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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling of photolysis rates over Europe: impact on chemical gaseous species and aerosols" *by* E. Real and K. Sartelet

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Answer to Reviewer 1:

We would like to thank both reviewers for their useful remarks which really help to improve the manuscript. In the following text, we wrote the questions of the reviewer and our associated answer.

General comments: This manuscript compares different models to simulate gas and aerosol concentrations over Europe. The authors first assess the impact of a new pho- tolysis scheme which uses updated cross-sections (the main

source of change from previous photolysis schemes). They then assess the effect of changing the way clouds are represented in the model, and then the addition of aerosols to the model (which act as attenuators of solar radiation and so alter photolysis rates and gas / aerosol con- centrations), using Polair3D. Results are presented in terms of monthly averages for July and November, across Europe. After comparing the different model outputs, the results are compared with measurements. The differences between models are generally smaller than the differences between any model and the measurements, and provide the expected outcomes i.e. including aerosols attenuates solar radiation (especially at the ground) and reduces photolysis rates. How this affects gas concentrations depends on the time of day, the existing concentrations and the competing photolysis rates that may influence a given gas.

Note 1 : This is all explained in great detail for all gases and aerosols considered, and all model comparisons. It makes the paper rather longwinded and I suggest the authors try to find more concise ways to present their arguments and results.

The manuscript was shortened in two ways. First, we decided to not present all the results in detail concerning the impact of cloud parametrisation because its impact on pollutant concentrations at the ground is low compared to the impact of aerosols. Section 3.2 was therefore shortened and does not contain sub-sections any more. Second, we also decided to focus on the month of November for section 3.2 (cloud parametrisation impact) and the month of July for section 3.3 (aerosols impact) in the detailed comparison of the impact on photolysis rates and concentrations, because there are the months for which the cloud parametrisation and aerosols have the most impact respectively. However, both months are kept when comparing to measurements and when quantifying the impact on pollutant concentration at the ground. Explanations of some specific features observed in figures of relative differences have

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also been shortened and are more clear now (section 3.3.2 on NO2 concentrations at 3-4 km and section 3.3.3 on HNO_3 ground concentrations). However, we have also included new features (tables and text) following reviewers suggestions. Therefore the paper is finally only reduced by 8 pages.

Note 2 : It would be helpful to have some more detail about the models and measurements (e.g. Uncertainties).

We did compare our results to the uncertainties of the model in 3.4.1 (comparison with Roustan et al, 2010 on model uncertainties). But this paragraph was not clear and hard to understand. For clarity, we decided to move this discussion to a new paragraph (3.4) which we named *"Comparisons of the impact of the modeling of photolysis rates to model uncertainties"*. To make the comparison easier, we provided a table with a quantitative evaluation of uncertainties, and we re-wrote this paragraph.

Specific comments:

Note 3: Abstract - the manuscript does not evaluate the effect of photolysis rates on air quality monitoring. The monitoring is used to assess the models. This work shows that the model with aerosols would reduce the number of predictions of air quality alerts, or prior warnings, it would not affect the actual, measured air quality (which may or may not agree with the prediction).

We do agree that monitoring was not the appropriate word. We replaced it, by modeling or simulation depending on the context.

Note 4 : P16698 The FastJX scheme. How are the 18 wavelength bins defined. For example, is there enough resolution to distinguish between UVA and UVB - ie to separate the effects on J(O_3) and J(NO_2). Please give more detail.

The selection of these reduced wavelength bins have been done by moving the bound-

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aries between the bins until a minimum in the relative root-mean squares errors is achieved for 4 photolysis rates (O_3, NO_2, HNO_3 and H2O2) and under different conditions (clear sky, cloudy) (Wild et al., 2000). The UV part of the spectrum (289-412 nm) is the most refined part. The effects on J(O_3) and J(NO_2) are therefore well separated. We added in the manuscript that the UV part is the most refine part of the spectrum in section 2.1.2.

Note 5: P16699 line 26 sensitivity, not sensibility

OK

Note 6 : P16704 + the authors describe the (small) changes in photolysis rates with on line cloud treatment, but do not explain why the changes occur (what is the physical mech- anism captured by the more complex treatment?).

