Atmos. Chem. Phys. Discuss., 2, S1018–S1021, 2002
www.atmos-chem-phys.org/acpd/2/S1018/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "A novel model to predict the physical state of atmospheric $H_2SO_4/NH_3/H_2O$ aerosol particles" *by* C. A. Colberg et al.

Anonymous Referee #1

Received and published: 10 February 2003

This is an important contribution to our understanding of what may control the physical state of aerosols in the atmosphere. The emphasis is placed on free tropospheric conditions, presumably because the authors are aware of the additional complexity of aerosols close to the Earth's surface.

The authors should be a little more careful with their assertion that these results are important for radiative forcing. The forcing applies to changes in the radiative properties since pre-industrial conditions. It is not clear in the introduction whether the different quantities being referred to are truly forcings or just changes in aerosol radiative effect due to changing the state of the entire aerosol column. Also, the bulk of radiative forcing comes from changes in aerosols below 600 mb, which is the focus of this study.

A more thorough description of the state of the art would be helpful. Are the authors

Full Screen / Esc
Print Version
Interactive Discussion
Discussion Paper

sure that the effloresecence/deliquescence cycle has not been included in 3-D air pollution models (such as those from Jacobson and others). Certainly it is a novel study, but it is also something that can be studied in a regional or global Eulerian aerosol model (see detailed comments below).

The description of the procedure is generally thorough and reasonably clear. However, it would be helpful if you could explain early on what you assume to happen when a crystal forms. A sentence like 'In these first simulations we assume only one solid phase to form (the one that first reaches its ERH). We then examine the effect of multiple solid phase'.

The paper gives the overall impression that these calculations are representative of the real atmosphere. In reality, because the atmospheric aerosol is sometimes considerably more complex, some of these results will be only plausible indications of what could happen, while others will be close to being 'universal' statements. It would be helpful to have your own view on the changes that one might expect in the real atmosphere. How might the presence of other major salts, such as NaCl, affect the overall conclusions? These are sensitivity studies which you can't perform, but the reader would appreciate some estimates of likely influence.

Minor points

1st sentence of abstract. I suggest 'The physical state of tropospheric aerosol is largely unknown despite... for the aerosol radiative properties'. And in the 2nd sentence you mix plural and singular aerosols.

At every point you need to decide between 'aerosol', 'aerosols' or 'aerosol particles' which you use interchangeably.

2450.7. Medium-Range

2450.9. THE APSM

2450.26. 'precursors to the terrestrial radiation budget' doesn't make sense - rephrase.

ACPD

2, S1018–S1021, 2002

2451.4. The term 'sulfate aerosols' is not an abbreviation for anything. It may commonly be assumed to imply a mixed aerosol, but 'ammoniated sulfate aerosol' is also commonly used.

2451.18. Suggest 'susceptible' rather than amenable.

2452.11. It isn't true that a Lagrangian model is needed. It can be done with an Eulerian model that carries two tracers (one for solid and one for aqueous).

2452.15. There seem to be missing words here.

2454.3. Use 'by which' in place of 'that'

2454.10. Replace 'In case' with 'If the'.

2455.5-15. The details given about the trap experiment are not central to this paper and could be deleted - you give plenty of references for the experiments. Just retain the table.

2456.15. Remove comma before 'that'.

2456.20. Suggest 'absence' in place of 'lack'.

2459.1 Wording is odd.

2460.2. 'Clearly' is not appropriate. Certainly we wouldn't expect efflorescence at the DRH, but it sounds like you are comparing it with a field observation.

2460.10. Again you assert that a 3-D model can't handle efflorescence/deliquescence. It can, so long as solid particles that are advected don't 'poison' neighbouring grid boxes. They would do this only if they strongly perturbed the water vapour or ammonia gas phase abundances, which doesn't happen. So a model simply has to track the fraction of particles that are in a given state and treat them separately.

2460. Section 3.1. It would be helpful to say WHY letovicite is the dominant phase, referring back to the phase diagrams.

ACPD

2, S1018–S1021, 2002

2460.25. Please caveat this statement by giving again the altitude range and season etc. It's not a universally true statement.

2462.4. Does 5% mean absolute change in DRH etc (e.g., 70-75%), or relative (70-73.5)?

2462.25. An ERH of less than 0% is arrived at by using your 'rule of thumb' 50% subtraction. It is not a theoretical value, but rather a simple empirical extension.

2464.8. Choose a better word than 'massively' - certainly not as massively as the salts you are talking about.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 2449, 2002.