Reply to Referee #3

The study "OMI tropospheric NO2 profiles from cloud slicing: constraints on surface emissions, convective transport and lightning NOx" by M. Belmonte Rivas et al. applies a cloud slicing technique to clouded OMI NO2 observations in order to derive a mean NO2 pseudoprofile. The study is well written and contains comprehensive analysis, which indicate (regional) model shortcomings for emissions, convection, advection, or lightning NOx, which is valuable information for the scientific community.

My main concern is that the study does not at all account for seasonality, while all involved components (NOx emissions (heating, lightning, biomass burning), NOx life- time, convection patterns, NOx profiles, and cloud characteristics) can vary strongly over the year. The value of an annual mean pseudoprofile is thus questionable, as the different cloud pressure levels and the corresponding NO2 columns are not at all equally distributed over the year.

Previous cloud slicing studies have considered seasonality (e.g. Liu et al. for CO and Choi et al. for NO2), and I see no reason why this study does not. Thus, I recommend to perform the cloud slicing on a seasonal basis. If statistics is too low for 3 months, the seasons from several years can be merged. This requires major revisions, but will yield better interpretable pseudoprofile and very likely strengthen the discussion of the model comparison.

May this manuscript serve as a proof-of-concept regarding the amount of information that may be extracted from otherwise discarded cloudy OMI (GOME or SCIAMACHY) data. The authors are already intent on applying the cloud slicing methodology on OMI data on a seasonal (and also interannual) basis. But due to the already excessive length of this manuscript, we would prefer to leave this material for a future paper. The object of this manuscript is to describe our methodology, its shortcomings, and what potential applications it may serve. We are aware that any statements regarding the comparison with the model remain at the level of plausible at this stage, in the hopes that the picture afforded is solid and convincing enough to motivate further studies and perhaps a global validation campaign.

Further comments:

8022/7: Here, OMI "cloud pressure" is introduced and related to the cloud midlevel. Later (Fig. 2, section 3.1), the terms "cloud top pressure" and "cloud top" are used. Please use consistent terms.

Agreed. The term Cloud Top Pressure and its acronym CTP are changed into CLP for Cloud Level Pressure where relevant across the manuscript.

8023/19: VMR is not a concentration.

Agreed. The terms volume mixing ratio and concentration are used interchangeably thorough the manuscript, though here for correctness the term volume mixing ratio will be used.

8023/24: What is the lesson learned from the trial runs? How far are the results depending on the chosen pressure grid? What are the reasons for choosing exactly this grid?

In general terms, profiles do not depend on the chosen pressure grid, though their appearance (particularly their vertical resolution) does. Initially, we started with three strata defined between the surface, 720 hPa, 500 hPa and the tropopause level as being representative of low, mid-level and high level clouds. At a later stage, each strata was further subdivided into two sublevels (distributing the number of samples per strata as uniformly as possible), as we noticed that the resulting amount of samples per strata was sufficiently large to provide new profiles with a smoother and realistic appearance.

8024/16-19: Clarify that VCD_above is the *tropospheric* column above cloud

OK.

8025/19: Units are missing.

The temperature correction T_{corr} is unitless.

8028/1: Before discussing the Pseudoprofile errors, please first introduce the term

Pseudoprofile in a dedicated subsection.

Agreed. The section has been rewritten and reorganized, also on request of Reviewer #2.

8028/10: model true -> model ("true")

Agreed.

8029/4: It is stated that the cloud modifies the profile, but how (and how strong) is not discussed. This aspect should be extended when introducing the Pseudoprofile.

Agreed. The following text is inserted in Section 2.1.3: “There are a number of ways in which the presence of cloud may modify the underlying profile: either directly, via lightning NOx production in the upper levels, or advection of (clean/polluted) air from the boundary layer at the lower levels, or more indirectly via suppression of biomass burning at the surface or decreased photolysis under the cloud. One can appreciate that the effect of cloud presence on the profile varies with cloud altitude, which is unfortunate, because we use changes in cloud altitude to sample the underlying profile. This state of affairs introduces a source of systematic error between the cloud-slicing estimate of trace gas concentration (i.e. the pseudoprofile) and the actual underlying profile, which we term pseudo-profile error.”

8030/18: Why is this comparison not shown? This figure might be provided as supplement.

Agreed. The annual mean NO2 VCDs above cloud from the TM4 model (the model counterpart to Figure 3) are provided as a supplement. See also the Appendix to this reply.

8036/25:
we have drawn ... classes defined according ...
-> we have defined ... classes according ...

Agreed.

8043/19: actualize -> update; please provide reference(s).

