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 1 :

Specific Comments:

- p. 9452 l. 16, The nucleation mode is sometimes defined as all particles with particle diameters of 1-10 nm, sometimes even up to 20 nm, but I never heard of nucleation mode particles with 25-28 nm particle diameter, please correct this statement.

This statement has been corrected and the sentence is now : 'The detection of the largest particle number concentrations occurred in air masses coming from Polar and Scandinavian regions for which an elevated number of fine mode (25 - 28nm) particles was observed and attributed to new particle formation over open sea. In the free troposphere (FT), typical observed N10 are of the order of 900cm^{-3} in polluted air masses and 400 - 600cm^{-3} in clean air masses, respectively'

- p. 9453 l. 9, the indirect effect is not only the modification of the "cloud distribution", it is also the modification of cloud properties.

This has been corrected in the manuscript.

- p. 9453 l. 14, I do not fully agree that the aerosol concentrations over "Europe are still not well quantified". There are many publications on this topic and the authors of the present manuscript should refer to these studies, here in the introduction, but also in the results section. This is a major criticism! Here are, far from being complete, a few potential references missing in the manuscript: Coen et al., 2011 ; Birmili et al., 2001; Tunved et al., 2005; Petzold et al., 2002.

Few references (26 in total), including the ones suggested by the referee, have been added in the introduction as well as in the result part. As these references are cited in different sections of the manuscript, we can't show the entire corrections but a list of the reference included :

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- p. 9454 l. 15, the aerosol-cloud-climate scale does not stop in the “millimeter” size range, please clarify what you mean or correct the range.

This statement has been modified in the last version of the manuscript: “To improve our understanding of processes related to atmospheric aerosol physics and chemistry, from aerosol formation to the overall aerosol-cloud-climate scale, the French ATR-42 research aircraft performed a total of 22 research flights between 2 May and 30 May 2008.”

- p. 9454 l. 19, the description of the measurement flights (and also of the measurement instrumentation below) should be detailed enough to understand the results of this manuscript without having to look into several other publications. It is not sufficient to provide abstract flight numbers. Moreover it is not clear at which altitude it was flown. Hence, please provide a description of both in THIS manuscript. This is, again, a major criticism.

A short description of the different flights is provided in this paragraph while the altitude range is given in the Table 1: “The 22 research flights were grouped into six different types of flights (Figure 1), according to flight plans, as described in Crumeyrolle et al. (2010). The scientific objectives of the RF_Type_1 flights were to specify origins and regional characteristics of the air masses sampled at Cabauw. The RF_type_2 flights, close to the coastline, were performed to better understand the nucleation event occurring near the coastline of the North Sea. The goals of RF_type_3 / RF_type_4 / RF_type_5 / RF_type_6 aimed at studies related to aerosol properties along ‘quasi-Lagrangian’ flight tracks: West-East and North-South transects, most of the time in coordination with the British Bae-146, DLR -Falcon 20 flights and ground sites. Fourteen (RF42, RF44, RF47, RF49, RF51, RF52, RF55, RF56, RF57, RF58, RF59, RF60, RF62, RF63) of the 22 flights performed during the campaign were used in this study while the other eight flights were dedicated to observation of new particle formation events Crumeyrolle et al. (2010).”

