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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurements and modelling of molecular iodine emissions, transport and photodestruction in the coastal region around Roscoff" *by* R. J. Leigh et al.

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Response to Referee comments for "Measurements and modelling of molecular iodine emissions, transport and photodestruction in the coastal region around Roscoff", R. J. Leigh et al.

Authors: R. J. Leigh and S. M. Ball. Date: 15th September 2010.

The authors would like to thank the referees for their constructive and helpful comments which have helped to significantly improve this paper through the review process.

All minor technical corrections requested by the referees have been made. Many fig-

ures have been re-plotted to improve their legibility, and we have taken care to place the figures at the right places in the manuscript (referee 2).

Both referees considered this work to be "scientifically highly interesting and timely" and to present "novel concepts, tools, ideas and data". However the referees raised concerns about the modelling of I2, specifically: Referee 1 commented that [not being from a modelling background] "it is hard to assess whether the results are indeed sufficient to support the interpretations and conclusions". Referee 2 commented that while the correlations [between model and measurement] indicate "at least a general understanding of the emission rates & sources", she/he would like to see "a stronger emphasis on the discrepancies and their potential explanations". In hindsight we accept the modelling and the modelling/measurement comparison could have been executed better, so we have made the following improvements:

1. Emission footprints have been re-calculated for each individual model time step (5-minute), rather than using a linear interpolation between a few set footprints. This allows changes in the footprints with tide height and day/night conditions to be more accurately captured.

2. Footprints are now averaged over 5 degrees in each direction around the wind vectors (as measured at the RHaMBLe site) to allow for some directional variability in wind vectors over each time step.

3. The mixing layer depth above the seaweed beds has been reduced to 15 cm. This now produces peak I2 concentrations immediately after the seaweed is first exposed to air that broadly agree with peak I2 concentrations measured in the lab studies of Ball et al (RHaMBLe special issue, ACP, 2010). For example, the peak concentrations of I2 modelled above newly exposed Laminaria digitata and hyperborea is 15 ppbv, in good agreement with studies by Ball et al.

4. Seaweed plants have been assigned species-specific heights (Table 1), allowing the taller species to be exposed (and to start emitting into the atmosphere) before the sea

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bed is completely uncovered.

5. Comparisons between the modelled & measure I2 amounts are now first discussed for night-time conditions. This serves to establish a baseline test of the model's performance (i.e. the I2 photolysis is decoupled from modelling of emission footprints/transport), prior to the introduction of photolytic destruction and any recycling mechanism with their attendant extra uncertainty.

6. Additional modelling of iodine and NOx chemistry has been undertaken using the model of Mahajan et al. Geophys Res Lett 2009 (who also first proposed I2 recycling in the semi-polluted marine atmosphere via the formation of the IONO2 reservoir). Specifically, the Mahajan model has been used to examine how I2 concentrations decay away downwind of an emission source in the presence of NOx (for an assumed [NO2] of 2ppbv, representative of the RHaMBLe conditions) and in the absence of NOx but for reduced I2 photolysis rates. Good qualitative and reasonable quantitative agreement was found for these two scenarios when the I2 photolysis rate is decreased to 10% of its typical daytime value, providing a validation for our simplistic (but we believe effective) approach of reducing the effective I2 photolysis rate to mimic the effect of I2 recycling(section 5). The result of this extra chemical modelling show that sequestering I2 as IONO2 and the subsequent "re-release" of I2 provides a mechanism for transporting I2 much greater distances from its source compared to the case where I2 is rapidly photolysed. Indeed without recycling, the model in this paper predicts that no day-time I2 would be observed at the BBCRDS measurement site (which is clearly not the case).

7. In figures showing measured & modelled I2, we now also show NO2 measurements from a chemiluminesence monitor (shoreline site) and co-retrieved NO2 measurements made with the BBCRDS system. These NO2 data have two purposes: (i) the extent of the I2 recycling via IONO2 depends on NOx concentrations, and this is discussed qualitatively in the text, and (ii) the generally good correspondence between the chemiluminescence NO2 and BBCRDS NO2 serves as an extra quality control check on the

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BBCRDS I2 measurements.

The above improvements in the execution of our modelling have also significantly improved the model's agreement with the LP-DOAS and BBCRDS measurements. Some discrepancies remain, and the various potential reasons for the discrepancies are now discussed in greater detail in the text including: uncertainties in the wind fields; uncertainty in I2 emission rates from the seaweed species, particularly for L. Ochroleuca which was not tested in the Ball et al seaweed lab study; uncertainties/mapping errors in the distribution of seaweed species particularly close to the measurement site.

The wider implications of applying our methodology to model atmospheric chemistry in inhomogeneous emission fields have also been discussed to improve the potential audience of this paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 21165, 2009.