

Answer to Reviewer # 1

The authors thank the reviewer for his pertinent and helpful comments on the paper, and they are grateful for this review work which is always rather time-consuming and cumbersome. The manuscript has been significantly modified according to the suggestions proposed by the reviewer but also from several discussions and iterations between the co-authors. The microphysical and optical properties of the primary and secondary wakes are now presented separately. These modifications have changed the manuscript mainly for clarity objectives. The remainder is devoted to the specific response item-by-item of the reviewer's comments :

1. Introduction

I think that a more clear description is needed of where this study fits with previously published results. In particular, although a brief description is given of other studies, it is never directly discussed why more measurements are needed or why the current study adds anything to the body of knowledge on contrail science. It seems to me to be a major oversight to not use the BAMS special issue on contrails as the basis for explaining why more information is needed on contrail particle formation and evolution. Without this type of discussion in the introduction, as well as in the concluding remarks, the reader is left wondering how these measurements advance the science.

As suggested by the reviewer, the introduction gives a better description on the paper objectives and now discusses why more measurements are needed or why the current study adds something to the body of knowledge on contrail science and refers to the BAMS special issue on contrails. The introduction has been re-written, see new manuscript:

2.1 Contrail particle probes

First of all, the title of the section is odd as it reads as if the particle probes are part of the contrail. Suggest removing the adjective “Contrail” and just entitle the section “Particle Probes”.

Done.

Secondly, I recommend that the term “Optical equivalent diameter” be used instead of “diameter” to make it quite clear the type of size parameter being defined. This can be clarified farther on when discussing the use of T-matrix to redefine the size bins. It is probably OK to then use “diameter” from this point on once there is no ambiguity in its usage.

Two diameters are discussed in the manuscript. The first one addresses the size-bins of the FSSP-300. Basically the FSSP measures scattering energy at forward angles which is governed mainly by diffraction and therefore depends on the projected area of the particle. Indeed, the size calibration for aspherical particles (see below) is expressed in terms of equivalent surface diameter, i.e. the diameter of a sphere that has the same projected area (see Mishchenko et al., 1997). In order to characterize the particle size distribution properties, we

define the effective diameter because that is the ‘optical equivalent diameter’ commonly used to assess cloud (contrail) optical properties as discussed by McFarquhar and Heymsfield (1998) and Schumann et al. (2011a). Here the effective diameter represents the volume to the surface ratio (see below) for a given size distribution (bin-scaled to either spherical ice particles or aspherical particles). (References as in the paper)

In lines 92 and 93, it is more correct to say that the sampling volume is defined by the cross sectional area of the beam multiplied by the distance traveled during the sample period.

Done.

I am completely mystified by the explanation in the section encompassed by lines 114-129. If the way in which the size channels are grouped is essential to the interpretation of the data then a much better description of what is being done is needed here. The relationship between Mie ambiguities and particle shape is not at all clear to me. I am fairly well versed in the Mie ambiguities with respect to spherical particles, refractive index, size and collection angle but don’t understand what the authors mean when they talk about shape information from the PN. The FSSP-300 is a 30 channel instrument so when the discussion is about 8 and 9 channel rebinning with different upper thresholds I am unable to decipher their meaning. I am supposed to know these things so if I can’t unravel the meaning I doubt other will either. When calculating extinction coefficients from the FSSP-300 size distributions are you assuming spherical ice crystals? What type of uncertainty is associated with these derived values?

We initially planed to discuss this issue in a forthcoming paper. Now, **Appendix A** has been added to the manuscript in order to explain how we combined FSSP-300 and Polar Nephelometer data to assess the size-bin calibration for spherical and aspherical ice particles.

2.2 Trace Gas Instruments

Line 168, what is a PFA?

PFA is the inlet material, i.e. a fluoropolymer. Now explained

Observations Why is effective diameter being used rather than some more definitive parameter to track particle growth like median volume diameter? If effective diameter is going to be used, then define how it is calculated and the estimated uncertainties. The usual definition has it proportional to the ratio of liquid water content to extinction, both are parameters that are likely to have large uncertainties for aspherical particles.

See section 2.1 above.

