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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observations of meteoritic material and implications for aerosol nucleation in the winter Arctic lower stratosphere derived from in situ particle measurements" *by* J. Curtius et al.

J. Curtius et al.

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We are very thankful for the thoughtful comments by the reviewers and by J. Renard. Our replies to the comments are given below. Text changes compared to the ACPD version are also indicated. We include the original comments (italics), our replies (normal font) and the text changes in the manuscript (bold). We hope, with these changes the manuscript is acceptable for publication in ACP.

Referee comment by reviewer 1:

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Curtius et al. describe here observations near and within the polar vortex of aerosol size and volatility from the Geophysica aircraft. These observations appear to indicate a high fraction of meteoritic material, consistent with previous studies. The quality of the data and the authors? insight are excellent and this is a well written and interesting paper. There are only minor issues that should be corrected.

Page 5044 Line 22: "the cut-off is slightly": Please quantify "slightly".

We added the following text to quantify the slight pressure-dependence:

In our laboratory experiments we found the cut-off for the COPAS instrument when operating with butanol and detecting sulfuric acid-water particles to be 9.7±1.6 at 70 hPa, 8.1±1.7 nm at 120 hPa and 5.8±2.0 nm at 200 hPa for one channel and 9.5±2.2 nm at 120 hPa and 7.4±2.2 nm at 200 hPa for the other channel.

Page 5046 Line 20: "the two instruments agreed within 20% or better.": This topic could use a little more discussion. Specifically the previous fits would seem to suggest a very close agreement, within a few %, and yet here the authors state 20%. This is directly related to credibility of the data. Is the fit normally better and 20% at worst?

To be more precise, we replaced the sentence "Generally the two instruments agreed within 20% or better" by:

We include the 95% prediction interval in Figure 1. For Figure 1a a pair of data points falls with 95% probability in an interval ±14% around the regression line, and for Fig. 1b the 95% prediction interval is ±22%, respectively.

The points scatter in a normal distributed way around the fit line and of course most of the data is much closer to the fit line than the 95% prediction line. Therefore, the data agree indeed normally much better than 20%.

Page 5054 Lines9-15: "f can be utilized as a directly measurable vortex tracer." Although f appears to be useable as a vortex tracer this statement should be qualified in that it goes a bit too far. It is clear from Figure 6 that f would be a very noisy tracer

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and is not of the quality of traditional tracers. See also the discussion just before and within the Summary: a discussion of other refractory particles near the polar vortex (Baumgardner) would seem to suggest the utility of this "tracer" would be limited in many situations.

Yes, we agree, we added some caveats and additional explanations to Page 5054 Lines 9-15 and to the discussion in the Summary (see also comment on this issue by D. Murphy):

(p. 5054 Lines 9-15):

Within limits, f can be viewed as a directly measurable vortex tracer, but because the correlation with the calculated vortex tracer has a substantial scatter, f does not allow a vortex identification as precise as identification from traditional correlations of long-lived gaseous tracers. We found the ratio f to be better suitable as a vortex tracer than the concentration or mixing ratio of the non-volatile particles alone. For the total particle concentration as well as for the non-volatile concentration a distinct dependence on the vertical coordinate (altitude, potential temperature, N_20, etc.) exists. For example, measured values of the mixing ratio of non-volatile particles outside the vortex at low potential temperatures can be identical to mixing ratios inside the vortex at higher potential temperatures (cf. Fig. 4c) and therefore a vortex tracer is more difficult to discern. For the ratio f the dependence on the vertical coordinate apparently cancels out and the simple correlation is found. Some of the scatter might be caused by variable numbers of volatile particles (e.g. enhancement due to mixing with air containing newly formed volatile particles from the tropical tropopause region), or to the fact that non-volatile particles are not necessarily all of meteoric origin (e.g. particles such as soot can influence the measurement of the non-volatile particles).

