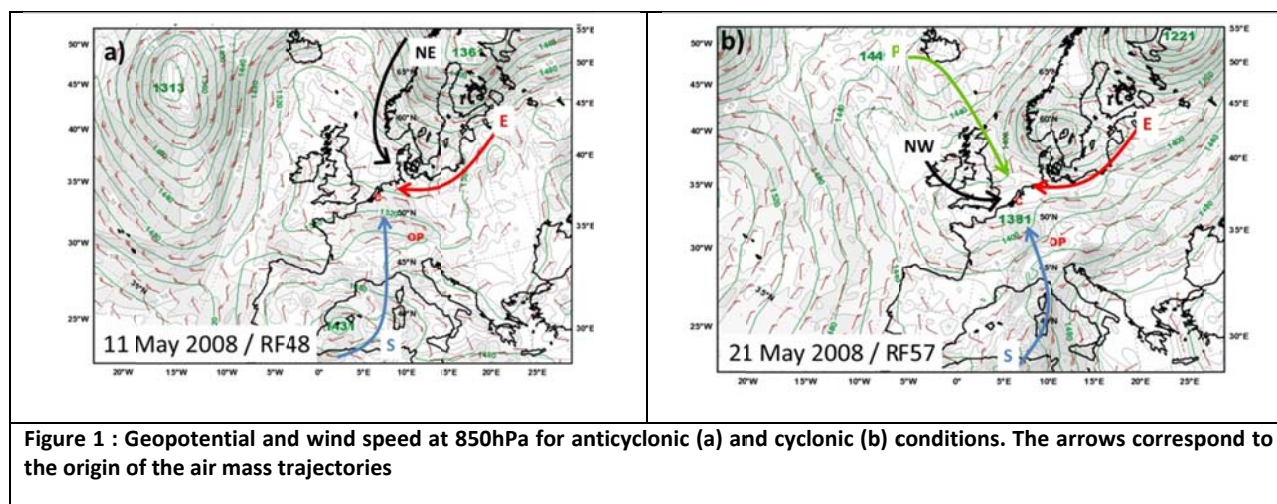


We thank both reviewers for their detailed and constructive comments on our manuscript. The authors apologize for all the edition mistakes the referees find in the paper which were due to a LATEX compiling issues within the editing office. The first author should have noticed that before publication on the ACPD site. We have revised the manuscript attempting to take into account all the comments raised by both reviewers.

## General Comments of Reviewer 2 :

1) Authors used FLEXPART for air mass origin analysis, although P sector includes London metropolitan area. Can authors clarify how this was taken into account that supposedly cleanest air mass sector includes huge pollution source nearby the most of the flights performed?

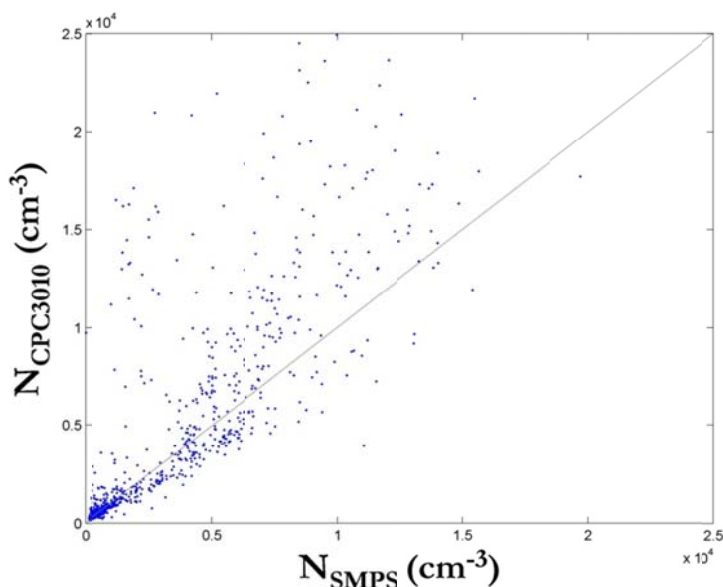
The referee may refer to the figure 1 to say that London is a part of the P sector. The figure 1 was a schematic view of the different sectors. Unfortunately, London was in the P sector on the figure but not on the actual classification. According to the referee 1 comments, we added in the manuscript the Figure 1 (a and b) which depicts the geopotential and the wind speed at 850hPa. The blocks as they appeared in the former picture are now replaced by arrows which represent better the air mass paths as they occurred.



