

Interactive comment on “Chemical and physical characteristics of aerosol particles at a remote coastal location, Mace Head, Ireland, during NAMBLEX” by H. Coe et al.

H. Coe et al.

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First of all, may we express our thanks to reviewer for their comments and appreciating the value of the data we present.

The responses to the general comments made by the reviewer are given below:

1) The reviewer comments that the data may be of use for other aspects of the NAMBLEX campaign and the data should be connected to other aspects of the NAMBLEX Special issue. The majority of the work conducted during NAMBLEX was focused on gas phase photochemistry, ozone formation/destruction and halogen cycling. The main links between these data and that included in the other papers in the special issue is the role of aerosol loss, and, the role of the shore in perturbing the loss locally. Both of

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these topics are covered in some detail in this paper, which, in the context of the special issue as a whole provides the context for the analyses conducted in some of the other papers. We will extend the relevant sections to more closely link these papers to our work by including discussion paragraphs. There are strong links between this paper and that of Dall'Osto et al in the same special issue and these are also discussed in this paper

2) The reviewer wishes us to reword the abstract to provide a clearer overview of the goals of the paper and to provide clarification in other sections of the paper. We accept that in places the paper did not clearly identify the goals of the individual sections. We will rewrite the abstract, the latter part of the introduction section and the section on the heterogeneous uptake calculations to improve the clarity of the overall aim of the paper.

3) The reviewer suggests that the section on the aerosol chemical composition should be extended. We have not extended this section of the paper. The main findings in our opinion are: 1) there is a significant background of mixed sulphate and organic aerosol on the western edge of Europe in westerly conditions, indicating a background input of aerosol to Europe; 2) that during periods of long range transport, identified by elevated CO and black carbon the organic fraction or the aerosol show strong similarities with measurements of aged continental organic aerosol observed in other locations, but in marine conditions this may not be the case. Links are therefore made to the work of O'Dowd and the paper by Dall'Osto et al who discuss this in more depth in this issue. This work is discussed in depth and relevant literature is used to compare and contrast with the measurements we present. The impactor data, showing chlorine depletion and nitrate in the coarse sea salt mode is not a new finding but supports other papers within the issue which assume sea salt is the main reacting surface of the particles. Hence we did not extend this approach.

The reviewer makes a series of specific points. These are addressed in the following ways:

- On page 11654 line 10, we will change the sentence to “For example A+W 1998 measured a lower limit of 0.2 for the uptake coefficient of HNO₃ to deliquescent NaCl”.

- On page 11657 line 1, the sentence will now read: “However, expression (1) is only valid in the molecular regime where the mass accommodation dominates the overall loss.”

- The reviewer questions our use of expression (3) as presented by Schwartz (1986). The expression assumes that transport rates as calculated in the liquid, in the continuum regime, and in the gas, in the gas kinetic regime, match at the droplet surface. This is not the case. Several flux matching approximations are available to overcome this and the approach of Fuchs and Sutugin (1971) is one such. However, Sander (1999) has shown that though the uptake expression (3) is never greater than the Fuchs and Sutugin flux matching approach the difference between these two approaches is negligible over most of the solution space. A maximum in the difference of only 15% only arises when the ratio of the mean free path of the gas to the particle radius, the Knudsen number, is close to one and the accommodation coefficient approaches unity. As Sander (1999) points out this is far less than the uncertainty in the accommodation coefficient in most cases and so does not present any practical reduction in accuracy over any particle size range. Furthermore, the expression used in this work has the added advantage of easily separating the gas phase diffusion rate from the accommodation rate at the surface of the particle. This aspect has been used extensively in our discussion of the next section of the paper. The above discussion will be included in the revised paper. The reviewer asks how surface and condensed phase chemical reactions may alter our conclusions given that HO₂ and HOI are highly reactive. The net flux across the particle surface, derived assuming the kinetic regime limitations, is proportional to the concentration difference between the species in the gas and solution phases as well as by the uptake coefficient. Whilst our analysis does not explicitly include chemical transformation, we have also calculated the loss rates with uptake coefficients of unity to demonstrate the maximum possible loss, whether the pathway is through physical or chemical routes. The following sentence will be included “When $\gamma = 1$ uptake of the gas at the particle surface cannot be faster,

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providing the upper limit to surface transfer rate, whether by physical or chemical loss.” In certain places we have used the term “accommodation coefficient”, which may have led to the reviewer’s question. This will be revised in any resubmitted paper to make it clear. Our analysis is about what limits overall uptake rate, either transfer of material to the surface from the gas or uptake across the surface itself. The purpose of this section was not to discuss whether uptake was reactive or purely absorptive. This will be different for different systems.

The reviewer wishes to know how “the diffusion limitations to mass transfer connect with the composition of the particles?” and then says “the gases addressed in each section are different”. These are really two different points. To address the first point: A single gas may have varying uptake rates to different particle surface types, whereas different gases may have variable uptake rates to the same surface type. The former is dictated entirely by changes in accommodation or reactive uptake rate, as derived from laboratory measurements; the latter is also dependent on the binary diffusion coefficient and average gas velocity, both of which are species dependent. However, the variation in both of these parameters is due almost entirely to their dependence on the inverse of the square root of the molecular weight of the gas. As a result, if both HO₂ and HOI are assumed to have the same uptake coefficients, the overall uptake rate for HO₂ will almost exactly be a factor of 2 greater than that for HOI for any given particle size. This is treated in more detail in the revised section which discusses this analysis in combination with other papers in the NAMBLEX special issue. The second point: “the gases addressed in each section are different” is not correct. In both graphs 5 and 6 and all the way through section 5 the analysis was performed for HOI. A separate discussion comparing with papers in NAMBLEX now explicitly extends this to HO₂.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 11643, 2005.

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