Lines 224-228: Why is it surprising that NOy and ice crystal concentration are not correlated. It seems to me that their production and removal processes and time scales are quite different. I am uncomfortable with trying to explain the lack of correlation with plume inhomogeneities. I think a discussion of the physio-chemical processes involved with the crystals and gases is needed in order to explain the lack of correlation. Using plume inhomogeneities is rather frail, in my opinion.

There are different physical and dynamical processes working on gaseous species and on the particles in the aircraft wake (such as phase change and sedimentation), hence it is not surprising that high NO_y concentrations indicative for the primary vortex are not fully correlated with high particle concentrations or IWC. The NO_y data are highly correlated with trace gas observations of SO₂ and HONO in the A380 contrail (Jurkat et al., 2011). Except for the first encounter at 12:15:37 UT, the primary vortex sequences with high NO_y mixing ratios are not correlated with the particle concentration or the IWC. The complex vortex dynamics acting on gaseous species and particles, the entrainment of ambient air as well as particle inertia could explain a separation of particles and trace gas fields in the primary vortices and the secondary wake of the aircraft exhaust. Most contrail ice particles may form at the outer edge of exhaust plumes while emission trace gases such as NO_y are concentrated inside the exhaust plume (Petzold et al., 1997).

Line 274 – What does “reliable values” mean? Do you mean, if we assume that these are reliable values? In the previous sentence you say these values can’t be representative. Why not just compute averages and standard deviations ?

As underscored in the revised manuscript we do not have enough data to calculate representative averages and standard deviations in the primary vortex of the contrail. Only maximum values are given. The small scale heterogeneities of the vortex properties is of the order of the horizontal distances of the probe mounting positions on the Falcon (5-10 m). This hampered the representativeness of the measurements and the derived mean contrail properties. For instance the transient peak of extinction (7.0 km⁻¹) measured by the Polar Nephelometer in the primary vortex is not correlated with the extinction derived from the FSSP-300 as shown on Fig. A1 (surrounded data). Because a representative data set has been obtained in the secondary wake (70 to 220 s contrail age) at a quasi-constant altitude near the A380 cruising altitude (10600 – 10700m), the mean microphysical properties are now indicated.

Line 277 – What does it mean to say “maximum values from 340-360, for example? Where are maximum values being defined?

See above.

Line 279 – Reiterating my previous question about the use of effective diameter instead of a more physically relevant diameter, if the purpose is to examine particle growth, what is the justification for using effective diameter?

We agree, the mean volume diameter should be a more physically relevant parameter to examine the particle growth rather than the effective diameter. Nevertheless, there is a close relationship between these two parameters as displayed on Fig. A2. Indeed, *Deff* increases with *MVD* over a smaller range of variation, but *Deff* well describes the growth of the particles in this case study.

Lines 309-324 - I am rather confused by the discussion in this section. There might be a problem of grammar use but it is not clear to me what is being discussed here. The measurements indicate quasi-spherical particles and it seems that the objective here is to explain why they are quasi spherical. It is my understanding that ice formation in contrails is likely a result of homogeneous freezing of water droplets. If that is the case, and if the sampling is only a few hundred seconds after contrail formation, why can’t these still be

spherical ice? There are lab studies (Golina et al., for example) that show that frozen droplet remain spherical for up to 20 minutes after they freeze. Why does sublimation need to be used here to explain the spherical shape?

This part has been rephrased (see below) and is discussed in the introduction.

Line 364 – Why is a refractive index of 1.29 the starting point? I was under the impression that 1.31 is the refractive index for ice, at least at the wavelength of the PN.

We recall that the size distribution was retrieved along with other parameters, i.e., the refractive index and the partitioning ratio. In particular, the real part of the refractive index of particles was found to be of 1.31 ± 0.002 , that is, the refractive index of ice at the wavelength of the PN. Generally, when a parameter is to retrieve, the inverse problem is solved over an interval that is larger for sure than the domain of a priori expected values of the parameter. In other words, the expected value cannot be a bound of the retrieval interval. The starting point of 1.29 satisfies this condition for the expected value of 1.31.

Line 377 – Given that laboratory and theoretical studies have shown that small water droplets freeze slower than larger, how robust is this assumption of ice fraction the same over all sizes.