The physical mechanism captured by the more complex treatment of clouds is essentially the possibility to have "superposed" cloud layers. It can also captured the non-linear response in deep cloud. This was partly described in section 2.1.3. but we added the following sentence to the text to make it more clear: '*"there are conditions under which the approximation is inappropriate, for example in the presence of multiple layers of clouds (as only a single layer is simulated) or in deep clouds, where attenuation is non-linear (Wild et al., 2001)."*

Are changes of this magnitude important? Are they within the general uncertainties of any measurement, or model? Are they "correct"? Similarly with the inclusion of aerosol. Figure 1 shows that R-ATT is most often closest to R-AERO (except at the ground when the two cloud-only models agree) - is the addition of two complexities in the modelling warranted by the modest differences observed between the most complex and the simplest treatment of cloud /aerosol? The authors are correct to illustrate the magnitude of the cloud and aerosol influences, but having done so, are the differences in photolysis rates significant?

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Firstly, the figures that illustrate photolysis rates changes arising from the different simulations have been changed. In the previous version of the manuscript, we chose to show the absolute value of all photolysis rates (clear sky, R-COnL, R-ATT, R-AERO) (see Figure 1) in order to be able to compare changes induced by each simulation. However in this figure, the magnitudes of those changes were not easily readable. According to comments of both reviewers we decided to show relative differences on photolysis rates between R-COnL/R-ATT and R-AERO/R-COnL (Figures 1 and 4). For R-COnL vs R-ATT, relative differences are small at the ground (around % 3 km) and reach 12% inside clouds, for R-AERO vs R-COnL, relative differences reach 14% at the ground. Secondly, the reviewer is asking about the significance of photolysis rates changes. Our paper focuses on air quality implications of clouds treatment and on aerosol inclusion in photolysis rate computation. Therefore to evaluate the significance of these treatments, we evaluate the impact on air quality pollutant concentration (whether than on photolysis rate values). The magnitude of this impact is now clearly estimated and compared to model uncertainties in section 3.4.1 (see our answer to Note 2)

Note 7 : Aerosols not only influences photolysis rates through changing the actinic flux, but can also form part of the following chemical reactions and so have a dual effect on gas con- centrations and aerosols. This is most pronounced where aerosols are high, as some of the figures show, although changes are large for a limited number of species. The tables comparing models and measurements show that the three models are always in better agreement with each other than with the measurements, implying that there is still something missing from the models (but see comment above about uncertainties).

The reviewer is right to say that the three 'models' are in better agreement with each other than with the measurements. The new section 3.4 (*"Comparisons of the impact of photolysis rates modeling to model uncertainties"*) give a range of mean concentrations that can be obtained using different parametrisations (i.e. different "models",

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for example by changing the vertical diffusion parametrisation or heterogeneous reaction) . The objective is simply to evaluate what are the species the most impacted by changes in photolysis rates calculation and to quantify this impact in comparison of other parametrisations. Photolysis rates calculation does not have the strongest impact on classical measurements (hourly O_3, NO_2, PM) and other model parametrisations are certainly more responsible for the bias between model and measurements (see new section 3,4). Photolysis rate calculation has a strong impact on OH concentrations but they are not measured regularly over Europe, and also systematically reduce O_3 high values (strong impact on O_3 bias compared to other parametrisations). The new comparison of modeled and measured exceedances of O_3 information threshold (see answer to Note 11) shows a strong reduction in the bias between measurement and model when including aerosols in photolysis rate calculation. This comparison (suggested by the reviewer) shows a real improvement when using aerosol in photolysis rate calculation.

Note 8: For the daily ozone peaks (important for air quality alerts) the model R-AERO is least representative of the measurements, implying that it would incorrectly reduce the num- ber of alert warnings. Perhaps hourly peaks would be represented differently (R-AERO is best for average hourly ozone) - could the authors provide this information.