Agreed. Two new references have been introduced:

Mijling, B., and R. J. van der A (2012), Using daily satellite observations to estimate emissions of short-lived air pollutants on a mesoscopic scale, J. Geophys. Res., 117, D17302, doi:10.1029/2012JD017817.

Ding, J., R.J. van der A, B. Mijling, P.F. Levelt, and N. Hao (2015). NOx emission estimates during the 2014 Youth Olympic Games in Nanjing. Atmos. Chem. Phys. Discuss., 15, 6337-6372, doi:10.5194/acpd-15-6337-2015

Fig. 5: Are there also negative VMR (over ocean)? If so, please mention & shortly discuss them.

Yes, there are some instances of negative VMRs but mainly related to column differences between poorly populated cells (i.e. at high latitudes, near the tropics at low altitudes, or around the subsidence regions). These instances are identified and dealt with by recourse to information from nearby cells, when available, or otherwise ignored A brief mention is inserted in Section 2.1.3 “Error analysis”.

Appendix – Annual mean NO2 VCDs above cloud from OMI and the TM4 model

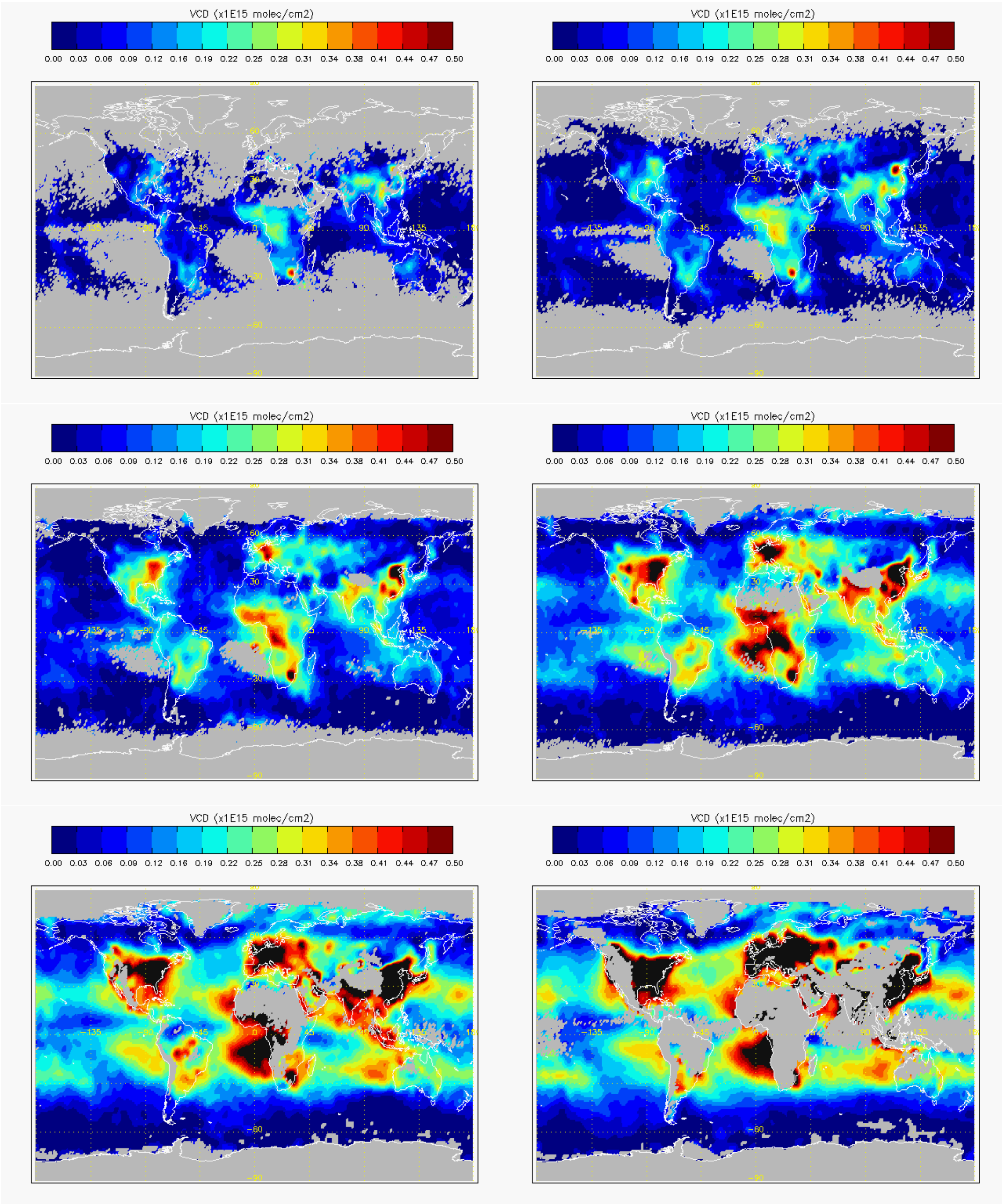

Figure R1 – OMI NO2 VCDs above cloud - average quantities for the year 2006: for high altitude clouds (top row, 330 and 450 hPa), mid altitude clouds (middle row, 570 and 670 hPa) and low clouds (bottom row, 770 and 870 hPa).