2) Table 2 brings doubts on quality of aerosol size distribution measurements. How N50 can be higher than N10 for S-EUR sector in BL during LP conditions? Similar discrepancy is for E-EUR in BL and E-EUR FT. Does it mean that Integral of the SMPS is not even closely corresponding to N10 measured by CPC? Why? Unless this is clarified, whole data analysis is questionable.

The CPC 3010 concentration, measured during the whole campaign, is represented as a function of the SMPS on the **Error! Reference source not found.** We can see that most of the time the total particle concentration measured by SMPS and the CPC 3010 agreed reasonably well most of the time. Using a linear fitting line, we highlight that the total number of particles measured by the SMPS is underestimated as compared to the one measured by the CPC by a factor of 30%. This result may be due to the presence of particles below the SMPS lower particle size of 20 nm. The relatively wide dispersion

is certainly due to particle concentration variations during one SMPS scan lasting 90 seconds. Thus, the particle concentrations from individual SMPS scans may show deviations from averaged total concentrations.



**Figure 2:** Total concentration measured by the CPC 3010 as a function of the total concentration by the SMPS. The grey color corresponds to the 1:1 line.

This difference in the acquisition frequency causes a lot of discrepancies between the CPC and the SMPS measurements, especially in an aircraft where spatial concentration variations of aerosol particles become important. Indeed, the measurements of the CPC are performed at 1Hz while the SMPS measurements are performed at 0.01Hz resulting in a large data set for the CPC measurements and a reduced number of SMPS measurements. To somehow better compare the total concentration measured by the CPC with SMPS concentrations from the integration of one scan, we apply a moving average to the whole CPC data set based on the measurement periods of the SMPS (90s). Only one value of the averaged CPC concentrations is compared to the SMPS values. The results (Table 1) are still showing some discrepancy between the CPC total concentration and the integration of the SMPS size spectra. The difference is lower than 8% of the value which is in average in the uncertainty of both instruments.

**Table 1 :** Median number concentration in the boundary layer (BL) and in the free troposphere (FT) during anticyclonic (HP) and cyclonic (LP) conditions. N<sub>10</sub> : CN concentration for particles larger than 10nm measured with the CPC3010 , N<sub>50</sub> : CN concentration for particles larger than 50nm measured with the SMPS and N<sub>500</sub> : CN concentration for particles larger than 500nm measured with the PCASP. The values in bracket are the former values.

|    |        | N <sub>10</sub> (cm <sup>-3</sup> ) |                | N <sub>50</sub> (cm <sup>-3</sup> ) |      | N <sub>100</sub> (cm <sup>-3</sup> ) |      |
|----|--------|-------------------------------------|----------------|-------------------------------------|------|--------------------------------------|------|
|    |        | HP                                  | LP             | HP                                  | LP   | HP                                   | LP   |
| BL | NW-EUR | 1370<br>(1300)                      | --             | 150                                 | --   | 128                                  | --   |
|    | S-EUR  | --                                  | 5080<br>(4800) | --                                  | 5400 | --                                   | 4556 |
|    | NE-EUR | --                                  | 7171<br>(5970) | --                                  | 2900 | --                                   | 1739 |

|    |        |                |                |      |      |      |      |
|----|--------|----------------|----------------|------|------|------|------|
|    | E-EUR  | 5870<br>(5490) | 5444<br>(3135) | 3240 | 5820 | 2246 | 3754 |
|    | P      | --             | 7090<br>(7090) | --   | 1390 | --   | 980  |
|    |        |                |                |      |      |      |      |
| FT | NW-EUR | --             | --             | --   | --   | --   | --   |
|    | S-EUR  | --             | 1048<br>(930)  | --   | 660  | --   | 589  |
|    | NE-EUR | 601 (660)      | 438 (400)      | 80   | 260  | 33   | 159  |
|    | E-EUR  | 1505 (865)     | 1297<br>(920)  | 680  | 940  | 472  | 809  |
|    | P      | --             | 466 (440)      | --   | 325  | --   | 292  |

3) Water vapor/humidity, ozone, CO were present onboard of ATR-42. Why these parameters have not been used. They will make the conclusions and claims more robust.

