



**The Met. Office**

**MRF Technical Note No. 31**

**Corrections to be applied to the PSAP and nephelometer for accurate determination of the absorption coefficient, scattering coefficient and single scattering albedo.**

**by**

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10<sup>th</sup> July 2000

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## 1. Introduction

The Meteorological Research Flight (MRF) C-130 aircraft has two dedicated instruments that are used to measure the scattering and absorption coefficients in aerosol studies. The aerosol absorption coefficient ( $\sigma_{ap}^\lambda$ ) is measured at a single wavelength ( $\lambda = 0.567\mu\text{m}$ ) by the Particle Soot Absorption Photometer (PSAP) designed by Radiance Research. Scattering is measured at three wavelengths ( $\lambda = 0.45, 0.55, 0.70 \mu\text{m}$  with a bandwidth of  $0.04 \mu\text{m}$ ) by the TSI integrating nephelometer (model 3563). The nephelometer provides the total scattering and hemispheric backscattering coefficients for each wavelength, denoted as  $\sigma_{sp}^\lambda$  and  $\sigma_{bsp}^\lambda$  respectively. General performance information of the nephelometer can be found in Anderson *et al.* (1996). The coefficients from both PSAP and nephelometer have the units of inverse distance.

An important optical property of atmospheric aerosols that is used in radiative transfer calculations is the single scattering albedo ( $\omega_o^\lambda$ ) which is defined as:

$$\omega_o^\lambda = \frac{\sigma_{sp}^\lambda}{\sigma_{sp}^\lambda + \sigma_{ap}^\lambda} \quad (1)$$

This note describes recommended correction factors to apply to the PSAP and nephelometer raw recorded data as described by Anderson and Ogren (1998) and Bond *et al.* (1999) respectively to deduce  $\omega_o^\lambda$  as accurately as possible. Section 2 describes correction factors to be applied to the PSAP, section 3 describes correction factors applied to the nephelometer, and section 4 describes an additional correction necessary for the most accurate determination of  $\omega_o^{0.55}$ . These correction factors are then applied to *in-situ* measurements made with the MRF C-130 in section 5. Conclusions and suggested improvements to the operating procedures/calibrations of the instruments are given in section 6.

## 2. PSAP

The PSAP provides a continuous measurement of particulate absorption by monitoring the change in transmission of  $0.567 \mu\text{m}$  LED radiation across a fibrous glass filter. For the PSAP, the state of hydration of sampled aerosol is not important because of the negligible effect on absorption due to any water deposited on the filter. The equation governing the operating principles of the PSAP gives an absorption coefficient,  $\sigma_{ap}^{0.567}$ :

$$\sigma_{ap}^{0.567} = \frac{A}{V} \ln\left(\frac{I_o}{I}\right) \quad (2)$$

where  $A$  is the area of the spot caused by the filtered aerosol,  $V$  is the volume of air drawn through the filter in a given time period, and  $I_o$  and  $I$  are the filter transmittances before and after this time period, respectively.

From this equation Bond *et al.* (1999) deduces that errors in the PSAP can arise from:

- 1) Inaccurate knowledge of spot size and hence  $A$
- 2) Inaccurate knowledge of flow rate and hence  $V$
- 3) Spurious absorption due to the misinterpretation of scattering as absorption by purely scattering particles in measurements of the transmittance
- 4) Overestimate of absorption due to external mixture of absorbing and scattering particles causing a degree of multiple scattering.

## 2.1 Correction for variations in spot size

This error is inherent because an erroneous spot sample area is assumed in the manufacturers' calibration. According to Bond *et al.* (1999), the manufacturers' calibration assumes a spot area of  $1.783 \times 10^{-5} \text{ m}^2$ , which is equivalent to a circular spot diameter of approximately 4.765mm. This is the diameter of the circular hole in the filter holder. However, the exposed area of the filter is somewhat larger owing to the use of O-rings in the filter holder to provide an airtight seal. Laboratory measurements of the variation in spot size using a number of filters that have been exposed in-flight reveal a spot size diameter of 5.19mm with a standard deviation of 0.245 and a standard error of 0.06 (n=16). Thus the manufacturers' calibration diameter does lie within two standard deviations of the mean measured spot diameter. However, Bond *et al.* (1999) examine many filters and suggest a spot diameter of 5.1mm for their reference PSAP. J. Ogren (*personal communication*) suggests spot diameter of approximately 5.2 based upon results from a number of different PSAP instruments. Therefore, the following correction ( $C_{ss}$ ) relating the measured absorption coefficient,  $\sigma_{ap\ meas}^{0.567}$ , to the absorption coefficient corrected for spot size,  $\sigma_{ap\ ss}^{0.567}$ , is applied using the MRF data:

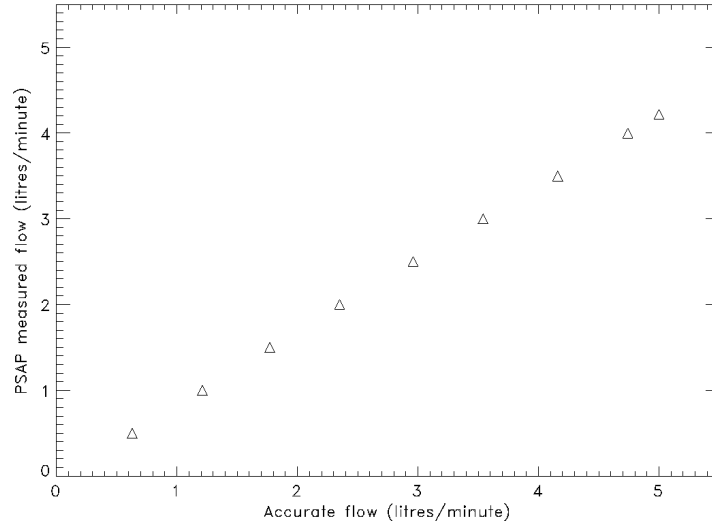
$$\sigma_{ap\ ss}^{0.567} = C_{ss} \sigma_{ap\ meas}^{0.567} = \left( \frac{4.765}{5.19} \right)^2 \sigma_{ap\ meas}^{0.567} = (0.84 \pm 0.02) \sigma_{ap\ meas}^{0.567} \quad (3)$$

## 2.2 Correction for inaccuracies in the flow rate

Bond *et al.* (1999) measured the flow rate indicated by internal flow meter of the PSAP against an accurate bubble flow meter, and found that the PSAP flow meter may be inaccurate by up to 20%. The accuracy of the flow meter in the MRF PSAP was tested using a bubble flow meter. Figure 1 shows that the flow meter in the PSAP systematically underestimates the actual flow. Hence a correction to the flow rate,  $V$  in equation (2) leads to a correction ( $C_{flow}$ ) in the absorption coefficient of the form:-

$$\sigma_{ap\ flow}^{0.567} = C_{flow} \sigma_{ap\ meas}^{0.567} = (0.84 \pm 0.02) \sigma_{ap\ meas}^{0.567} \quad (4)$$

Note that it is coincidence that  $C_{flow}$  and  $C_{ss}$  are identical.



**Figure 1.** Graph of the actual flow versus the PSAP measured flow. The slope determines the correction factor  $C_{flow}$  in equation (4).

### 2.3 Correction due to scattering being misinterpreted as absorption

Bond *et al.* (1999) performed a series of tests to determine to what degree scattering was misinterpreted as absorption by the PSAP. These tests included the use of laboratory controlled purely scattering aerosols in the PSAP. The results suggest that about 2% of the scattering was interpreted as absorption. Thus a purely scattering aerosol would give a single scattering albedo of 0.98 when measured by the PSAP. It is impractical at the present time to carry out such tests on the MRF PSAP. Therefore this correction will be applied here.

### 2.4 Correction due to multiple scattering

For a partially absorbing aerosol, the degree of absorption will be enhanced due to the effects of multiple scattering within the sample. Measurements by Bond *et al.* (1999) suggest that the PSAP overestimates the absorption (once the factors i-iii have been accounted for) by a further 22%. It is impractical at the present time to carry out such tests on the MRF PSAP. Therefore this correction will be applied here.

### 2.5 Summary of corrections applied to the PSAP

The required adjustments and uncertainties are summarised thus (Bond *et al.*, 1999):-

$$\sigma_{ap\,corr}^{0.567} = \frac{C_{ss} C_{flow} \sigma_{ap\,meas}^{0.567} - K_1 \sigma_{sp}^{0.567}}{K_2} \quad (5)$$

where  $C_{ss}=0.84 \pm 0.02$ ,  $C_{flow}=0.84 \pm 0.02$ .  $K_1 = 0.02 \pm 0.02$  and  $K_2 = 1.22 \pm 0.20$  represent the response of the instrument to scattering and absorption, respectively.  $\sigma_{sp}^{0.567}$  is the scattering coefficient as measured by the nephelometer. Because of the use of nephelometer data in producing the  $K_1$  term, the correction to the PSAP is ultimately limited by the accuracy of the nephelometer.

