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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Satellite NO_2 observations and model simulations of tropospheric columns over South-eastern Europe" *by* I. Zyrichidou et al.

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REPLY TO THE COMMENTS OF REFEREE#1

We would like to thank referee #1 for the comments and suggestions that contribute to the improvement of our manuscript. In the following, the referee's comments are shown first and are followed by the author's responses.

Response to general comments

In the revised version we show all geolocations selected in the area and when available we also use surface in situ observations of NO2, representative of mainly local surface

emissions, to compare with satellite estimates. An appropriate figure and discussion has been inserted in the revised manuscript. We also discuss in more detail the expected impact of lightning and biomass burning on the tropospheric NO2 loading. As mentioned also later in our response to referee #1 comments the model run used in this study was performed in the frame of CECILIA EU project and at the moment the model is not yet optimized to include operationally these two mechanisms in the simulations. This is an ongoing effort. However in the discussion we provide literature evidence for their expected impact.

Abstract, Line 22: Typo, should be -0.1% not -0.1.

The value -0.1 is referred to the mean bias whose mathematical expression is MB= 1/N*(Σi(Pi-Oi)) and is not reported as %. (Pi: the prediction at time and location i, Oi: the observation at time and location i and N: the total i). We have added in the text the units, in 1015 molecules/cm2, that were omitted.

Page 12173, Line 5: A more recent reference in addition to Crutzen, 1979 would be beneficial.

We further cited Murphy et al, 1993 and Finlayson-Pitts and Pitts, 2000.

Page 12173, Line 9-10: Please insert references regarding the OH radical as the cleansing agent of the troposphere and the toxic effects of NO2 and O3.

We added a reference regarding the OH radical of van Noije et al., 2006.

Page 12174, Line 4: Typo. It is CHartographY.

CartograpHY was corrected to CHartographY.

Page 12175, Line 8: Style. Shouldn't this be 'polluted air masses. . .. .Black Sea, leads to an increase in NO2'.

In the corresponding paper of Ladstätter-Weißenmayer et al., 2007 they are discussed as "air masses", but from the context it is easily concluded that the authors are referring to “polluted air masses”. The corresponding addition was made in the text.

Page 12175, Line 26: Please explain why the spatial distribution is not ‘proper’. Are there any stations at all in this region?

The ground-based stations that exist in the region are managed by the various local authorities, such as municipalities, prefectures, and so on. As a result, such stations exist either only in the capitals of states and in largely populated cities, or near airports. As an example, according to the European Air Quality data base (Airbase) (http://www.eea.europa.eu/) for Greece only 24 ground stations (16 urban, 7 suburban and 1 rural) provide NO2 measurements, most of which (14) located in the Greater Athens area and 4 in the Greater Thessaloniki (the second biggest Greek city). The same pattern is found for the rest of the Balkan states considered and therefore from the air-quality/environmental ground-based network one cannot deduce the NO2 variability over the Balkan Peninsula. An appropriate discussion has been inserted in the revised manuscript.

Page 12176, Line 18: What are the criteria for determining an ‘unpolluted city’?

We agree with the comment. The word should have been “sites” and not “cities” and was corrected. An unpolluted site is a site with no significant known local sources and located far from industrial and urban sources.

Page 12177, Line 5 and thereafter: Capitalization. It should be Equator not ‘equator’.

Corrected as requested.

Page 12179, Line 24: What do you mean by un-flagged? Please be clearer.

According to the TEMIS HDF data file user manual for GOME, GOME2 and SCIAMACHY, the flag component in the hdf files indicates if the tropospheric retrieval is meaningful (yes= 0, no= -1). So for these instruments we kept only the measurements with flag =0. As for OMI HE5 files we kept only the measurements with even flag values that correspond to meaningful tropospheric retrievals. A comment to this effect

was added in the text.

Page 12179, Line 31: Why were only cross-track pixels 10 to 50 used in the OMI retrieval?

The Intrinsic Field-Of-View (IFOV) is defined as the FWHM of the pixel response curve obtained when moving a point source in swath and/or flight direction. The IFOV of a sub-satellite OMI CCD-pixel is about 3 km in the swath direction and about 10 km in the flight direction. In the swath (across track) direction, 4 or 8 pixels are binned to a ground pixel of 12 or 24 km. Futhermore, OMI measurements are co-added during 2 seconds. With a ground speed of the Aura spacecraft of about 7 km/s, this results in ground pixel size of 13 km in the flight (along track) direction. The pixel-size in the swath-direction increases from 13 $\times$ 24 km^2 (exact nadir position) to 13 $\times$ 128 km^2 at the most outer swath-angle (57 $^\circ$). So, in order to make sure that the pixels used in this work are on the near-nadir position and hence the best horizontal resolution is utilized, we have kept cross-track pixels numbers 10 to 50. An appropriate explanation has been added in the revised paper.

Section 2.3: I found the description of CAMx model somewhat confusing. For instance, the run is based on a ‘coarse grid spacing’ which has a spatial resolution of 50x50 km2 (i.e., of order 0.5x0.5 degrees which is still quite fine for chemistry-transport model). Also I think the lack of lightning and biomass burning NOx emissions within the CAMx model is a significant shortfall. The authors should at least provide some information on the typical periods and intensity of biomass burning and also on the lightning statistics over this region.