The authors decided to remove all the Angstrom exponent section which didn't improve the quality of the paper. And as no evidence of mixing has been found, all the discussion about the mixing of both layers (BL and LFT) has been removed.

Detail comments:

Diffusional losses and associated corrections are not mentioned in manuscript :

Using quite simple calculations according to Baron we may have lost up to 10% of the 10 nm particles and some 4% of the 20nm particles in the as well in the single CPC as in the SMPS systems (anyway the systems start to measure at 20nm). We did not mention that in the text.

P9458, L20: Why state variables measured onboard of the airplane were not used? How do they compare to soundings?

The state variables measured onboard the ATR-42 were used to highlight the boundary layer height.

Chapter 3: What exactly is meant here by FT? What altitude range? Is the altitude coverage and distribution of the measurements in FT the same for every sector in absolute values and with respect to BL height?

The free troposphere is used to refer to the lower free troposphere. As both referee were suggesting, we added the ATR-42 altitude range in the table 1 (in the manuscript). The values are pretty similar from sector to sector (2500-3000 m) except for the NE-EUR (>3000 m). Thus, the results for this air mass sector will not be necessarily compared to the results of the other air mass sectors.

P9459, L11-14: How do you know this? From FLEXPART?

Again here, the first author would like to apology for the LATEX compiling issue. This sentence was not supposed to appear here. This has been corrected in the actual version.

P9459, L17: What motivation did you have to derive N500 aerosol number density? What one can learn from it?

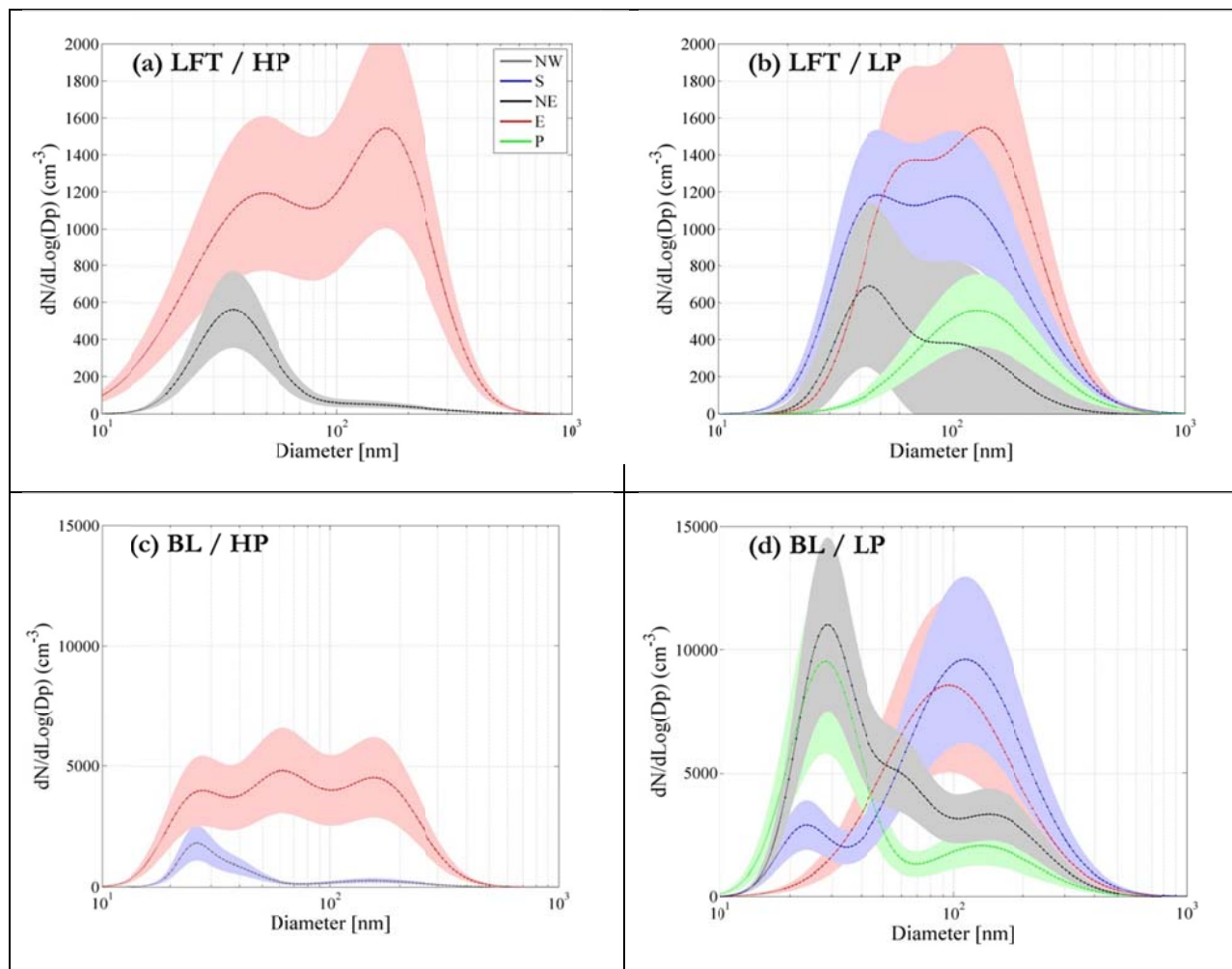
The authors motivation was to show any particular event involving plume of large particles that may influence the scattering coefficient as well as the CCN concentration. As both referees suggested and according to Asmi et al (2012), the CCN/N<sub>100</sub> ratio is more appropriate than the CCN/N<sub>50</sub> ratio. Thus the N<sub>100</sub> will be display instead of the N<sub>500</sub>.