Compared to EMEP measurements, R-AERO is least representative of measurement in July (O_3 peaks are under-estimated). However in November, ozone peaks are best represented by R-AERO. To expand the discussion, we added a comparison to another European database stations: airbase (Table 5) and we added the following comment to the manuscript: *"Both O_3 peaks and hourly O_3 are better reproduce with R-AERO for EMEP and Airbase. The bias is systematically reduced by several per cent for both months and both networks. The errors (RMSE and NME) and correlation coefficients are respectively decreased and increased with R-AERO excepted in July for O_3 peaks. In that case, R-AERO leads to an under-estimation of those peaks when in all other*

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case, the reference model over-estimates hourly and peak O_3, explaining the better reproduction of O_3 measurements with R-AERO." The reviewer is suggesting to look at hourly peaks, but it is already the case (it was not specified but O_3 peak are O_3 hourly peak. It is now specified in the text).

Note 9: P16717 Please say how many EMEP (and AERONET) stations were included in the comparison with measurements)

Done, in table captions.

Note 10: Table 4 caption - clarify that this is the average hourly O_3 for the month, and also for ? Stations

Done.

Note 11: Table 5 - similarly, the average daily peak O_3 for the month and ? stations. In the case of table 5 the average daily peak ozone, averaged over a month and several stations, is not very instructive. Would it not be better to show how many daily peaks (for all stations in the month) exceeded limits - for measurements and for each model.

This is a very interesting suggestion that we followed. We added a table to compare exceedence of O_3 information threshold measured and simulated at Airbase stations in July. The following comment was added to the manuscript: *"The number of O_3 exceedances measured and simulated with and without including aerosols in photolysis calculation is compared at the Airbase stations in Table 7 (as EMEP are"background" stations, exceedances of pollutant threshold are rare). This number is overestimated with R-COnL with a bias of 23%. R-AERO simulation reduces bias to 2%. The RMSE is also reduced. However, the NME is larger with R-AERO than R-COnL and the correlation coefficient is lightly lower. The significance of the bias reduction (larger than any other changes on computed statistics in Table 6 and Table 7) and the RMSE reduction, suggests that R-AERO better reproduces O_3 peaks and exceedance of O_3*

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information threshold."

Note 12: Figure 12 - what is the unit on the coloured scale, is it really difference in number of exceedances, or is it percent difference? The difference in number is hard to interpret without knowing the absolute number of exceedances from one of the models.

We finally think that this figure does not bring any new or really interesting information, since we added a comparison of exceedances of O_3 information threshold to measurements. Therefore, we decided to remove it.

Note 13: Technical corrections: The use of English could be improved. There are many minor grammatical errors. These are not sufficient to prevent understanding of the text, but they are annoying and detract from the general impression of the manuscript. The errors are too numerous to list here, but pay attention to plurals, and use, or not, of the definite article (the).

We tried to correct grammatical mistakes.

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Answer to Reviewer 2:

We would like to thank both reviewers for their useful remarks which really help to improve the manuscript. In the following text, we wrote the questions of the reviewer and our associated answer.

The authors use two different schemes to compute photolysis rates in the presence of clouds and aerosols. They do this within an European-scale air quality model, and assess the consequent changes in gaseous pollutants. Attempts are made to explain some of these differences, and to compare with measurements. Neither the photolysis schemes nor the air quality model are new. The analysis and explanation of the results is detailed and long, but much of it may be more speculative than robust, with very little supporting evidence. The comparisons with measurements are minimal and show essentially no difference between using the different schemes. Overall, the manuscript appears appropriate as an internal progress report on model development, but not as a publication in a scientific journal. If a major revision of the paper is attempted, I would suggest shortening substantially, focusing the discussion on a few robust results, and examining the reasons for these results in detail with support from sensitivity calculations and measurements. Some specific comments follow.

The manuscript was shortened in two ways. First, we decided to not present all the results in detail concerning the impact of cloud parametrisation because its impact on pollutant concentrations at the ground is low compared to the impact of aerosols. Section 3.2 was therefore shortened and does not contain sub-sections any more. Second, we also decided to focus on the month of November for section 3.2 (cloud parametrisation impact) and the month of July for section 3.3 (aerosols impact) in the detailed comparison of the impact on photolysis rates and concentrations, because

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there are the months for which the cloud parametrisation and aerosols have the most impact respectively. However, both months are kept when comparing to measurements and when quantifying the impact on pollutant concentration at the ground. Explanations of some specific features observed in figures of relative differences have also been shortened and are more clear now (section 3.3.2 on NO_2 concentrations at 3-4 km and section 3.3.3 on HNO_3 ground concentrations). However, we have also included new features (tables and text) following reviewers suggestions. Therefore the paper is finally only reduced by 8 pages.