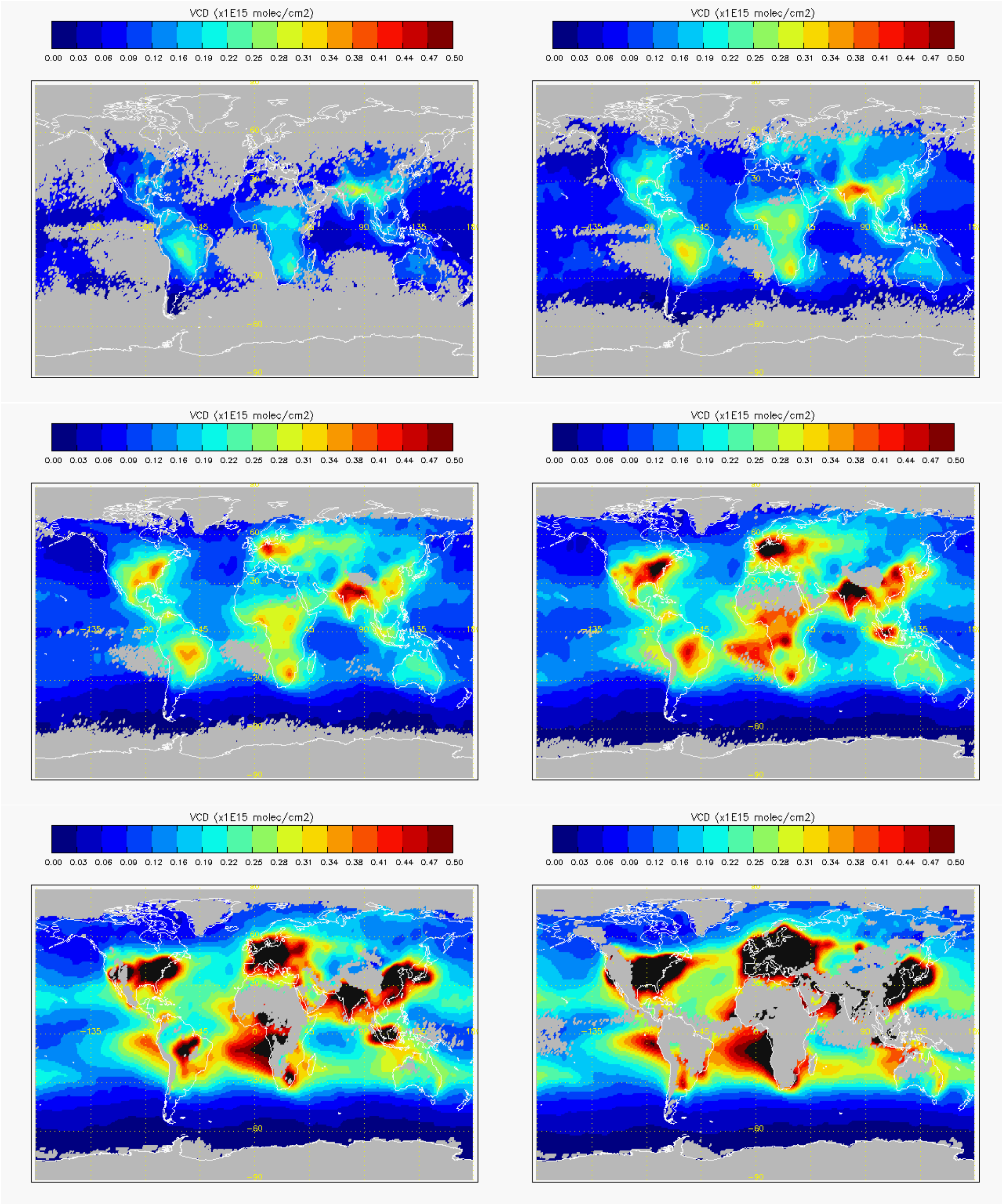

Figure R2 – TM4 model NO2 VCDs above cloud - average quantities for the year 2006: for high altitude clouds (top row, 330 and 450 hPa), mid altitude clouds (middle row, 570 and 670 hPa) and low clouds (bottom row, 770 and 870 hPa).