P9460, L2-3: It is only one possibility. It can this be during LP conditions also due to spatial heterogeneity and intensity of removal processes

The referee is right and the sentence has been corrected accordingly: 'N<sub>10</sub> concentrations are more variable in the BL (especially for air masses coming from E-EUR sector during LP conditions) mainly due to the spatial heterogeneities of particle sources and the intensity of the removal processes.'

P9460, L23-24, Fig.3: Can you add standard deviation into the graph to see what is the variability of the size distributions or show median size distribution with quartiles?

The standard deviations have been added to the size distributions and are represented by the shading areas in the Figure 3.



**Figure 3 :Number particle size distribution ( $10\text{ nm} < D_p < 500\text{ nm}$ ) as a function of the air mass origin (NW : North West Europe, S: South Europe, NE: North Europe, E : East Europe, P : Polar) in the lower free troposphere during anticyclonic (HP, a) and cyclonic (LP, b) condition as well as in the boundary layer during anticyclonic (HP, c) and during cyclonic (LP, d) condition. The shading areas are corresponding to the standard deviation of the measurements.**

Fig. 3: Nucleation mode usually represents newly formed particles and conditions of recent or ongoing nucleation. How do you explain that size distribution show “close” shape with no particles present in small sizes? Can Dnucl can be also result of primary emissions (traffic for example)?

In clean air masses (P) the ultra fine particles mode is only due to secondary aerosol formation but in the case of polluted air masses (E-EUR) the fine particle mode may be due to the secondary aerosol formation as well as some primary emissions. This has been corrected throughout the manuscript.

P9461, L 3: Caption of Fig.3 shows that 3a is FT, but here it is stated as BL?

The caption of the figure 3 is right, thus we corrected the sentence by: ‘The nucleation mode, only observed in the boundary layer, is centered between 22 and 28 nm during anticyclonic conditions (Figure 3c) as well as during cyclonic conditions (Figure 3d).’

P9461, L25-28: Cloud processing of aerosol usually results in bi-modal size distribution (or even tri-modal when nucleation is present). On what basis authors attribute monomodal size distribution to cloud processing?

In this sentence the author wanted to asses that during their lifetime particle properties and then the size distribution may change because of various processes including cloud processing. If the particles resided a longer time into the atmosphere then they have a better chance to cross a cloud path and being processed. Nevertheless, the cloud processing is not the only process that may occur. The growth of particles by coagulation or gases condensation may dramatically change the size distribution shape and clear all evidences of cloud processing. As the argumentation in this sentence was not understood we rephrased it in: ‘These observations indicate that particles resided a longer time in the atmosphere during which they may grow by interacting with clouds, coagulation and gases condensation.’

