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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Direct evidence for coastal iodine particles from Laminaria macroalgae - linkage to emissions of molecular iodine" *by* G. McFiggans et al.

G. McFiggans et al.

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Response to Referee 1

The authors would like to thank the referee for the positive comments and comprehensive review. Addressing each in turn:

We have only referred to the experiments involving CH2I2 since we were concerned with the atmospherically relevant systems. As we understand it, CF3I was used in the Burkholder et al. experiments to simplify the experimental interpretation due to the cleaner I atom source and consistent I atom production rate throughout the experiment and for its more straightforward handling. To our knowledge CF3I has neither been isolated in the atmosphere, nor implicated in coastal new particle formation. Whilst it is an interesting technique to isolate the participation of iodine oxides in the mechanism, it is

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not felt appropriate to refer to it more extensively in the current work, which deliberately attempts to focus directly on the atmospheric system.

The hygroscopic behaviour has been described more fully in the abstract.

The requirement for TSCs in the ambient atmosphere is not resolved. Whilst in Burkholder et al., Jimenez et al. and the current study no TSCs were needed to form the particles, in all cases no pre-existing aerosol were introduced into the laboratory systems. Pre-existing aerosol will always be present in the atmosphere and will provide a condensation sink for the precursor iodine oxides. It is well established that heterogeneous nucleation is favoured over homogeneous since the energy barrier is lowered by a pre-existing soluble or insoluble nucleus. The competition for iodine oxide vapours with the pre-existing accumulation mode aerosol may not be won by a homogeneous process such as those in the current studies, but may be won by heterogeneous stabilisation of background TSCs. We prefer to leave the participation of TSCs as an open question. The argument has been presented as above in extending the discussion in section 6 of the paper.

The light levels used may be described as variable. They were generally not conducted in direct sunlight. The Mace Head experiments were done on reasonably bright but overcast days, inside an open sea container. The experiments in the laboratory at UMIST were conducted over several months under typical Manchester ambient daylight 2 metres below a partially glazed roof. Further controlled light experiments are planned.

The experimental technique section has been reordered and modified.

The algae were different specimens cropped in batches as described in the paper. They were kept for several months in the longest case, and stored as described in the paper. The storage procedure section has been modified in response to referee 2.

We would like to thank the referee for the error in our sulphate mass fragmentation

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attribution. This has been rectified.

References to the works on I2 + O3 have been added and comparisons made.

To clarify the argument concerning the TDMA measurements: HTDMA measurements were made for the I2 + O3 system in this study. Jimenez et al. made measurements on the CH2I2 + O3 system. We did not make measurements on particles formed in the macroalgae + O3 system, but as the referee rightly points out Vakeva et al (2002) reported measurements made during Mace Head particle bursts. A discussion of a comparison between the measurements has been made and is included in the revised script, but does not add greatly to understanding of the system.

Concerning the DOAS retrieval of I2, the section has been modified and extended, and the reader is pointed to the additional reference of Allan et al., 2001 for a discussion of the technique.

Regarding section 6, we believe that the TSC question is not independent of whether the particles grow to CCN size. Various recent publications by other groups also state this to be the case. The ability to overcome a coagulation barrier by allowing TSCs to grow by condensation of vapours other than those forming them will determine the number of viable particles competing for available vapour to become CCN size. Alternatively, the homogeneous formation of iodine-only particles will change the dynamics such that a different population of viable particles is competing for the same vapours. Aerosol dynamics models (such as AEROFOR, e.g. Pirjola et al., 2000) show that the population of CCN is determined by nucleation rate, pre-existing particle condensation sink and condensable vapour production rate. Thus at a given vapour production rate and background population, the Şapparent nucleation rateŤ or rate of production of stable particles determines the CCN population. This production rate of viable particles of sizes above the coagulation barrier is entirely dependent on the mechanism. This is reflected in the amended section 6. The cluster dynamics (relying on identification of the intermediates by molecular dynamics simulations or spectroscopic determination)

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Interactive Comment

and competition with stabilisation of pre-existing TSCs is the subject of an extensive ongoing study and a complete discussion must remain beyond the scope of the current study.

The grammar, spelling and formatting issues have been addressed.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 939, 2004.