Atmos. Chem. Phys. Discuss., 8, S10597–S10603, 2009
www.atmos-chem-phys-discuss.net/8/S10597/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Technical Note: Formation of airborne ice crystals in a wall independent reactor (WIR) under atmospheric conditions" *by* E. Fries et al.

E. Fries et al.

Received and published: 22 January 2009

Comments on review

Thanks to both Referees for their suggestions. We have added some experimental results of the prototype and computational fluid dynamic (CFD) simulations by Fluent™ software, showing that ice nuclei are activated and will grow to macroscopic ice crystals inside the prototype. Here it was necessary to add Björn Nillius and Ulrich Bundke, who have done this additional work, to the coauthor list.

In the following we want to answer to the referees comments in detail.

Referee 1

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Discussion Paper

Additional laboratory work was done and is added to the paper. Here measurements using the Individual-particle Circular dEpolarization Fast Ice Nuclei DEtectoR (ICE FINDER) demonstrate that the wall independent reactor is working and ice cystals were produced by ice nuclei (IN) activation. These measurements should be part of a paper demonstrating the performance of the WIR instrument.

General comments

Additional measurements using ICE FINDER showed that IN are activated. Moreover additional CFD simulations showed that individual particles were exposed to ice supersaturated conditions all along their trajectories inside the WIR. The principle of WIR using relatively warm, wet walls and a colder aerosol particle is a well known principle that was realized. According to the comment about water vapour distribution inside WIR we have added FLUENT calculations. To confirm ice crystal formation inside the reactor measurements of ICE FINDER were added demonstrating that WIR is working as expected.

Specific comments:

Page 13018, lines 22-23: this comment is correct; the value 2.82 mg is the total mass of the water vapor formed by molecular and convective diffusion per time unit in the reactor. We have changed it in the final paper. Page 13020 lines13-15: the object of this study is presentation of “a new wall independent reactor (WIR) to simulate the formation of airborne ice crystals”. The study of interaction of hydrocarbons with airborne ice crystals by use of the WIR will be possible but it is not the aim of this paper. The interaction of hydrocarbons with ice crystals could be investigated during ice crystal growth in the WIR in the presence of gaseous AH in the air flow (similar to the experiments described in detail by Fries et. al., 2007). The sentence (lines 15-16) has been deleted.

Page 13021, line 5: Our preconditioning unit consists of a two step procedure. In a fist step a cold trap at -10 °C removes most of the water vapor. In a second step a 5

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m long helical flexible aluminum vent inside the chamber at -20 °C operates as a cold trap and keeps the probe flow at 100% rel. humidity with respect to ice. This second cold trap is very effective. A Fluent calculation of the second cold trap shows that after 40 cm excess water vapor is completely removed and relative humidity remains at saturation condition at the particular wall temperature. We introduce the two step procedure because the first step cold trap can be easily deiced in contrast to the second cold trap inside the chamber. Page 13021, lines 10-11: the aerosol probe was generated using bypass air taken out of the conditioned (dew point -20°C) flow. Page 13021, lines 12-13: we have changed this misleading formulation. The sentence (Page 13023, line 11-12): it has been improved in the following way: Therefore the amount of water vapour and it transport in the reactor is characterized by molecular and convective diffusion at the same time and should not limit the vapour condensation. The distribution of water vapour concentration was calculated from the measured temperatures at a relative humidity of 100% with respect to water across the profile of the reactor. The accumulated amount of water vapour formed in the reactor is given by Eq. (4)

Page 13024, lines 6-8: the Equation (6) is pure mathematical addition. The concentration and temperature gradients inside the WIR were inconstant. The reactor can be divided into several small elements wherein the gradients were constant. The amount of water vapour of such element can be calculated by Nernst equation (Levich, V.: Physicochemical Hydrodynamics, 1962) (1) QM is water vapour formation rate (mg/s); lm is the thickness of the molecular diffusion layer, which can be calculated by Einstein equation (2) where td is diffusion time (s) and can be defined as the quotient of reactor height (h,m) and air flow velocity (m.s-1) in the WIR. (3) Replacing the lm in the Eq. (1) using Eq. (3) we get (4) Ash , (m2) is the inner shell surface of the considered section given by its radius R (m) and its height h (m), (5) Substituting Eq (5) into Eq (4) and further simplifying it, we get

(6) The equation (8) was suggested by Reist for calculation of supersaturation (Introduction to Aerosol Science, Macmillan Co., (1984).) The value of Csat is the concen-

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tration of water vapour at saturated conditions (100% relative humidity) with respect to water at measured temperatures over cross-section in the different chamber’s depth. The value of C is water vapour concentration in the air flow calculated basing on the water vapour mass formed in the WIR by molecular diffusion and transported by convection per time unit in the different chamber’s depth (the 5th column in the Table 1). We have clarified it in the final paper. We have changed the sentence (page 13026 lines 20-21) to: We suppose that this residence time under experimental conditions including air flow rate, the inlet and outlet temperatures, water vapour mass formed in the WIR, is sufficient for the activation of ice nuclei inside the chamber. To the reference (page 13026, lines 21-24): In this paper, laboratory experiments are presented to investigate the uptake of AH by ice crystals during vapour depositional growth. Inside a walk-in cold chamber, ice crystals were grown from an ambient airflow in the presence of gaseous AH. With this approach, falling ice crystals were simulated during crystal growth. Experimentally defined ice growth rates are compared to predicted ones. Selected compounds for the uptake experiments at different temperatures were benzene, and alkylated benzenes, namely: benzene (C6H6), toluene (methylbenzene, C7H8), the C8H10 isomers ethylbenzene, o-, m-, p-xylene (dimethylbenzenes), the C9H12 isomers n-propylbenzene, 4-ethyltoluene, 1,3,5-trimethylbenzene (1,3,5-TMB), 1,2,4-TMB, 1,2,3-trimethylbenzene (1,2,3-TMB), and the C10H14 compound tert.-butylbenzene. Concentrations of AH were measured in ice samples by solidphase micro-extraction (SPME), followed by gas chromatography/mass spectrometry (GC/MS) and uptake coefficients were determined