Tab. 4 and associated text on page 9462: Why there is no discussion of very high aerosol volume concentrations in S-EUR and E-EUR during LP conditions? Those values are many times higher than the rest.

The discussion on the aerosol volume concentration was really short so we went into more detail in the corrected version of the manuscript :

‘The S-EUR air masses are strongly influenced by a dust particles event that stayed over the Netherlands for more than 3 days. The S-EUR volume concentration is the highest concentration observed in the BL ( $47.8\text{ }\mu\text{m}^3\text{ cm}^{-3}$ ). In the lower free troposphere, the aerosol volume concentration is high ( $3.48\text{ }\mu\text{m}^3\text{ cm}^{-3}$ ) comparable to the values reached for polluted air masses (E-EUR  $>3.95\text{ }\mu\text{m}^3\text{ cm}^{-3}$ ).’

Since the authors submitted this manuscript, a new study has been published (Begue et al. 2012) on this dust case. This has also been included in the corrected manuscript. The E-EUR air masses are highly concentrated during LP and HP conditions due to the passage over industrial and densely populated areas. This has been clarified in the manuscript.

P9462, L 13-24: Based on what authors claim that these particles are dust or sea salt?

The referee is right and we removed this statement.

P9464, L 1-5: Most of the NE-EUR sector is represented by Scandinavia and it is well known source of secondary organic aerosol from BVOC oxidation (e.g. (Tunved et al. 2006). Reference to Rinaldi paper is irrelevant as this article presents observations from Mace Head in Ireland and similar is valid for Asmi 2010 paper dealing with measurements from Antarctica. Both studies dealing with very different environments compared to NE-EUR sector. Authors should show robust analysis or argumentation and not speculation.

The chemical composition of the aerosol observed in the NE-EUR in the FT is totally different from the other ones and is also really particular. In the literature, the authors find few references showing similar aerosol chemical composition. These studies show similarities with the measurements presented in this manuscript. As the referee suggest that referring to these references is irrelevant we removed these sentences and replace it with : “Moreover, the organic absolute concentrations for NE are low ( $> 0.05 \mu\text{g}\cdot\text{m}^{-3}$ ) as well as the total aerosol concentration ( $\sim 1 \mu\text{g}\cdot\text{m}^{-3}$ ) which is comparable to results of a study on background rural aerosol particles (Hock et al., 2008). In situ measurements performed at Mace Head (O’Dowd et al, 2010) highlight that extended particle formation and growth events are not unusual over the Atlantic Ocean. Thus, the particular NE chemical composition might be linked to nucleation events occurring along the air mass transport.”