Summary (p.5060, l.24):

The fraction of non-volatile residual particles also serves as an experimentally accessible estimate of a vortex tracer. Besides meteoric smoke, particles of different nature such as light absorbing soot particles (Baumgardner et al., 2002), may influence the measurement of f and therefore limit the use of f as a vortex tracer.

Page 5055 Line 18: "Both size distributions peak near the smallest channel " It would appear this is a peak related to instrument performance and not necessarily the physical size distribution. Please clarify.

Yes, we agree, we added (Page 5055 Line 18):

but these peaks are likely to be caused by an instrumental under estimation of the particles in the lowest channel. The peaks therefore probably do not represent a maximum of the aerosol size distribution.

Editorial:

Page 5047 Line 4: This sentence needs to be rewritten. It should be "In this way " and eliminate "anymore". Rfi should be defined.

corrected

Referee comment by John Plane:

This paper describes a series of high altitude flights through the Arctic polar vortex. Particles greater than 10 nm (diameter) were measured using several low pressure condensation counters, including one with a heated inlet channel to evaporate volatile particles. Chemical tracers (N2O and CFC-11) were also measured. The particle measurements are analysed through a series of correlations - with potential temperature, chemical tracer mixing ratio, and "vortex tracer" index. The major conclusion is that there is a source of particles from the upper stratosphere. Because these particles contain a non-volatile core, the authors conclude that these are meteoric smoke particles. There is evidence that these provide nuclei for the heterogeneous condensation *of H_2O and H_2SO_4, which in turn could have potentially important effects on ozone in the lower stratosphere inside the polar vortex. The relative absence of these non-volatile particles outside the polar vortex also implies that most of the meteoric smoke is swept by the meridional circulation in the mesosphere to the winter pole, before descending in the vortex. One recent reference that should be discussed in the paper is Gabrielli et al., Meteoric smoke fallout over the Holocene revealed by iridium and platinum in Greenland ice, Nature, 432, 1011-1014, 2004. This study showed that the flux of meteoric smoke in Greenland snow is about 5 times larger than expected if the downward flux of meteor debris is uniform over the earth, a further illustration of the focusing effect of the meridional circulation. In order for the particles to avoid sedimenting out of the mesosphere within the 2 - 4 weeks that it takes to transport them to the polar vortex, the particles must be smaller than about 4 nm in diameter. A new meteoric smoke model presented by Gabrielli et al., which updated the seminal paper by Hunten and co-workers from 1980, shows that this should indeed be the case if the global daily input of interplanetary dust particles into the atmosphere is 50 tonnes or less. However, an interesting problem remains: will these very small particles coagulate sufficiently quickly during their descent in the vortex to reach a size greater than 10 nm (and thus be detectable) in the lower stratosphere? The paper is a very clearly written account of an important study. It contains the level of detail required to satisfy the reader that the performance of the particle counters was carefully checked. I therefore recommend publication in ACP after consideration of the (minor) points listed below:*

John Plane raises several interesting points here. The question about the size of the non-volatile cores is certainly important. His argumentation questions whether the meteoric smoke particles coagulate sufficiently quickly while being transported downward in the vortex to reach a size of about 10 nm detectable with our instrument. On the other hand, Dan Murphy argues in his review (see below) that the meteoric cores should have a size of about 80 nm to be consistent with current estimates of the global meteor flux. An exhaustive solution to this apparent contradiction is -although very important and interesting- beyond the scope of this paper. The CN-Counter measurements give no corroborated evidence of the non-volatile particle size, except that these particles have to be larger than the lower cut-off diameter of around 10 nm. Future experimental studies should certainly address a measurement of the size distribution of the non-volatile particles (but unfortunately currently no instrumentation exists for airborne high-altitude measurements of the size distribution of 2-200 nm particles, neither for the total nor for the non-volatile particles). Furthermore, detailed modelling of the coagulation processes and the concurrent uptake of gaseous sulphuric acid by the meteoric smoke particles are needed to explain our measurements. Generally, a coagulation of several hundred meteoric smoke particles cm-3 of 1-5 nm particles at 60 km altitude (0.2 hPa) as given by Gabrielli et al., 2004, to about 10 particles cm-3 of sizes larger than 10 nm at 20 km altitude (p = 55 hPa) within the transport times of months is reasonable, but detailed modelling including growth of the particles by uptake of gaseous sulphuric acid and water and coagulation with the stratospheric sulphuric acid water aerosol distribution should be performed in the future.

