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Atmospheric Chemistry and Physics Discussions

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

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Saiz-Lopez et al. present a very comprehensive and careful analysis of three different techniques (in situ and remote sensing) to measure I_2 and associated aerosol nucleation events and a detailed comparison with a constrained numerical box-model. The data was taken during the NAMBLEX campaign in July and August 2002 at Mace Head, Ireland. The main conclusion is that at this location the most likely precursor for new particles is I_2 and not organic iodine compounds. The potential for growth of fresh nuclei to CCN is explored, which seems, however, to be small based on their model

results.

The paper falls very well into the scope of ACP, is clearly structured, title and abstract are appropriate. I suggest to publish the paper after minor revisions.

Specific comments:

p 5406/5407: Regarding the reaction of DMS with IO "this reaction is slower than believed hitherto": only one publication had reported a high reaction rate coefficient, all other assessments had already concluded that this reaction is rather slow.

p 5407, 2nd paragraph: To my knowledge the iodine enrichment in particles was first reported by Duce et al., 1963, "Atmospheric iodine, bromine, and chlorine". J. Geophys. Res. 68, 3943-3947. This reference or a review (e.g. Carpenter, 2003, von Glasow and Crutzen, 2003) might be more appropriate than the cited model study by Vogt et al.

p. 5407, l. 17: Correct citation of Jimenez et al. The study of Burkholder et al. (2004) might also be cited in this context.

p. 5407, last paragraph: I_2 has a lifetime of about 10s, yet the time resolution of the instruments used here are between 20 and 60 minutes as mentioned later in the paper. Can we expect the I_2 concentrations to be constant during the measurement time? If not - what consequences does this have for the concentrations derived from the measurements, could peak values that might determine the chemistry, not be even higher than the numbers given in the paper?

p. 5407, line 29: Is there a reference for possible open ocean sources of I_2?

Section 2, Model description: It would be useful to have a list of the species that are constrained in the model and those that are not, as well as a list of reactions that are included in the model. This could be added as electronic supplement. Are any heterogeneous reactions leading to the recycling of iodine from the particles included?

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p. 5409, l. 26: What is the fate of I_2O_2 in the model?

p. 5411, l. 8: These high mixing ratios of NO_2 and NO_3 might be typical for Mace Head but I wouldn't refer to these conditions as "clean MBL", given the short lifetime of NO_x.

p. 5411, l. 12: "simple 1-dimensional approach" - I don't quite see why you refer to this as a "one-dimensional" approach as you don't have information on the vertical structure of the BL. Entrainment of outside air is definitely an important process but this doesn't change a zero-dimensional approach into a one-dimenional one. Use of terms like "box model without entrainment" and "box model with entrainment" would be more precise. This comment applies to all other occurrences of this term in the text as well, for example Fig. 7, p. 5419, l. 27, p. 5422, l. 2.

p. 5411, l. 20 - 25: Is there a reference for these micro-meteorological measurements?

p. 5416, l. 10: The model uses constrained values for many important photochemical species, so no complete budget of the chemical ozone destruction and production rates can be made. In order to avoid confusion among readers I suggest to rephrase "O_3 depletion rate" to something like: "iodine oxide related ozone destruction rate". The ratios of $[HO_2]/[OH]$ and $[NO_2]/[NO]$ are affected by iodine chemistry and an assessment of O_3 photochemistry would require the unconstrained evolution of these (and other) species.

p. 5417, l. 4-6: See previous comment.

p. 5417, l. 18/19: Measurements with the BBCRDS instrument were carried out "for a period of four days", yet "Throughout the campaign" you have information on I_2 from that instrument. Maybe rephrase to "on all these measurement days..." and add that these were not consecutive days.

p. 5418, l. 20 and p. 5419, l. 26: To me the use of "profile" implies some vertical information, maybe rephrase to "evolution with time".

p. 5418, l. 7: "September 2003" - on p. 5408 you wrote that the campaign took place

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in 2002.

p. 5418, l. 17: "new particle formation events were obsereved on almost every day.." - this is not evident from Figure 5, is this due to the absence of daytime low tides on some of the days?

p. 5420, l. 7: "an O_3 destruction of 0.25 ppb is predicted": During the time when O_3 decreases, the entrainment of air without iodine and enhanced O_3 is already happening, so the mixing ratio of O_3 only shows the net effect of chemistry and transport. The chemical destruction is certainly higher and it would be helpful to include some information on the strength of the entrainment vs. chemistry, maybe citing the moles of O_3 (total, not in concentration or as mixing ratio) chemically destroyed vs. those that are entrained.

p. 5420/5421, discussion of correlation of I_2 and particle peaks: The particle burst on Fig 9b (typo on p. 5421, l. 2: change "Fig 9a" to "9b") starts BEFORE low tide and I_2 increases AFTER low tide - given that there is roughly a 6h time lag, I think it's unlikely that this burst originated at another location along the coast from intertidal emissions, to me this time lag would rather be an indication of a different nucleation process as mentioned on p. 5421, l. 5. However, I don't see from the plots that the particles have "clearly grown to sizes well above 10nm" - at the beginning of the high-tide nucleation event the maximum diameter is 8nm. Could an assessment of particle growth rates help to identify possible sources, or the distance of sources from the measurement location?

p. 5421, discussion of timescale for particle growth: You mention that "the time required for the newly formed particles to evolve to bigger sizes is normally longer than the transit time" - it would be helpful if you included the times required in the text.

p. 5422, l. 5: Did you use the verticalturbulent mixing coefficients that were derived at Mace Head (see p. 5411, l. 20 - 25)? If so, do you have an indication that the mixing proceedes at the same rate 14km inland?

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p. 5422, l. 16-22: The statement that the iodine containing particles are "fully mixed vertically up to 1 km" is NOT a conclusion but this is how the model was setup (see p. 5422, l. 5).

p. 5422, l. 16-22: In terms of the numbers that you list, the contribution of iodine particles would indeed be high, but would that still hold if coagulation of the iodine clusters with background particles were included in the model?

p. 5423, l. 8/9: See previous comment: I'm not convinced yet that you can make this conclusion from your model study.

Figures:

Fig. 3: Maybe change "the model initialises with" to "the model was initialized with".

Fig. 3, 6, 10: It would be helpful to explain the meaning of δt in the caption of the figures as well and not only in the text.

Fig. 4: Were the measurements taken in Sept. 2003 or at the same time as NAMBLEX in 2002?

Fig. 7: "O_3 depletion" as description of the ordinate: This should rather be "mixing ratios" as "depletion" would either be cumulative (for total chemical O_3 destruction) or a rate.

Fig 9 and 10: The color code of the DMPS is probably the logarithm of dN/dlogDp, not dN/dlogDp itself.

Fig. 11: The caption is hard to understand, maybe rephrase.

References:

Bitter et al. - is it ACP policy to include papers "in preparation" in the reference list or should they rather be cited in parentheses in the text?

Burkholder et al.: published in ACP not in J. Phys. Chem.

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The following names have typos:

Mössinger in McFiggans et al., 2002

Ladstätter-Weißenmayer in Himmelmann et al, 1996

von Glasow in Vogt et al., 1999

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