There are two additional factors to take into account when estimating the error in  $\sigma_{apcorr}^{0.567}$ . Firstly, unit to unit variability of the instruments (Bond *et al.*, 1999) leads to an uncertainty (2 standard deviations),  $\epsilon_{utu}$ , defined as:

$$\epsilon_{utu} = \pm 0.06 \sigma_{apmeas}^{0.567} \quad (6)$$

Secondly, instrumental noise (Anderson *et al.*, 1999) leads to an uncertainty (2 standard deviations),  $\epsilon_{noise}$ , defined as:

$$\epsilon_{noise} = \pm 0.18 \times 10^{-6} \sqrt{\frac{\tau_0}{\tau}} \quad (\text{m}^{-1}) \quad (7)$$

for sub-micron aerosol which depends on the averaging time  $\tau$  compared to  $\tau_0$  (24 mins). The corrections to be applied to the nephelometer scattering coefficient are now investigated.

### 3. Nephelometer

Scattering measurements are subject to three correction factors that need to be taken into account for accurate determination of the single scattering albedo,  $\omega_o^\lambda$ . Firstly, the nephelometer performs a truncation of the scattered radiation, cutting off radiation at angles of less than  $7^\circ$  scattering angle. Combined with the slightly non-cosine weighted intensity distribution of illumination light provided by the opal glass diffuser, this leads to a correction factor that accounts for the total scattering,  $C_{ts}$ , (Anderson and Ogren, 1998). Secondly, the nephelometer is calibrated at STP and adjustments need to be made to account for variations in temperature and pressure from STP via a correction factor,  $C_{STP}$ . Thirdly, the aerosol sample within the nephelometer is dried to a low but variable relative humidity. Thus a correction factor,  $C_{RH}$ , should be applied.

#### 3.1 Correction for the missing forward scattering

The scattering coefficient corrected for total scattering,  $\sigma_{spcorrts}^\lambda$ , is related to the measured scattering coefficient at each wavelength,  $\sigma_{spmeas}^\lambda$ , by a correction factor,  $C_{ts}^\lambda$ , defined by:-

$$C_{ts}^\lambda = \frac{\sigma_{spcorrts}^\lambda}{\sigma_{spmeas}^\lambda} \quad (8)$$

Anderson and Ogren (1998) have investigated the relationship between the Ångström coefficient,  $\mathring{A}(\lambda_1/\lambda_2)$ , measured by the nephelometer and the correction factor,  $C_{ts}^\lambda$ . The Ångström coefficient is defined by:-

$$\mathring{A}(\lambda_1 / \lambda_2) = - \frac{\log(\sigma_{sp\ meas}^{\lambda_1} / \sigma_{sp\ meas}^{\lambda_2})}{\log(\lambda_1 / \lambda_2)} \quad (9)$$

The aerosol inlet outside of the aircraft skin and the aerosol transport method within the aircraft to the PSAP and nephelometer strongly imply that the aerosol measured is predominantly sub-micron because of the very low efficiency in sampling large ( $> 1 \mu\text{m}$ ) particles (J. Heintzenberg, *personal communication*). Therefore it can be assumed that for the purposes of corrections to the nephelometer, the sub-micron (as opposed to super-micron correction or an average of the two) corrections presented in Anderson and Ogren (1998) will be used in this report. Anderson and Ogren (1998) find a reasonably linear relationship for  $-0.1 < \mathring{A} < 2.5$  between  $C_{ts}^\lambda$  and  $\mathring{A}(\lambda_1/\lambda_2)$ . For the central nephelometer wavelength of  $0.55 \mu\text{m}$ ,  $\mathring{A}(\lambda_1/\lambda_2)$  is determined for  $\lambda_1 = 0.45 \mu\text{m}$  and  $\lambda_2 = 0.70 \mu\text{m}$  and the relationship is summarised by:-

$$C_{ts}^{0.55} = -0.044 \mathring{A} + 1.152 \quad (10)$$

Anderson and Ogren (1998) report that this correction is reasonable for the other wavelengths of the nephelometer. The error in the correction is difficult to determine but is estimated to be approximately  $0.33 C_{ts}^{0.55}$ .