The information content of the PN data is inadequate to retrieve a size-dependent partitioning ratio. In other words, variations of the shape and/or the refractive index (from 1.31 to 1.33) of small particles lead to variations of a phase function that are lower than the PN measurement errors. That is why the partitioning ratio is assumed to be constant over the full size range. And, this assumption is robust from optical point of view.

Line 389 - Please expand on this derivation. It seems that this is a circular calculation and will always provide an excellent fit? You get size distribution from the phase function but derive phase function from the derived size distribution? This needs a lot more clarification.

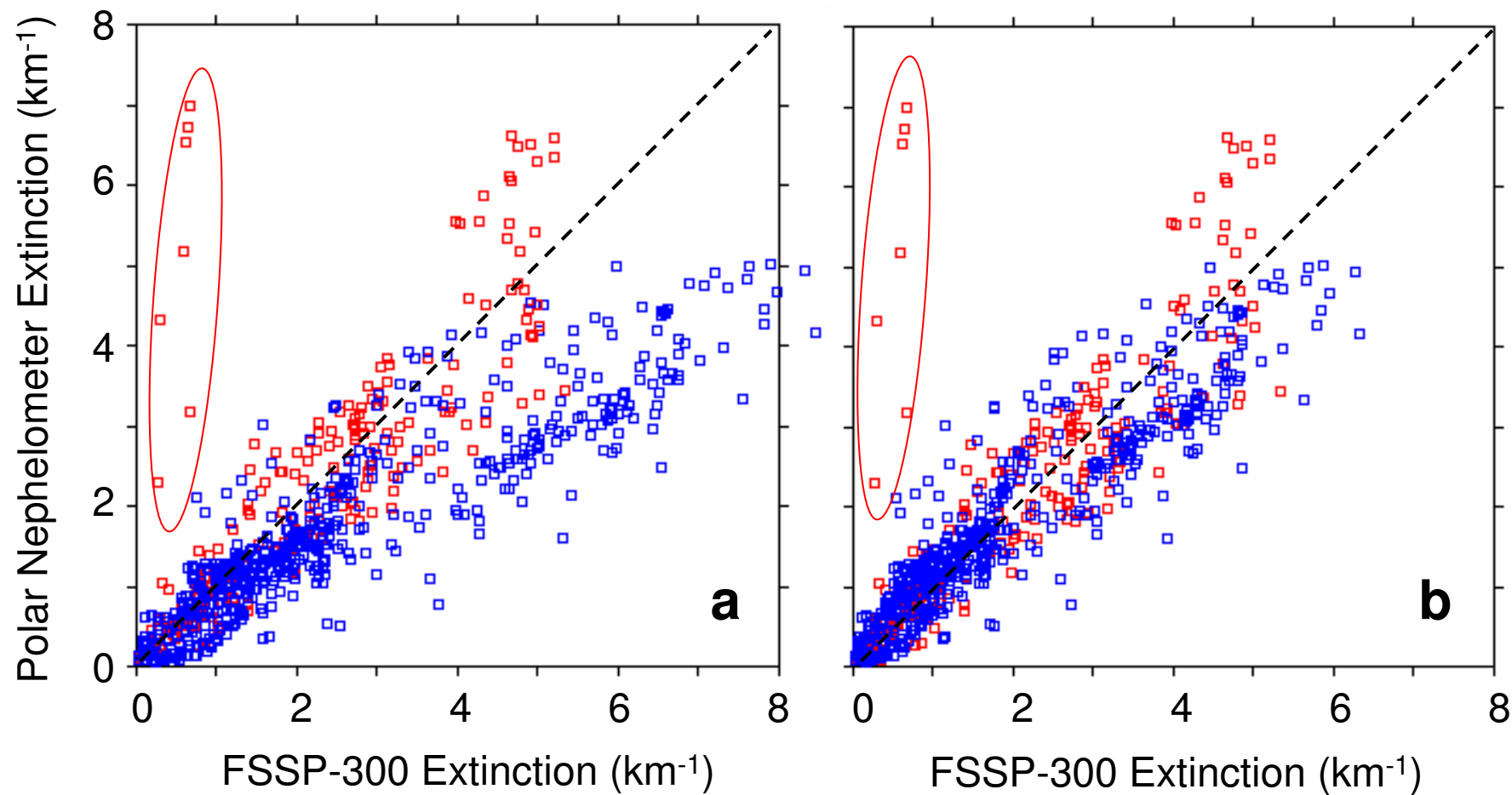
The small values of the residuals (see Table 1) signify that the employed model fits well the measured phase functions. That is, a mixture of spheroids with a size-independent partitioning ratio and the retrieved value of the refractive index are in good (not excellent) agreement with the recorded optical data.