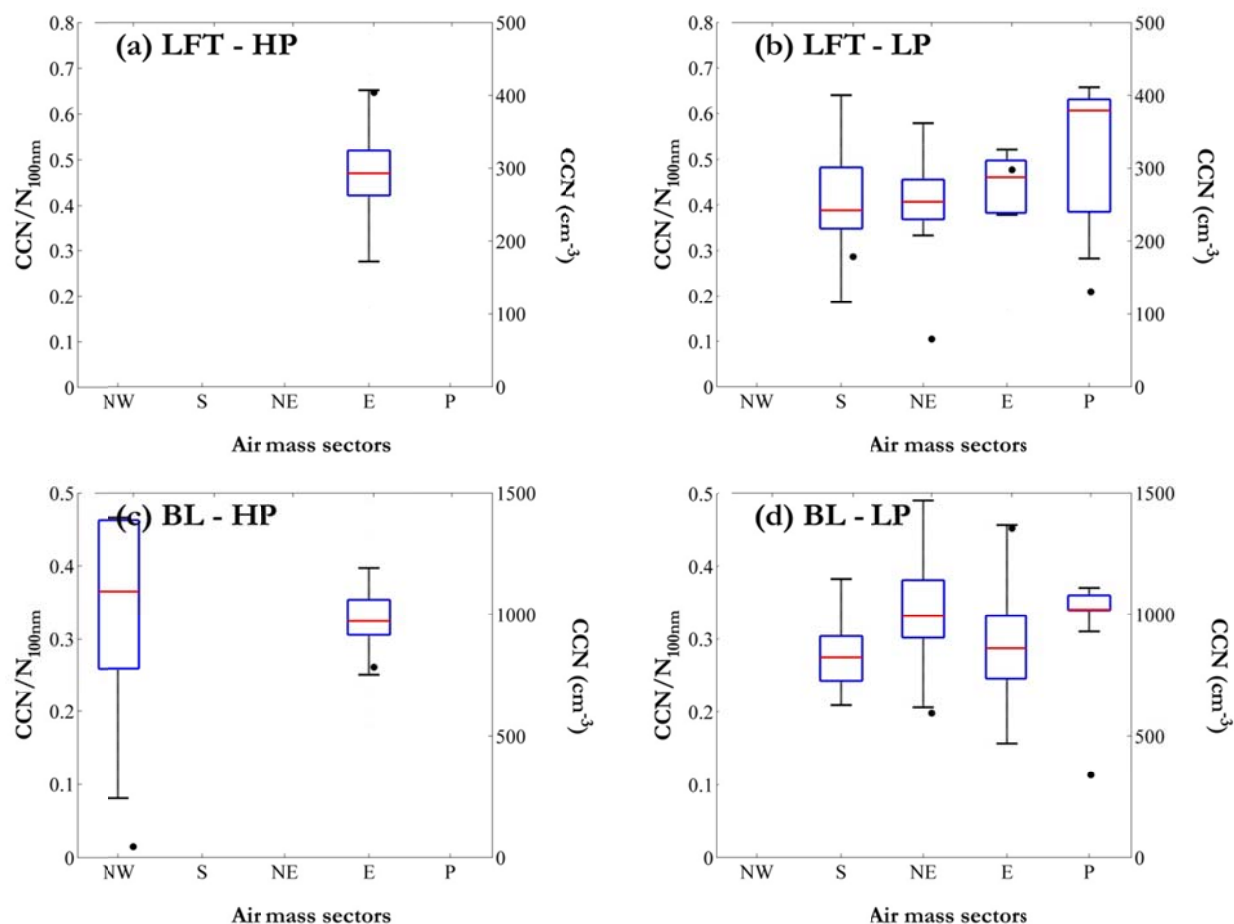
P9465, L4: what about size? It is like more important parameter than hygroscopicity of particles.

The size and the chemical composition are driving the hygroscopicity of the particles. In this study we try to describe the hygroscopicity of the particles using the CCN/CN ratio. The size distribution and the chemical composition are then used to understand the tendency observed on the CCN/CN ratios.

P9465, L7: Why 50 nm is representative for continental conditions, especially for rather polluted Europe? Based on what authors made this assumption? How does the picture change if it will be 70 or 90 nm instead?

The use of a higher cut off diameter changes the figures dramatically (**Error! Reference source not found.2**). As both referees suggested and according to Asmi et al (2012), the CCN/ $N_{100}$  ratio is more appropriate than the CCN/ $N_{50}$  ratio.

If the activation diameter is set at 100nm in the boundary layer, then the results change significantly (Figure 2). During high pressure conditions, the CCN/ $N_{100\text{nm}}$  ratios are about 0.17 and 0.34 respectively for NW-EUR and E-EUR (formally 0.23-0.22). During the low pressure conditions, the CCN/ $N_{100\text{nm}}$  ratio is found between 0.28-0.35 while it was formally in between 0.17-0.20. By taking into account only the particles larger than 100nm, the clean air masses (P, NE-EUR) are then associated to the largest values (0.35) consistent with the fact that marine aerosols are expected to be more soluble than aerosols of continental origin. The authors, thus, decide to include this figures and data into the manuscript as suggested by the referees



**Figure 4 :** CCN/N<sub>100</sub> ratios as a function of the air mass origin origin (NW : North West Europe, S: South Europe, NE: North Europe, E : East Europe, P : Polar) in the lower free troposphere during anticyclonic (HP, a) and cyclonic (LP, b) conditions as well as in the boundary layer during anticyclonic (HP, c) and cyclonic (LP, d) conditions.

P9468, end of section 4.5: Why authors did not use in situ data about LWC and trace gases to support their claims?