### 3.2 Correction for STP

The nephelometer measures the total scattering (i.e. the scattering due to the aerosol and the gaseous constituents of the sampled air). The degree of scattering of the gaseous components will depend upon the density of the air. The nephelometer is calibrated using at STP and therefore, in a similar way to the correction for missing forward scattering, the scattering corrected for differences from STP,  $C_{STP}^\lambda$ , may be defined by (Anderson and Ogren, 1998):-

$$C_{STP}^\lambda = \sigma_{sp\ corrts}^\lambda \frac{T}{273.2} \frac{1013.2}{p} \quad (11)$$

where  $T$  is the absolute temperature and  $p$  is the pressure (in hPa).

### 3.3 Correction for the effects of relative humidity

The relative humidity within the nephelometer under typical operating conditions (pump rate  $100 \text{ L min}^{-1}$  with the inlet heaters turned on) is variable. The relative humidity of the sampled air is reduced from ambient relative humidities by heaters in the inlets, heating by the instrument electronics, and dynamic heating due to deceleration of the sampled air. The relative humidity inside the instrument is recorded automatically by the instrument. An analysis of the nephelometer relative humidity,  $\text{RH}_{\text{neph}}$ , reveals a typical range of between close to 0% (corresponding to the free troposphere) and 50 % (corresponding to an almost saturated marine

boundary layer). This creates problems when attempting to deduce a ‘dry’ scattering coefficient comparable to the ‘dry’ absorption coefficient measured by the PSAP. This is because typical boundary layer aerosol particles such as ammonium sulphate and sea-salt will still hold deliquesced water and therefore be larger than fully dehydrated particles. An ammonium sulphate particle on the drying branch of the metastable region of deliquescence at 50 % RH is larger in size by a factor of approximately 1.22 relative to the crystallised state (Tang, 1996). This implies that the measured  $\sigma_{sp}^\lambda$  and  $\sigma_{bsp}^\lambda$  will be larger than their dehydrated counterparts. Based on model calculations, for a reasonable ammonium sulphate aerosol size distribution (log-normal distribution with mode radius of 0.05 $\mu\text{m}$ , and a geometric standard deviation of 2.0),  $\sigma_{sp}^{0.55}$  reduces by a factor of roughly 0.65 when the aerosol is dried from 50 % to below 37 % i.e. an RH below the crystallisation point. While no uniform correction factor is suggested to account for this feature, it is suggested that future measurement procedures should attempt to minimise this effect (section 6).

#### 4. Determining the single scattering albedo, $\omega_o^{0.55}$

A further adjustment that should be included in calculating  $\omega_o^{0.55}$ , is that the absorption coefficient measured by the PSAP is at a wavelength of 0.567 $\mu\text{m}$  rather than 0.55 $\mu\text{m}$ . The variation in the absorption coefficient with wavelength depends firstly upon the size distribution and secondly upon the refractive index, therefore no systematic correction may be applied. It is recommended that Mie scattering calculations are performed using the size distribution determined by the Particle Measuring System (PMS) PCASP (Passive Cavity Aerosol Spectrometer Probe) and PMS FSSP (Forward Scattering Spectrometer Probe) using suitable refractive indices to determine the spectral dependence of the absorption coefficient between 0.55 $\mu\text{m}$  and 0.567 $\mu\text{m}$ . Tests using size distributions from TARFOX (Tropospheric Aerosol Radiative Forcing eXperiment), SHAREM (SHip Antisubmarine warfare Readiness/Effectiveness Measuring exercise) and MOTH (Measurements Of Tropospheric Humidity) and suitable refractive indices show that the magnitude of such a correction may lead to an increase in the absorption coefficient by up to a factor of 1.06.

### 5. Examples

#### 5.1 MOTH transits (Flight A677)

Flight A677 took place on 8 May 1999 from Ascension Island to Tenerife. Saharan dust aerosol was encountered, the size distribution determined using the PCASP and the FSSP, and *in-situ* measurements of the absorption and scattering coefficients were made using the PSAP and nephelometer.

