**Atmospheric
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Comment

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

Anonymous Referee #2

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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 calcula-

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tions and measurements. Some specific comments follow.

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? Do you make a correction for the actinic flux inside the droplets? How is Mie theory also used for ice particles? 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. How are core/shell aerosols modeled? 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 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.

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. 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.

It is surprising (Table 4-7) that no improvement in the comparison with measurements is found for any of the methods. Other studies (e.g. Pour-Biazar et al., JGR 2007) found that assimilating cloud data from satellites gives improved prediction of surface O3. 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 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 JNO2 is to decrease the lifetime on NOx, by shifting NO to NO2 and making it available for OH+NO2 -> HNO3. Effects from reducing the NO3 photolysis are mentioned, but these seem unlikely during the day because it is nearly instantaneous anyway. 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 HO2 with the square root of jvalues at low NOx, and the linear dependence at high NOx (see for example Ridley et al., JGR 1992, or Kleinman et al. JGR 2005). The chemical responses should be analyzed in the framework of these (and other) previous studies.

Style: I agree with referee #1 that the use of the language should be improved. Line 16694/Eq.2 and related discussion: J is actually the photolysis rate coefficient, not the photolysis rate. 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.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 16691, 2010.

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