Page 13037 and 13038: the small asymmetry seen in the plots is a measurement artifact. Scans of the temperature were made by moving the temperature sensor on n horizontal planes. Thus the observed ‘asymmetry’ may result out of a long term temperature drift during the scans. We have added a velocity profile contour plot using FLUENT calculations (see Fig). Page 13039, Fig 3: measurement errors are added in the text. Page 13042, Fig 6: this figure demonstrates the stability of the aerosol generation which is necessary to get stable conditions for airborne ice

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formation.

Technical comments are corrected.

Referee 2

We added computational fluid dynamic calculation using FLUENT™. These calculations are in accordance with analytical calculations. Also measurements using ICE FINDER were added detecting ice crystals on singe particle basis. These measurements showed that WIR is working like expected.

General remarks:

Page 13020, line 6: We have specified these wall effects in the final paper. Page 13020, line 16: We can not distinguish between both effects. But former experiments to study the adsorption of organic material on growing ice crystals show almost no scavenging efficiency on “pure “ ice crystals during growth (see Fries et. al., 2007)

Experimental setup:

Page 13020, lines 21-22: Inlet and outlet of the chamber have to be held constant at cold temperatures: a) to generate the cold aerosol probe and b) to avoid melting of grown crystals for further analysis. This is now clarified in the text.

Page 13021, lines 1-4: this comment is correct. Here only the filter efficiency can be described. The efficiency for the removal of organics is part of future lab work. In the text we have clarified the function of the cleaning device for future applications

With respect to humidity profile within the reactor: the relative humidity has to be related to the actual measured temperatures of the air flow over the cross sections along the WIR. We have averaged the seven temperature values, which we have measured in the radial plane excepting the wall temperature at distance 0.15 m from reactor center. Therefore the vapour concentrations across the diameter should be not changed

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significantly. Moreover by adding the FLUENT™ calculations the situation now is much clearer to the reader. The conditions along the trajectories of particles in the inlet air stream are presented in form of diagram (Relative Humidity with respect to ice along 18 individual particle tracks starting all over the cross section of the aerosol inlet as function of the path length in the WIR) which is added in the final paper. The comment to the value Qmi is correct. That gives a mass of the water vapor formed by molecular diffusion in the reactor and can represent a potential ice grow rate. It has been changed in the final paper. Fluent calculations were added. A) Fluent calculations showed low amounts of turbulence. Residence times were also computed. B) DeMott has published transfer times in the order of 10-30 sec within his CDFC chamber. Our residence times were in the same order. Because we now have the experimental proof we can compare the residence timescales.

Measurements of ice crystal number concentration using ICE FINDER (Bundke et al 2008) were added. Measurements were in accordance with fluent calculations.

D-Model calculation of WIR

We have performed model calculations using the computational flow dynamic software FLUENT ™ . Using a 3D Model of the actual WIR geometry steady states of temperature and relative humidity profiles were calculated using model equations for laminar flow. Along 18 individual calculated tracks of 2μm particles, starting from all over the cross section of the inlet, the relative humidity is plotted as function of flight time inside WIR. Fig. 7 demonstrates that the maximum supersaturation seen by individual particles at the particular conditions (aerosol sample flow=30 l/min, aerosol temperature= 253 K, wall temperature= 268 K) reached149 % ś 2% RHi and remained complete above ice saturation inside the reactor. In Fig 2a and 2b the velocity field and the Reynolds number field is plotted on two orthogonal cross section to get a 3-D impression of flow conditions of the inlet section. The model calculations are in accordance with temperature measurements shown in this paper.

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First Test Experiments of WIR

Main aim of the initial experiments was to show that inside the prototype WIR instrument ice nuclei activation takes place. Therefore we use the Ice detector ICE FINDER (Individual particle Circular dEpolarization Fast Ice Nucleus DEtectoR) of the Ice nuclei counter FINCH (Fast Ice Nulceus Chamber) described in Bundke et al., 2008. In summary this detector measures the circular depolarization ratio of light scattered by individual particles, larger 3μm diameter. With ICE FINDER it is possible to count single particles and distinguish between super cooled droplets and ice particles activated inside WIR. Circular depolarization of scattered light is independent of particle orientation and multiple scattering. Thus, this principle of ice detection is at the moment the most robust measurement technique available. In Fig. 8 the relative frequency of occurrence as function of the normalized circular depolarization ratio P44/P11 is plotted. Here P44 and P11 are elements of the scattering matrix of the individual particles. The depolarization ratio was measured, integrated in the scattering angle range of 100-130 degrees. Water droplets are observed in this plot close to zero depolarization (P44/P11=0 ś0.2. In the range of P44/P11 between 0.2 and 0.4, large not activated particles can be found. Depolarization ratios larger 0.4 are associated with ice particles. The ICE FINDER software regularly removes not activated counts using scattering intensity as a second criterion using the circumstance that ice crystals are much larger than not activated particles. This optional function was deactivated during these initial tests.

Fig.5. Relative frequency of occurrence as function of the depolarization ratio P44/P11. Ice particles are seen for ratios P44/P11 $>$ 0.4.

These initial tests demonstrate that WIR is operating as expected.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 13017, 2008.

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