We have also added more details on model uncertainties and the impact of the photolysis rate calculation method on ground concentrations compared to other model parametrisations. The discussion on those uncertainties was moved it to a new paragraph (3.4) which we named *"Comparisons of the impact of the modeling of photolysis rates to model uncertainties"*. To make the comparison easier, we provided a table with a quantitative evaluation of uncertainties, and we re-wrote this paragraph. We also added a comparison with measurement from another data network (Airbase). In particular we compared the number of simulated and measured exceedances of O_3 threshold and show a real improvement when including aerosol on photolysis rate calculation. In this manner, we showed the importance of including aerosols on photolysis rates calculation for air quality and made the manuscript more valuable.

In the methods, it would be nice to see much more detail on processes that contribute the most relevant uncertainties: How are clouds produced in the model, and how well do they agree with cloud observations?

CTM does not compute meteorological fields. They are computed off-line and here reanalysis of ECMWF are used. To diagnose a cloud, the following common method is used: Relative Humidity (RH) from ECMWF is compared to a Critical Relative Humidity (CRH) which varies with pressure and ground pressure. If RH $>$ CRH, a cloud is

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diagnosed. We did not compare clouds from ECMWF to measurements, but reanalysis of ECMWF meteorological fields are believed to be the best available meteorological data for Europe.

Do you make a correction for the actinic flux inside the droplets?

In Fast-JX, droplets modified the actinic flux as part of cloud. Clouds optical depth and optical properties are calculated depending on the mean droplets radius. As there is no cloud resolved model in our CTM, there is no treatment of single droplet.

How is Mie theory also used for ice particles?

The paragraph 2.1.3 describing our treatment of clouds and particles optical properties was confusing. Mie theory is not used for ice particles. Fast-J contain prescribed input values of OP for water clouds with different droplet sizes and for ice clouds. We used these prescribed values in our study. We changed the text in paragraph 2.1.2 (Fast-J description) and 2.1.3 (cloud attenuation parametrisation) and added the following sentences: *"For clouds, pre-calculated values of OP are included in Fast-J for several cloud droplet sizes and ice crystal shapes."* (2.1.2) *"Here, we use the Fast-JX prescribed values (see section 2.1.2) for cloud droplets of 10 μm and irregular-ice crystals."* (2.1.3)

For the aerosols: How well does the model estimate secondary organic aerosols? According to Figure 5b the SOA is an order of magnitude smaller than sulfate, while measurements (e.g. see Jimenez et al., Science 2009) show sulfate and SOA are comparable.

The SOA model used probably underestimates SOA, as mentionned by reviewer. A paper on the modeling of SOA in Polair3D is under review in JAWMA (Journal of the Air and Waste Management Association). Therefore, the impact of SOA on photolysis rates is also probably underestimated as it is now mentioned in the paper. We added

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the following sentence at the end of paragraph 3.3.1 : *"Also, SOA concentration are underestimated with the SOA model SORGAM (Kim et al., JAWMA 2010, under review) and the global impact of SOA is probably underestimated"*

How are core/shell aerosols modeled?

Aerosols are modeled as core/shell only for the refractive index calculation. The modeling is based on Tombette et al., 2008 that used the Maxwell-Garnett approximation (for core in a solution) to calculate the refractive index of non-mixed aerosols. All details are given in Tombette et al., 2008.

The method of Mallet should be described. How does it differ from that of Chang et al.? Wild et al. actually found that the Chang et al method worked quite well, with a few exceptions (leaving the reader with a different impression than given by the authors of the present paper).