We concur with referee 1 on this comment. In the first place, the model spatial resolution of 50x50 km2 is quite fine indeed. So, we corrected the particular point in the text and we added some useful information about the CAMx model in order to provide a more elaborate description. As for the biomass burning statistics over the Balkan region, we discuss in the text that according to Figure 4 (right panel), derived from the World Fire Atlas (http://wfaa-dat.esrin.esa.int/), the typical period of large biomass burning events, which over the Balkan Peninsula are mostly determined by fires, is the warm season (from June to September). Moreover, we added a reference to Ellicott et al., 2009, in which a statistical estimation about the diurnal variability of biomass burning, mostly over Africa and South America where biomass burning is most frequent, is provided. It shows that fire radiative energy (FRE), whose rate is proportional to the biomass consumed, records maximum values during midday, just like the diurnal pattern we show for CAMx emissions in Figure 4 (left panel) . We agree that the lack of lightning and biomass burning emissions within the CAMx model is a shortfall. However, there is a published report in the frame of NATAIR EU research project (2007) (http://natair.ier.uni-stuttgart.de) which reports that NO emissions from lightning for Europe contributes 0.3% to the total emissions from other sources (Simpson et al., 1999). This proportion is quite small in order to consider lightning as important NO2 source for the area and to take it into account for the explanation of the discrepancies between the satellite measurements and the model predictions. An appropriate paragraph was inserted in the revised manuscript.

Section 3.1: I think a discussion of the variability over 4 locations, albeit that are representative of different conditions, is simply not enough. The main focus of the paper after all is the variability of NOx over this region. For example, showing the time series plots at all locations would make it easier for a reader to gain a better understanding of the NO2 variability.

Figure 2 and corresponding discussion has been revised following the reviewer's suggestion, including all geolocations.

Page 12185, Line 23: Typo. Correct the spacing between: '13:30 to 10:00 UT'. This also occurs in a couple of other places. Please check the rest of the manuscript carefully.

Spacings were checked and corrected.

Section 3.3: If the spatial resolutions of GOME and CAMx are not really comparable, why wasn't the comparison performed with just SCIAMACHY or OMI measurements for the just the TEMIS retrieval? I think adding a brief comparison of the model to these instruments would be beneficial to the paper.

We concur with the referee on the comment that a comparison of the model to the SCIAMACHY, OMI and GOME2 instruments would be beneficial as well. CAMx run from years 1996 to 2001 were utilised as those model run had already been performed by some of the co-authors as part of a different project (CECILIA) and were available in the time frame of this paper (see also our response to general comments). This limited us to use the GOME instrument for the comparison.

Page 12187, Line 24: Typo. I think there is a missing x10ˆ15 at the beginning of the line.

We added the missing x10ˆ15.

Page12188, Line 3-5: Please define the normalized mean bias. Also there is significant difference between the TEMIS and Bremen retrievals over industrial areas (-4% versus 30%). I think this warrants more explanation.

The normalized mean bias is defined as: NMB=Σi(Pi-Oi)/ΣiOi and is reported as %. (Pi: the prediction at time and location i, Oi: the observation at time and location i and N: the total i). We agree with the referee 1 that the difference between TEMIS and Bremen retrievals over industrial areas need more discussion. This difference is probably due to the fact that although the consistency between the two algorithms is satisfactory over the Balkan region (see Fig.6) with a correlation coefficient R=0.81, the mean NO2 GOMEbremen retrievals over industrial areas are lower than CAMx (CAMx: 2.72$\pm$1.22 x 1015 molecules/cm2 vs GOMEbremen: 2.09$\pm$1.40 x 1015 molecules/cm2), whereas GOMEtemis retrievals are a little bit higher than CAMx ones (CAMx: 2.72$\pm$1.26 x 1015 molecules/cm2 vs GOMEtemis: 2.84$\pm$1.62 x 1015 molecules/cm2). Considering however the large scatter these differences are hardly statistically significant. Moreover in addition to possible algorithm issues that require further investigation, these differences can also be partly attributed to the different sampling, since the two algorithms do not always provide simultaneous estimates for the same pixel. An appropriate paragraph has been added in the discussion.

Page 12188, Line 15: Typo. Insert space between '(left). Each. . .'

Space was inserted.

Page 12188, Line 25-27: The authors state that the EMEP emissions over Istanbul were unrealistically low and subsequently that this location was excluded from Fig 7. I think this is where the real science of the paper is. Surely, the authors should be using the satellite observations to constrain the model emissions. At the very least they should provide a better estimate over Istanbul by perturbing the emissions (in a brute-force approach) to obtain the best match between the model and satellite NO2 columns.

Model runs for the whole Europe of various models including CAMx performed in the frame of the GEMS EU project for the year 2008, using a finer resolution for the surface emissions prepared by TNO (Visschedijk et al., 2007) and based on updated information compared to EMEP inventory, show much better agreement (within 30%) with satellite data (OMI) during winter but still most models underestimate significantly the satellite observations that correspond to the area (Huijnen et al, manuscript in preparation), which is also attributed to the summer a priori profiles used in OMI retrievals and is related to the free troposphere versus boundary layer sensitivity of tropospheric NO2. An appropriate discussion has been included in the revised text.

Figure 7: 31 is not the number of 'observations' it is number of locations. Also I think this figure would be better if 'raw' concurrent measurements were plotting instead of the daily average over 1996-2001 (since the averaging will smooth out the variability and therefore the true performance of the model). Nobs definition in Figure 7 was corrected. Nobs is indeed the number of geolocations. We agree that this averaging

smoothes out the temporal variability. However, we feel that the true performance of the model on the temporal scale has already been discussed in Figures 4 and 5 and analysis thereof. In Figure 7 we wish to depict the spatial distribution between predicted and observed tropospheric NO2 columns as a tool to verify the spatial distribution of the emissions used in the model simulations. As one can imagine, a figure containing 6 years of data for all 31 geolocations with 'raw' concurrent measurements will make the plot very busy and hence extremely difficult to decipher.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 12171, 2009.