We added the following text (p.5053, l.26):

Recent calculations with a one-dimensional model describe the development of the meteoric smoke particles in the mesosphere including processes of formation, coagulation, condensation and gravitational settling (Gabrielli et al., 2004). At 60 km altitude particle concentrations of $\sim$700 cm^{-3} for particles of 0.4 nm size to $\sim$20 cm^{-3} for particles $>$4 nm are modelled.

and (p.5054, l.8):

A remaining question is whether the meteoric smoke particles coagulate fast enough with each other during their downward transport in the Arctic vortex to reach sizes larger than $\sim$10 nm to be detectable by our CN-counter. A detailed modelling of the coagulation of the meteoric smoke particles with each other, simultaneous coagulation with stratospheric sulfuric acid-water aerosol and condensation of gaseous sulfuric acid and water vapor is beyond the scope

of this study, but the time scales of several month are reasonable for at least a fraction of the meteoric smoke particles to reach sizes of the non-volatile cores of $>$10 nm.

The second point is the focusing effect of the meridional circulation and the enhancement of meteoric debris found in the Greenland ice core (Gabrielli et al, 2004). Once the particles are released from the Arctic vortex in the lower stratosphere, it is not entirely clear to us that there is a direct connection to deposition of these particles in the Arctic ice. Even if the meteoric smoke particles leave the lower stratosphere by stratosphere-troposphere exchange at high latitudes they will most likely be transported by isentropic transport to mid latitudes. There should then be some enhancement in the extra-tropics compared to the tropics, but it is not clear to us whether really a factor 5 enhancement in the Arctic ice can be expected. Furthermore, a fraction of the particles is removed from the atmosphere by precipitation and it would have to be studied in how far the proposed enhancement in the ice core is influenced by wet deposition of the particles at the Greenland site compared to the extra-tropical average during the Holocene (but from precipitation maps one might rather expect a depletion than an enhancement for Greenland?). Nevertheless, the Gabrielli et al. study as well as our study both support the hypothesis that the deposition of meteoric smoke particles is not uniform over the entire surface of the Earth but is expected to be enhanced in the extra tropics due to the focusing effect of the mesospheric meridional circulation and the downward transport in the stratosphere through the Arctic vortex.

We added the following text (p.5053, l.18):

Gabrielli et al. (2004) study iridium and platinum concentrations in Greenland ice cores. A major fraction of the Ir and Pt is found to have an extraterrestrial origin. Due to the focusing effect of the meridional circulation in the mesosphere and the downward transport of meteoric smoke particles in the polar vortex an enhancement of cosmic material deposited at the Earth surface poleward of 55° latitude is postulated, in comparison to the global average assuming a uniform

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influx. Our conclusion of the enhanced non-volatile material in the vortex being meteoric smoke particles therefore supports these findings, and vice versa our findings are supported by the Gabrielli et al. (2004) study.

1. The terms meteoroid, meteor and meteoritic are quite specific. A meteoroid is a dust particle entering the atmosphere; this may produce an optical or radio signal, the meteor; and if it survives entry and reaches the ground, it becomes a meteorite. Hence, the use of "meteoritic" in numerous places in the paper (including the title) is not strictly correct. Refer instead to "meteoric" or "meteor" smoke particles and debris. On page 5042, l. 25, use "meteoroids", not "meteorites".

We changed the term "meteoritic" to "meteoric" in all instances and changed "meteorites" to "meteoroids".

2. The Gabrielli et al. reference should be included.

See above.

3. page 5045, l. 2 "the present volcanically quiescent period".

corrected

4. page 5051, l. 24. The sentence starting "Additionally " needs clarification. What does "temporal" mean here?