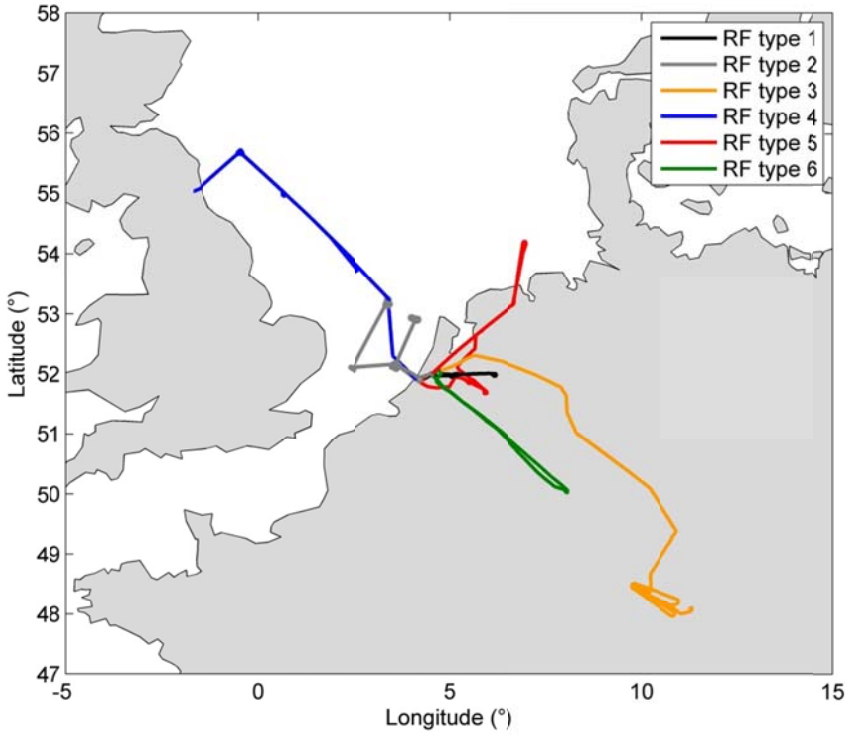

Figure 1: Different types of research flight (RF) plans performed during the EUCAARI intensive observation campaign in May 2008.

- p. 9455 l. 2, “isokinetic” includes isoaxial, because it means that one has the same velocity vectors (magnitude AND direction) in the free stream and the inlet tip. So, please remove the bracket term.

The referee is right. Indeed, when the isokinetic condition is fulfilled the isoaxial condition has to be fulfilled. Then, the bracket term has been removed.

- p. 9455 l. 8, depending on the, for the reader unknown, flight altitude the dp50 of CPCs shifts with operating pressure up to several nanometers. Was this the case for your flights?

During the EUCAARI campaign, the altitude range of the ATR-42 (see Table 1), between 0-5000m, was not as wide as other aircraft studies and thus the change in the cut-off diameter has been less important. However, the referee is right to highlight this uncertainty. Unfortunately, no laboratory measurements have been done with the CPCs used during the EUCAARI campaign. In order to take that comment into account, we corrected the sentence into : 'The 50\% detection diameter is larger than 3 and 10 nm for each CPC'

- p. 9455 l. 17, please provide a reference for the thermo-desorption column or described it more in detail, in particular the particle residence time in the column. Is it large enough that the volatile mass fraction had enough time to evaporate?

The thermo-desorption column with residence times and whole evaporation process has been modeled with FLUENT (Villani et al., 2007). Subsequent use of the thermodenuder has been presented in Matsuki et al. 2010; Quennehen et al. 2011) where the particles already have been heated up to 280^{O}C. Villani et al. (2007) described the design and calibration of the heating units for the conditioning of a selected aerosol sample while minimizing sample losses due to thermophoresis and diffusion. The column design was based on the modeling of the profiles of temperature and velocity and the behavior of a monodisperse aerosol in the heating units, using Computational Fluid Dynamics (CFD) Flow Modeling Software. This allowed for estimations of the heater dimensions and also calculation of the minimum length of heating tube needed to completely evaporate the aerosol particles at high temperature with sufficient residence time, as well as to cool the aerosol sample down to ambient temperature.

- p. 9456 l. 1, a residence time of 612 s or more than 10 min would correspond to a spatial resolution of about 61 km. How representative is this? By the way, 10 min seems to me unrealistic long, is this correct?

Again, the authors want to apologize for the compiling issues. In this case the '-' disappear in between the 6 and 12s. So it's not a 10 min period but a period in between 6 and 12 seconds.

- p. 9456 l. 24, is "Squirrel" commercial software? Please provide a reference.