In fact Mallet used the same method as in CMAQ (Roselle et al., 1999). It is adapted from the method of Chang et al., except for the cloud optical depth where empirical formula from Stephens (1978) is used. We should have mentioned Roselle et al. instead of Mallet et al. We replaced our sentence: *"In the standart version of Polyphemus, the impact of clouds on photolysis rates is calculated through an attenuation coefficient Att applied to clear-sky photolysis rates (Mallet, 2005). In this parametrisation, similar to the technique used by Chang et al. (1987) ..."* by *"In the standart version of Polyphemus, the impact of clouds on photolysis rates is calculated through an attenuation coefficient Att applied to clear-sky photolysis rates (Roselle et al, 1999). This method is adapted from the method of Chang et al., except for the cloud optical depth where empirical formula from Stephens (1978) is used"*. We do agree with the reviewer and with Wild et al. that the method of Chang et al works quite well outside clouds. We mentioned it several time (in the abstract : *"Outside clouds, differences are small"*, in section 3.2.1 : *"below clouds, differences are much*

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lower, not higher than a few percents on average" or in the conclusion : *"At the ground those differences are small"*). But inside clouds, differences can be large, as also mentionned in Wild et al : section 4.2: *"For the deep cloud, the influence of a full scattering treatment is more evident. .. With two layers of clouds, the benefits of a full scattering calculation are again clear"* section 4.3: *"The Chang technique has closer agreement with fast-J, but underestimates photolysis rates in the mid-troposphere for the tropics and mid-latitudes."* . As mentioned previously, we decided to focus on the impact on air quality (i.e. at the ground) and as the impact of cloud scattering treatment at the ground is small, we shortened the paper by removing detailed explanation of the differences between the simulations with the two different cloud scattering treatments.

In the results section, Figure 1 shows that the average differences in J values between clear sky, aerosol, and cloudy cases are really quite small. But then it is misleading to say that the differences between the R-Att and R-COnL models are of the same order of magnitude as differences between R-ATT and simulations with no clouds (abstract/7, and p.916704/20,21) , when in fact all of these differences are very small on average.

We agree with the reviewer that Figure 1 gives the reader the feeling that differences are small. Therefore we decided to replace it with a figure showing relative differences (instead of the absolute values) between R-COnL, R-Att and Clear-sky runs. In this figure, it is clear that differences between R-COnL and R-Att are small at the ground and reach around 10% inside clouds. We also added a figure showing relative differences between R-Aero and R-COnl (Figure 4) that shows mean differences around -14% at the ground, when taking into account aerosols.

It would be interesting to see the frequency distributions of the changes , rather than domain-averaged or monthly averaged values. This is where clouds have the most effect, and the results might be sensitivity to the different cloud schemes.

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We have plotted the frequency distributions of the changes between R-COnL and R-Att at the ground (sse attached figure). This figure shows that most of the differences are lower than 2%. It also shows that during 3% of the time, they are larger than 50%. As we mentioned above, we decided to reduce the length of the discussion on cloud attenuation parametrisation, and therefore we did not include this figure in our paper, but we added the following sentence to section 3.2: *" At the ground, differences are most often lower than 2% (during 80% of the time) but they can sometimes be larger than 50% (3% of the time)"*

Line 16705/10-14: Contrary to the assertions made here, the drop radius should have a large effect on photolysis rates and radiation in general (e.g. the Twoomy effect). The fact that only a small difference is found here should be discussed.

The test at the end of section 3.2.1 was badly made. We did perform a sensitivity test by changing the droplet radius when computing OP (i.e. we chose different OP pre-calculated in Fast-J for different droplet radius). However, we did not change the OD (needed as input of Fast-J) although it should be modified with droplet radius. Therefore our discussion is incorrect. We decided to reduce the length of the discussion on cloud attenuation parametrisation, and therefore we decided to remove this discussion on the impact of droplet radius on photolysis rates.

It is surprising (Table 4-7) that no improvement in the comparison with measurements is found for any of the methods.

We do agree that model/EMEP-measurement statistics at the ground are not improved when using a better cloud parametrisation (R-COnL vs R-ATT). Those statistics are slightly improved when including aerosols in photolysis rates calculations. In the new version of the manuscript, we added comparisons with Airbase data (734 stations over Europe). For these stations, larger improvement is obtained when including aerosols (R-AERO vs R-COnL) for hourly and peak O_3, as well as for the number of

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exceedances of O_3 information threshold.