We replaced the sentence by:

Additionally, Wilson et al. (1990) observed a shift in the correlation with time: In February average CN mixing ratios were measured to be ∼30% higher than in January for corresponding N_2O. Such a shift in the N_2O-particle correlation was not observed in our measurements.

Referee comment by Dan Murphy:

This paper presents valuable observations of the volatility of particles in the lower polar

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stratosphere. The discussion is solid and well-referenced. I have some suggestions for improving the clarity and extending the discussion:

- Figure 2 could be eliminated. Figure 3 shows the same data in a more useful format.

We agree that the information is somewhat redundant but we prefer to include Figure 2 for two reasons: the non-expert reader might have a better comprehension of the context of the measurements when the data are also displayed as a function of altitude and not only potential temperature. Secondly, the color coding by flights allows the reader to get an impression of the flight-by-flight variability (temporal variability). Especially for the discussion of the variability of the particle concentration in the lowermost stratosphere (section 3.4) this might be helpful.

- Figure 8 would be more informative if it showed particles mg-1 rather than cm-3. The former is analogous to mixing ratio and is the more conserved quantity. There are systematic changes in air density with potential vorticity that make it difficult to understand what is causing the relationships shown in this figure using cm-3.

We changed Figure 8, now displaying particles mg-1. Text and Figure caption were adjusted accordingly.

- In the discussion, the authors suggest that the fraction of nonvolatile particles is a tracer of vortex air. There is a tracer there, but should one use the fraction of nonvolatile particles or the mixing ratio of nonvolatile particles? The question is which quantity is more quantitative about mixing vortex air with low-latitude air that might contain variable numbers of new (volatile) particles from the tropical tropopause. Some critical discussion of which is the better tracer would strengthen the paper.

We agree that the ratio might be influenced by a variable number of volatile particles, in fact this might be a reason for the observed scatter in the correlation, but the mixing ratio of non-volatile particles is not as suitable as a tracer because absolute mixing ratios outside the vortex at a lower potential temperature/altitude are just as high as mixing ratios inside the vortex at higher potential temperatures/altitudes (cf. new Fig. 4c, see below). Because of this additional dependence a correlation with the vortex tracer is more difficult to discern than for the ratio f where the altitude dependence apparently cancels out. For the text changes in the manuscript see above (comment by reviewer 1).

- To follow this, I?d like to see added to Figure 4 a separate panel showing non-volatile particles as a function of N2O.

We included the requested panel (Fig. 4c) showing the non-volatile particles mg-1 as a function of N2O.

- Figure 4a allows some rough estimates of the source strength of meteoritic smoke particles. Combining Figure 4a and Figure 5, it appears that below 200 ppbv of N2O there is a slope of about 1 particle mg-1 per ppbv of N2O. If this is representative of the stratosphere, then one could multiply this slope times the global sink of N2O to get a source strength (Murphy and Fahey, JGR, 1994). Using a stratospheric sink of N2O, the global high altitude source of non-volatile particles would be about 2e25 per year. The authors can do better calculation from their actual data than my eyeball fit. Inserting the lower limit diameter from the manuscript (26 nm) gives a lower limit global incoming meteor flux of 0.4 Gg per year, with some uncertainty due to the density of the particles. A more realistic diameter of about 80 nm for the meteoritic cores gives an annual flux of order 10 Gg per year. This is consistent with independent estimates of the global flux of meteors and is further support for the authors? contention that the non-volatile cores are meteoritic material.

We included a regression line for the N2O non-volatile particle correlation for N2O smaller than 200 ppbv (Fig. 4c). Indeed a slope of 0.8 particles mg-1 per ppbv of N2O is observed. We are reluctant to derive an estimate of the global meteor flux based on this slope because a) we are not certain whether this correlation observed inside the vortex in one winter is representative for the global average or at least for the

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Northern hemisphere. The meridional differences in the influx from the mesosphere, for example, might limit the concept. b) The assumption of a mean particle size of about 80 nm seems fairly large, especially with respect to the above comment by John Plane, claiming that the particles from the mesosphere should be rather small.