As this section of the manuscript did not improve the understanding of the manuscript and as no prove of mixing has been found, the authors decided to remove the last part of the optical section (Angstrom exponent discussion) from the manuscript.

P9469, L10-11: Can this be supported with trace gas measurements onboard of ATR-42?

P9469, L10 and L13: First you say that it is maybe and three lines later that the occasional mixing is confirmed. How is it possible? Unless you have clear independent parameters supporting this claim, nothing is confirmed.

As the referee 1 suggested and as the referee 2 made a lot of comments on the conclusion section, the authors corrected it :

“A comprehensive set of instruments performing meteorological, cloud microphysics and aerosol physico-chemical and optical measurements was integrated on the French research aircraft ATR-42 for the EUCAARI intensive observation period. The obtained measurements document clear relations between aerosol properties and air mass origins. Based on backward calculations with a Lagrangian

particle dispersion model, the observed air masses were classified into five sectors according to their predominant residence times in these sectors. Additionally, measurements performed under anticyclonic (during first half of the campaign duration) and cyclonic (during second half of the campaign duration) synoptic conditions were analyzed separately, also showing distinct characteristics.

The observations reveal a strong difference in  $N_{50}$  particle number concentrations between the boundary layer (BL) and the lower free troposphere (LFT). In particular,  $N_{50}$  concentrations are about five times higher within the BL as compared to those observed within the LFT. Observed size distributions are trimodal in the BL (ultra-fine, Aitken and accumulation modes) and generally bimodal in the LFT (Aitken and accumulation modes), which is consistent with previous studies (Birmili et al., 2001; Asmi et al., 2011). Moreover, the aerosol chemical composition observed during the EUCAARI campaign show large differences between boundary layer aerosol (BL) and lower free troposphere (LFT) aerosol. In the BL, the nitrate and organic, mainly originating from anthropogenic sources (refinery, ship tracks), are dominating the chemical composition while in the LFT the sulfate and organics are the main components. These results support a growing body of research that as aerosols age, they undergo an increase the sulfate fraction compared to organic fraction, which renders particles more hygroscopic, especially in the LFT.

Polluted air masses are characterised by high total number particle concentrations and low concentrations of ultrafine particles ( $N_{10-50}$ ). In the BL, the total mass concentration as well as the aerosol chemical composition are similar to those observed in Mexico City (DeCarlo et al., 2008). The relative chemical composition of particles within polluted air masses is dominated by organics (about 50%) and nitrates (20%) with notable amounts of sulfate.

Non-polluted air masses, in general originating from polar and Scandinavian regions, are characterised by high total particle concentrations most likely due to new particle formation events occurring over the sea. The chemical composition of particles in the BL are characterised by significant amounts of chloride (most likely from  $NH_4Cl$  originating from marine sources) and nitrate species (most likely from ship tracks (Lauer et al., 2007)), while, in the LFT, the sulfate and ammonium are largely dominant (>90% of the AMS components) linked to the presence of high contents of condensable gases over Scandinavia and over ocean (Gondwe et al., 2003; Lana et al., 2011).

The dust plume observed within the air masses coming from South Europe are characterised by high total and organic concentrations consistent with observations shown by Falkovich et al. (2001). These dust plumes are transported over urban areas where organic gases are condensed onto the dust particles. “

Fig: 2, 3, 5, 6, 7: quality is bad, barely visible. Concerning language, there are many typos, missing words and unclear sentences and I suggest careful check and language corrections.

The quality of the figures has been improved. Numerous typos, missing words were due to Latex compiling issue. To resolve this problem and avoid any others, we transform the manuscript into a word file.

Overall at present level manuscript does not bring a new science and fulfil quality level suitable for ACP and major revision is needed.

Synergetic measurements of size distributions, chemistry, optical and CCN properties in an airborne measurement campaign over Europe were never performed before, to our knowledge. We now emphasis in the manuscript what clear differences in BL and LFT aerosol particle size and chemistry can



be drawn, and which consequences these different characteristics have on the aerosol optical and CCN properties, directly linked to their direct and indirect radiative impact. Moreover, we outline that the vertical distribution of aerosols is not always a decreasing function of altitude, and that, independently of the transport of dust or sea salt, aged particles can also be found more concentrated at the intermediate level of 1-3 km.