The mean data and estimated standard deviations were recorded from runs 1.1 and 1.2 at a height of approximately 8000ft in a relatively homogeneous Saharan dust layer. The runs were of 10 minutes duration, the mean pressure was 752.5 hPa, and the mean air temperature was 289.3 K. The ambient relative humidity was approximately 25%, and  $\text{RH}_{\text{neph}}$  was less than 4%. The scattering and absorption coefficients are:

$$\begin{aligned}\sigma_{ap\,meas}^{0.567} &= 4.41 \times 10^{-6} \text{ stdev } 0.53 \times 10^{-6} \text{ m}^{-1} \\ \sigma_{sp\,meas}^{0.450} &= 2.96 \times 10^{-5} \text{ stdev } 0.35 \times 10^{-5} \text{ m}^{-1} \\ \sigma_{sp\,meas}^{0.550} &= 3.04 \times 10^{-5} \text{ stdev } 0.29 \times 10^{-5} \text{ m}^{-1} \\ \sigma_{sp\,meas}^{0.700} &= 2.75 \times 10^{-5} \text{ stdev } 0.26 \times 10^{-5} \text{ m}^{-1}\end{aligned}$$

### 5.1.1 Correction to PSAP data

Using equation (5):-

$$\sigma_{ap\,corr}^{0.567} = \frac{C_{ss} C_{flow} \sigma_{ap\,meas}^{0.567} - K_1 \sigma_{sp\,meas}^{0.567}}{K_2}$$

where the mean values and standard errors are assumed to be:-

$$C_{flow} = 0.84 \pm 0.02$$

$$C_{ss} = 0.84 \pm 0.02$$

$$\sigma_{ap\,meas}^{0.567} = 4.41 \times 10^{-6} \text{ m}^{-1}$$

$\sigma_{sp\,meas}^{0.567} = 3.01 \times 10^{-5} \text{ m}^{-1}$  (by linear interpolation. Note that this correction derived by Bond *et al.* (1999) uses the uncorrected nephelometer scattering coefficient.)

$$K_1 = 0.02 \pm 0.02$$

$$K_2 = 1.22 \pm 0.20$$

$$\epsilon_{utu} = 0.06 \sigma_{ap\,meas}^{0.567} = 2.65 \times 10^{-7} \text{ m}^{-1}$$

$$\epsilon_{noise} = 0.18 \times 10^{-6} \sqrt{\frac{\tau_0}{\tau}} = 1.25 \times 10^{-7} \text{ m}^{-1}$$

leads to a corrected value,  $\sigma_{ap\,corr}^{0.567} = 2.06 \times 10^{-6} \text{ m}^{-1}$ . A basic error analysis (including all the +/- standard errors above and accounting for  $\epsilon_{utu}$  and  $\epsilon_{noise}$ ) for deriving the standard error suggests  $\sigma_{ap\,corr}^{0.567} = 2.1 \pm 0.6 \text{ Mm}^{-1}$ .

### 5.1.2 Correction to nephelometer data

Using equation 9:

$$\hat{A}(\lambda_1 / \lambda_2) = - \frac{\log(\sigma_{sp\,meas}^{\lambda_1} / \sigma_{sp\,meas}^{\lambda_2})}{\log(\lambda_1 / \lambda_2)}$$

$$\text{Gives } \hat{A}(450/700) = 0.166$$

Application of equation 10:

$$C_{ts}^{0.55} = -0.044 \dot{A} + 1.152$$

gives  $C_{ts} = 1.14 \pm 0.$ . Thus  $\sigma_{sp\ corr\ ts}^{0.55} = (1.14 \pm 0.) \sigma_{sp\ meas}^{0.55} = 35 \pm 12 \text{ Mm}^{-1}$ .

Application of equation 11:

$$C_{STP} = \sigma_{sp\ corr\ ts}^{\lambda} \frac{T}{273.2} \frac{1013.2}{p}$$

gives  $\sigma_{sp\ corr}^{0.55} = 50 \pm 15 \text{ Mm}^{-1}$  (assuming the correction to STP introduces errors of less than 5%).

In this case,  $RH_{neph}$  is 4% and therefore no correction factor due to the effects of relative humidity,  $C_{RH}$ , needs to be applied.