As stated before, the measurement of the size, shape and density of the meteoric inclusions would be an important subject of future research.

We added (p.5052, L.7):

The mixing ratio of non-volatile particles as a function of N_2O is shown in Fig. 4c for comparison. Similar to the total particles, the non-volatile particles increase markedly inside the vortex with decreasing N_2O for N_2O $<$200 ppbv. A detailed discussion of this correlation will be given in the following section.

and p.5054, l.8:

In principle, the slope of the N_2O-non-volatile particle correlation for N_2O $<$200 ppbv (cf. Fig. 4c) could be used to obtain a global source strength of the extraterrestrial influx through meteoric smoke particles (Murphy and Fahey, 1994). We did not apply this concept here as we do not know in how far the observed correlation from inside the polar vortex is representative for the stratosphere in general (e.g. meridionally inhomogeneous influx from the mesosphere could change the correlation at lower latitudes) and because we do not know about the average size and density of the meteoric smoke particles from our measurements which would be necessary to determine the extraterrestrial mass influx.

Interactive Comment by J. Renard:

The paper presents new and interesting results for the presence of meteoretic dust in the stratosphere. Two (minor) comments could be taking into account by the authors : 1) The detection of such material, at least for the larger grains, have been performed by our team using ballon-borne counter and UV-visible spectrometer (Renard et al.

Applied Optics vol44 N 19 4086-4095, 200). 2) The satellite data should be used cautiously, since they can exhibits large bias, in particular using priori hypothesis (log-normal size distribution, refractive index) as we have discussed in Renard et al. Applied optics vol41 N 36, 7540-7549, 2002.

We included both references and discussed them:

p.5041, l.19:

Renard et al. (2005) report balloon-borne stratospheric extinction measurements. At ∼30 km altitude unexpected spectral structures around 640 nm wavelength were interpreted as signals from large particles (larger than several hundred nanometers in size) of most likely extraterrestrial origin. Recently, Baumgardner et al. (2004) reported large fractions of light absorbing particles such as soot in the lowermost Arctic stratosphere at altitudes of 9–12 km.

p.5043, l.19:

Note that the results from satellite observations are potentially biased as some a priori hypotheses about the shape of the aerosol distribution and the refractive indices have to be included (Renard et al., 2002).

Besides the clarifications and additions requested by the referee reports and the interactive comment, we added a paragraph about the ACPD paper by Engel et al., published online in August 2005.

p.5054, l.8:

Very recently, Engel et al. (2005) published balloon-borne tracer observations (SF_6, CO, CO_2, etc.) and model calculations describing the transport of mesospheric air in the Arctic vortex in the winter 2002/2003. They find comprehensive evidence for the presence of mesospheric air in the polar vortex. Interestingly,

the largest fraction of mesospheric air is found in a layered structure descending from ~30 km altitude in January to ~22 km altitude in March. This observation of a distinct layer of mesospheric air might also explain the CN layers observed by Hofmann (1988, 1990) in the polar vortex to be in fact layers of meteoric smoke particles, rather than freshly nucleated sulfuric acid water particles. Furthermore, the results by Engel et al. support our hypothesis that meteoric smoke particles of mesospheric origin cause the observed enhancement of the non-volatile particle fraction in the Arctic vortex. Due to additional gravitational settling, the meteoric smoke particles, at least the largest ones, are potentially transported downward in the vortex even faster, reaching lower altitudes earlier than the corresponding mesospheric gaseous compounds.

Engel, A., Möbius, T., Haase, H.-P., Bönisch, H., Wetter, T., Schmidt, U., Levin, I., Reddmann, T., Oelhaf, H., Wetzel, G., Grunow, K., Huret, N., Pirre, M.: On the observation of mesospheric air inside the arctic stratospheric polar vortex in early 2003, Atmos. Chem. Phys. Discuss., 5, 7457-7496, 2005.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 5039, 2005.