Other studies (e.g. Pour-Biazar et al., JGR 2007) found that assimilating cloud data from satellites gives improved prediction of surface O_3. I guess the problem here is that the clouds are still predicted by the model, so they don't correspond to real clouds. This of course is a much bigger error than the radiative transfer calculations.

The model we used is a CTM and clouds are diagnosed using ECMWF data. We agree with the reviewer that large error may come from the simulation of clouds, much larger than the impact of radiative transfer calculations in, above and below clouds. We added the following sentence in the cloud parametrisation discussion :*" A better representation of clouds by assimilation of cloud data from satellite for example will have a larger impact on surface O_3 (Pour-Biazar et al., JGR 2007)"*.

The explanations of how chemical concentrations respond to changes in j values are very qualitative and not obviously correct. For example, it is well known that one of the consequences of decreasing JNO_2 is to decrease the lifetime on NOx, by shifting NO to NO_2 and making it available for OH+NO_2 -> HNO_3. Effects from reducing the NO_3 photolysis are mentioned, but these seem unlikely during the day because it is nearly instantaneous anyway.

We do agree that decreasing JNO_2 will shift NO to NO_2 making it avaliable for OH+NO_2 $\rightarrow$ HNO_3. However, the shift from NO to NO_2 is lower than the large decrease in OH due to the JO^1D decrease. This OH decrease limits the transformation of NO_2 to HNO_3 and HNO_3 concentrations mainly decreased as mentioned in the paper.

More generally, there have been theoretical studies looking at the sensitivities of chemistry to changes in j-values, e.g., the well known scaling of OH and HO_2 with the square root of j values at low NOx, and the linear dependence at high NO_x (see for example Ridley et al., JGR 1992:, or Kleinman et al. JGR 2005). The

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chemical responses should be analysed in the framework of these (and other) previous studies.

We did not find clear evidence of the *"scaling of OH and HO2 with the square root of j values at low NOx, and the linear dependence at high NOx"* in Kleinman et al. JGR 2005 :"A comparative study of ozone production in five U.S metropolitan areas". We found that Bloss et al., 2005 showed that at a remote location, measured OH concentrations are strongly linearly dependant on JO^1D. They also showed that their global chemistry model evaluated a strong correlation between OH concentrations and JO^1D (correlation coefficient > 0.9) in the whole troposphere. The work of Lefer et al. 2003 also showed a linear relationship between OH and JO^1D. They used aircraft measurements taken in the troposphere to run a box model. They explain this linear relationship by the following argument: *"the direct production of OH from O^1D) and H_2O was the dominant source of OH, while the primary OH sinks (i.e., OH + CO, OH + CH_4) did not vary with changes in photolysis frequencies"*. Furthermore the sink of OH through NO_2 reaction (to form HNO_3) varies by change in photolysis frequencies (more NO_2) but as NO_2 concentration is low on average over Europe, this does not play a strong role (as shown in Kleinman et al. JGR 2005, at low NO_x only a very small part of OH is converted to HNO_3 by the reaction with NO_2). What Lefer et al. 2003 did show is that HO_2 / JO^1D relationship is best described by a power function with almost a square root dependence. We added the following sentence to paragraph 3.2: *"In general OH concentrations vary linearly with JO_3. This was also observed by Bloss et al. (2005) and Lefer et al. (2003) that found a strong linear relationship between OH and JO_3 both in the measurement and in their models. This can be explain by a strong dependence of OH source on photolysis rates whereas OH sinks are less dependant on those rates. For example, the sink reaction of OH with NO_2 to form HNO_3 is influenced by photolysis rate modifications but changes in NO_2 concentrations are small on average."*

Style: I agree with referee 1 that the use of the language should be improved.

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Line 16694/Eq.2 and related discussion: J is actually the photolysis rate coefficient, not the photolysis rate.

We replaced “photolysis rate” by “photolysis rate coefficient” each time we though it was necessary.

Line 16699/13: Specifying 4 or 5 significant digits for the coefficients a and b seems excessive, since the precision to which tau is determined is probably no better than 10-20%, and often worse.

As we use the method of Pozzoli et al (2008), we used the same number of digits as in their work. We do agree that precision is probably excessive and we reduced it to 2 digits.

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