori profiles would be appropriate here.

P3, lines 7-8: earlier studies by McLinden et al. [2012] showed that the oil sand signals was detectable also for retrievals using coarse-gridded a priori profiles. The statement should be nuanced in that the spatial signatures are more realistically resolved with higher resolution a priori profiles.

P3, L13: also include here a reference to the DOMINO retrievals using 0.5 x 0.67 profiles from GEOS-Chem over Europe from Vinken et al. [2014]. These retrievals also use diurnal profiles.

P3, L17: the NASA retrieval is usually indicated as the Standard Product v2 or SP v2. OMNO2 (actually OMNO2A v1) refers to the software for DOAS spectral fitting.

P4, L30: 13 x 24 km2

P4, L32: 'theoretical' daily global coverage is a strange term. Coverage was nearly global every day until the row anomaly, and after that, coverage is more or less global every 2 days.

P5, L6: please provide the name and appropriate version of the NO2 SCD product used. I think it is OMNO2A v1. This product has recently been evaluated in Marchenko et al. [2015] and van Geffen et al. [2015]. It would be appropriate to cite those papers here.

P5, L17: apart from albedo, please also provide details on the cloud information (effective cloud fraction, pressure) used in the BEHR-approach.

P5, L27: which version of the WRF-Chem model is used?

P6, section 2.3: please provide some more details on the WRF-Chem model such as what is the NOx emission total over the US in the period of interest, and on the meteoro-

logical and chemical boundary conditions used. It is unclear how realistic WRF-Chem simulations of NO2 profiles are. Have these been validated against surface and aircraft measurements of NO2?

P7, L19: please clarify what the vertical range is of the 'first five layers' of WRF-Chem.

P8, L6: is it really necessary or relevant to refer to a Matlab function, when explaining how you do the fit? If it is, please provide some more information on why you chose this particular fit approach over other alternatives. As a non-Matlab user, the sentence does not mean much to me. Later on the same for 'fmincon'.

P8, 12-17: this part is very technical and should be moved to an appendix or supplement.

P10, L13: that lightning is not included in this WRF-Chem set-up should not be mentioned only here, but already in the model description section 2.3.

P12, L18: sigma_x represents the width of the Gaussian plume, but also the spatial smearing of the signal caused by the satellite pixel extent, and the fact that cities are covered by different satellite footprints from day to day.

P13, L14-19: this whole section presumably discusses Table 4, but that is not obvious from the text. I'm confused by the statement that the "choice of a priori leads to statistically different emissions for all five cases", whereas Table 4 shows emission values that all overlap within the quoted uncertainty estimates.

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