Retrievals with the model of only spherical particles provide some size distributions. In contrast to the mixture of spheroids, the phase functions, computed from the retrieved size distributions of spheres, have high values of residual in the cases B and C (Figs. 2 and 4). This should now be clearer from the revised paper.

Line 417 - I don't understand what this means? Why split between oblate and prolate. What does this accomplish?

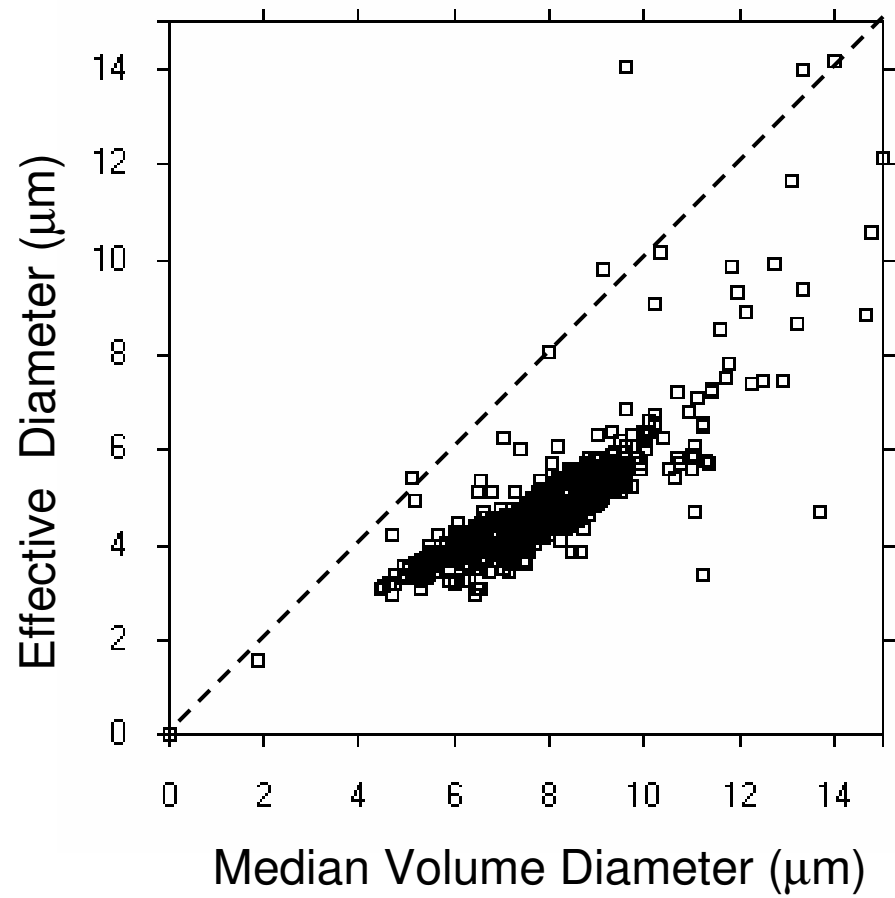
The phase functions of the oblate and prolate spheroids are not identical (see, e.g., Mishchenko et al., 2002). At the same time, the information content of the PN data is inadequate to retrieve the percentage of each class of particles. In the absence of better

knowledge, we assumed that half of particles were plate-like (oblate spheroids) and the other half were column-like (prolate spheroids).



Comparison between extinction coefficients from FSSP-300 and Polar Nephelometer probes. (a): FSSP-300 size calibration for spherical particles, (b): Size calibration for spherical (red data) and aspherical (blue data). The red and blue square symbols refer to asymmetry parameter > 0.85 and < 0.85 , respectively.

Figure A1



**Effective diameter versus Median Volume diameter
derived from the FSSP-300.**

Figure A2