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Interactive comment on "Modelling molecular iodine emissions in a coastal marine environment: the link to new particle formation" *by* A. Saiz-Lopez et al.

Anonymous Referee #2

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General comments: This paper first presents an analysis of the spatial distribution of iodine species around Mace Head by combining model simulations with in-situ and long-path-averaged observations. The scientific methods and assumptions in this part are valid and clearly outlined. The conclusion that the emission of I2 mostly occurs in the intertidal zone is reasonable, which has not been tested by the in-situ observations before. The authors extend their modeling into the simulation of new particle formation due to the production of iodine oxides in the atmosphere. In this part, more explanation of the model is needed especially regarding the treatment of the particle nucleation. I suspect that the assumption that I2O2 participates in the new particle formation, which

is not usually assumed in other studies, is key to the successful reproduction of the observed particle size distributions. If it is true, the authors should mention that the results are sensitive to this assumption and present the results of model sensitivity runs without producing small particles from I2O2. Also, it is meaningful to point out that more study is necessary in the future to clarify whether I2O2 participates in the particle formation as well as I2O3 and I2O4. The paper fits into the scope of Atmospheric Chemistry and Physics and I recommend publication after minor revision.

Specific comments: 1. page 5409, line 19. The authors should include more description regarding how the particle nucleation is modeled. The sentence "it is assumed that the smallest particles in the first size bin can be either I2O2, I2O3 or I2O4" is not sufficient. Do the iodine oxides, once formed in the gas phase, completely undergo the formation of the small particles? Or the reversible processes between the gas phase and the particle phase (condensation and evaporation) are adequately considered similarly to O'Dowd et al. Nature, 2002 and Burkholder et al., ACP, 2004? How is the number density of the smallest particles determined? In relation to this, the authors mention that the uptake rate of IO, OIO, I2O2, I2O3, I2O4 and larger iodine oxide particles onto background marine aerosol is calculated using uptake coefficients gamma of either 1 or 0.1. (page 5411, lines 2-4) It is not clear to which species gamma=1 is applied and to which gamma=0.1 is assumed.

2. page 5410, line 23. The authors note the difference between their assumption and that made by Burkholder and coworkers being the formation of particles from iodine oxides other than the dimerisation of OIO. How sensitive are the results of this study to this assumption?

3. page 5411, lines 2-4. Is the uptake of HOI considered in the model as well as those of iodine oxides? Its uptake on the sea salt particles would lead to the autocatalytic release of halogen molecules, making the iodinated species circulate between the gas and aerosol phases for a certain period. I would suspect that gaseous iodine species would survive longer than shown in Figure 3 and 7 if this regeneration mechanism

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works efficiently. This might also affect the discussion of the [IO]/[I2] ratio below.

4. page 5411, line 9. It is not reasonable to assume that the HO2 level is constant because its loss could be highly controlled by its fast reaction with IO, whose concentration changes by more than an order of magnitude in the model runs (Kanaya et al., GRL 2002, Bloss et al., GRL 2005). If the above-mentioned role of HOI is significant, more careful treatment of HO2 is necessary.

5. page 5413, section 3.3. The starch denuder collection followed by the ICP/MS detection of I2 is new and technically attractive. Is this technique free from interference?

6. page 5416, line 26. In the model run with the localized I2 emission, a box of 320 m length is assumed. How is this length compared with the typical horizontal scale of the intertidal zone covered with seaweeds possibly emitting I2? Related to this, “a box 360 m long” mentioned in the Figure caption (Figure 3b) might be a typographical error.

7. page 5418, line 19. Particle number density, not particle mixing ratio.

8. page 5420, lines 11. Why is the new particle formation faster in the run assuming a larger J(OIO) value? This trend is opposite to that described by Burkholder et al., ACP 2004, where significant particle production only occurred with low J(OIO) values. Does this suggest that the ultrafine particle formation is dominated by the self-reaction rate of IO (i.e. the I2O2 formation rate) rather than the production rates of I2O3 or I2O4 in this study?

9. page 5421. For the model runs whose results are shown in Figure 10a, how do the authors assume the I2 mixing ratio? Is it assumed that I2 emission follows a Gaussian distribution with a width of 22 min as mentioned before? In Figure 10a, is the unit of delta t (x-axis) “hour”, as opposed to Figures 3b and 6 (maybe “minutes”)?

10. page 5422, line 16. Please double check the size-integrated number density of the ultrafine particles of 2x10**3 cm**-3 after the model integration of an hour. I would suspect it is lower than 2x10**3 cm**-3 as seen from Figure 11.

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11. Reference, Page 5424, line 33. Atmos. Chem. Phys., not J. Phys. Chem. A.

12. Figure 7. The right axis title should be O3 (not O3 depletion)

13. Figure 9. Is the x-axis in Local Time? The sunset time should be indicated by broken vertical lines.

14. Figures 5, 9, and 10b. The unit of the color-coded dN/dlogDp should be included. (1e+0, 1e+1, ..., 1e+6 should be used rather than 0, 1, ..., 6 for consistency with Figures 6, 10a, and 11)

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 5405, 2005.