### 5.1.3 The single scattering albedo

Application of Mie theory using the refractive indices and size distributions detailed in Haywood *et al.* (2000), reveals that the absorption coefficient should be increased by a factor of approximately 1.03 to account for the differences between the absorption at  $0.567\mu\text{m}$  and  $0.55\mu\text{m}$ . Thus, using the corrections detailed above, the following values for single scattering albedo were deduced:-

$$\omega_{o\ uncorrected}^{0.55} = \frac{\sigma_{sp}^{0.55}}{\sigma_{sp}^{0.55} + \sigma_{ap}^{0.567}} = \frac{3.04 \times 10^{-5}}{3.04 \times 10^{-5} + 4.41 \times 10^{-6}} = 0.87$$

$$\omega_{o\ corrected}^{0.55} = \frac{\sigma_{sp}^{0.55}}{\sigma_{sp}^{0.55} + \sigma_{ap}^{0.55}} = \frac{5.0 \times 10^{-5}}{5.0 \times 10^{-5} + 2.1 \times 10^{-6}} = 0.96 \pm 0.02$$

Thus the single scattering albedo is significantly larger when the corrections are accounted for. This is because the majority of the corrections made above decrease the magnitude of the absorption coefficient while increasing the magnitude of the scattering coefficient. The magnitude of the error in  $\omega_o$  is relatively small when applying the corrections noted above. However, this is misleading as it is the co-albedo (i.e.  $1 - \omega_o$ ) that determines the degree of absorption. Thus it should be noted that the estimated error in the co-albedo when applying these corrections may be 50%. Note also that the estimated error does not include uncertainties due to atmospheric variations in the measured absorption and scattering coefficients, merely the uncertainties associated with application of the correction factors listed above.

## 6. Conclusions and suggestions for future improvements

It is relatively straightforward to apply the correction factors described in section 2 to the PSAP. The flow correction factor,  $C_{flow}$ , and the spot size correction factor,  $C_{ss}$ , have been shown to be in reasonable agreement with those derived by other groups using different PSAP instruments. However, the two important correction factors  $K_1$  and  $K_2$  in equation (5) can only be determined by careful laboratory calibration using artificially generated absorbing and non-absorbing aerosol. In the absence of

laboratory calibration, it is suggested that the coefficients  $K_1$  and  $K_2$  from Bond *et al.* (1999) be implemented. There is, to date, only one single MRF comparison of absorption coefficients derived from the PSAP against other methods. Hignett *et al.* (1999), calculated an uncorrected  $\sigma_{ap}^{0.567}$  of approximately  $3.9 \times 10^{-6} \text{ m}^{-1}$  from the TARFOX campaign compared to a value of approximately  $2.8 \times 10^{-6} \text{ m}^{-1}$  from the integrating sandwich technique using exposed Nuclepore filters (Clarke, 1982). Application of the correction factors  $C_{ts}$ ,  $C_{flow}$ ,  $K_1$  and  $K_2$  (the nephelometer was not operational hence  $\sigma_{sp}$  cannot be accurately determined, but may be estimated from the single scattering albedo reported) lead to a PSAP corrected absorption coefficient of  $1.8 \times 10^{-6} \text{ m}^{-1}$ . Thus the agreement between the two methods is not improved. However, the absolute value of the absorption coefficient from the integrating sandwich technique may also be subject to correction factors analogous to the correction factors  $K_1$  and  $K_2$  applied to the PSAP data, so the results are inconclusive. Future measurement/comparisons should help determine  $\sigma_{ap}$  and  $\omega_0$  to a higher degree of accuracy.

The recommended correction factors to apply to the nephelometer include a correction to account for the radiation scattered in the forward direction,  $C_{ts}$ , and a correction to account for the departures from STP,  $C_{STP}$ . Both of these factors may be readily included. A correction to account for the effects of relative humidity effects on hygroscopic aerosols will depend upon the chemistry of the aerosol particles, which is difficult to determine using the present instrumentation of the C-130. A recent flight (A770 on 7 July 2000) contained some nephelometer tests that included variation of the following parameters:

- (i) *nephelometer flow rate*– the pumping rate was set at either 100 L min<sup>-1</sup>, 30 L min<sup>-1</sup>, or zero pumping where the effect of ram air only was used.
- (ii) *air sampling pipe deicing heaters*– either switched on or off

Permutations of the above settings in both humid (i.e. marine boundary layer) and relatively dry (i.e. free tropospheric) conditions show that the flow rate of the deicing heaters has little effect on the relative humidity inside the nephelometer. This indicates that most of the drying of the aerosol must be due to dynamic heating when the air sample is decelerated, and that the other effects mentioned are secondary in comparison. Therefore it is recommended that the inlet pipe to the nephelometer be heated by an additional external source such that the air sample relative humidity is reduced to around 20 % i.e. a reduction of 30 % from the highest humidity at present measured within the nephelometer.

Note here that the calibrations detailed above assume that the aircraft samples predominantly sub-micron particles. If the sampling efficiency of giant aerosol is to be improved in the future, then different corrections may be needed.

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