

4.051 A global modelling study of the release of ClNO₂ from tropospheric aerosol and its impact on tropospheric oxidation.

Presenting Author:

Paul Griffiths, Cambridge University, ptg21@cam.ac.uk

Co-Authors:

Alexander T. Archibald, Cambridge University, Cambridge, CAMBS, UK

N. Luke Abraham, Cambridge University, Cambridge, CAMBS, UK

R. Anthony Cox, Cambridge University, Cambridge, CAMBS, UK

John A. Pyle, Cambridge University, Cambridge, CAMBS, UK

Abstract:

Nitrogen oxides play a central role in the chemistry of the atmosphere, affecting levels of both ozone and OH. Heterogeneous removal of the NO_x reservoir, N₂O₅, onto aerosol particles can be a major loss route for NO_x with modelling work by Tie et al. (2003) suggesting that, at high latitudes, N₂O₅ hydrolysis can reduce NO_x levels by as much as 90 %. The reactivity of the aerosol towards N₂O₅ has been shown to be a complex function of ambient temperature and RH as well as aerosol composition.

Following measurements by Osthoff (2008), Thornton and co-workers demonstrated that the presence of chloride ions in the aerosol can release of nitryl chloride, ClNO₂, following uptake of N₂O₅. The night-time chemistry leads to a build-up of nitryl chloride, which can subsequently be photolysed to yield chlorine radicals, an atmospheric oxidant, and NO₂, regenerating NO_x. The yield of ClNO₂ depends on particulate levels of chloride and nitrate, as well as factors controlling initial N₂O₅ uptake. The production of ClNO₂ at the air-sea interface has been studied by Kim et al., and the important role of surface active organics demonstrated by Ryder et al.

We have included these processes in a chemistry-climate mode, the UK Met Office Unified Model, UM/UKCA-MODE, using a parameterised yield of ClNO₂ from N₂O₅ aerosol uptake. In this paper, the performance of the parameterisation is examined and the effect of the halogen chemistry on levels of e.g. ozone and particulate nitrate will be investigated. Comparison with field measurements e.g. Tang et al. will also be made, and the impact of ClNO₂ release on oxidative chemistry in the troposphere quantified.