Squirrel in the IGOR software used by the AMS Aerodyne community. It's a free software that you can download on the 'Jimenez research Group Wiki'. We changed the statement into : 'All flight data were treated according to procedures provided within the standard AMS analysis software 'Squirrel' (SeQUential Igor data RetRiEvaL version 1.48, Allan (2003)) implemented with Wavemetric's Igor Pro

(version 6.12) and in accordance to the standards defined and currently used by the Aerodyne AMS operators community at the time when these data were treated (http://cires.colorado.edu/jimenez-group/wiki/index.php).'

- p. 9457 l. 14, please specify what is meant with "small segments". Were the retroplumes only initialized when the aircraft changed altitude?

The segment sizes were variable as a function of each flight but they were in general not larger than 50km. This has been clarified into the manuscript. Retroplumes were initialized for a change in the position (latitude – longitude) as well as altitude.

- p. 9457 l. 25, the choice of the geographical sectors seems to me arbitrary. Please provide arguments why you did it in the way you did. And again, you are not the first on doing this kind of analysis, what does the literature tell which number and size of wind sectors makes sense? After explaining the choice of the sectors more in detail, please explain also more in detail how the air masses were attributed to the sectors (mathematical formalism?).

The choice of the sectors has been made as a function of :

1. Aerosol sources that vary a lot as a function of each sectors (Polar = clean, NE-EUR : Organics enriched, E-EUR : Industrialized areas, NW-EUR: industrialized areas)

2. Air masses frequency : The sector S-EUR was originally not that wide, but only few air masses were coming from the south. They were mainly coming from North-Africa and then went over the Mediterranean sea, Italy, East of France. The figure 2 (in the manuscript) does not appear anymore and has been replaced by the maps showing the geopotential and the wind at 850hPa (Figure 2 a and b in this document). Each air mass sector is now represented with an arrow and is thus less confusing than the blocks as it appeared in the former figure.

3. Observations made on the field (size distribution, number concentration, chemical composition). Indeed, during the post processing of the data we noticed large differences of the size distribution, number concentration and the chemical composition as a function of air mass sectors.

- Tab 1., the potential temperatures provided here are totally wrong, -159_C down to -216_C !!!

The referee is right, a bug has been found in the code used by the first author. The authors corrected the values in the table 1 :

Table 1. Averaged meteorological parameters observed in the boundary layer (BL) and in the free troposphere (FT) for different episodes during anticyclonic (HP) and cyclonic (LP) conditions. – = Not Observed

		RH (%)		Pot. T (K)		% of time occurring		ATR-42 Atitude range (m)	
		HP	LP	HP	LP	HP	LP	HP	LP
BL	NW-EUR	64	--	290.4	--	33	--	<1500	
	S-EUR	--	76	--	299.3	--	13		<1400
	NE-EUR	--	66	--	286.4	--	65		<1600
	E-EUR	45	22.5	297.2	292.5	67	19	<1600	<1200
	P	--	74	--	285.8	--	3		<1200
FT	NW-EUR	--	--	--	--	--	--		
	S-EUR	--	54	--	306.5	--	54		1600-3000
	NE-EUR	--	20	310.2	297.4	4	23	3000-5000	2500-3000
	E-EUR	43	67	300.5	306.8	96	18	1600-4000	2500-3000
	P	--	10	--	298.7	--	5		2800-3100

- p. 9458 l. 16, please provide two weather map figures displaying the differences in the meteorological conditions more clearly. You could and should use these maps also to show the major air mass pathways you distinguish. How representative are the two weather conditions (and hence the aerosol data) for the spring season over Europe?

The geopotential and the wind at 850hPa are represented on the figures below for 11 May 2008 (Anticyclonic situation) and the 21 May 2008 (Cyclonic situation). These pictures have been taken from the EUCAARI LONG Range EXperiment internet site (http://www.pa.op.dlr.de/aerosol/eucaari2008/).

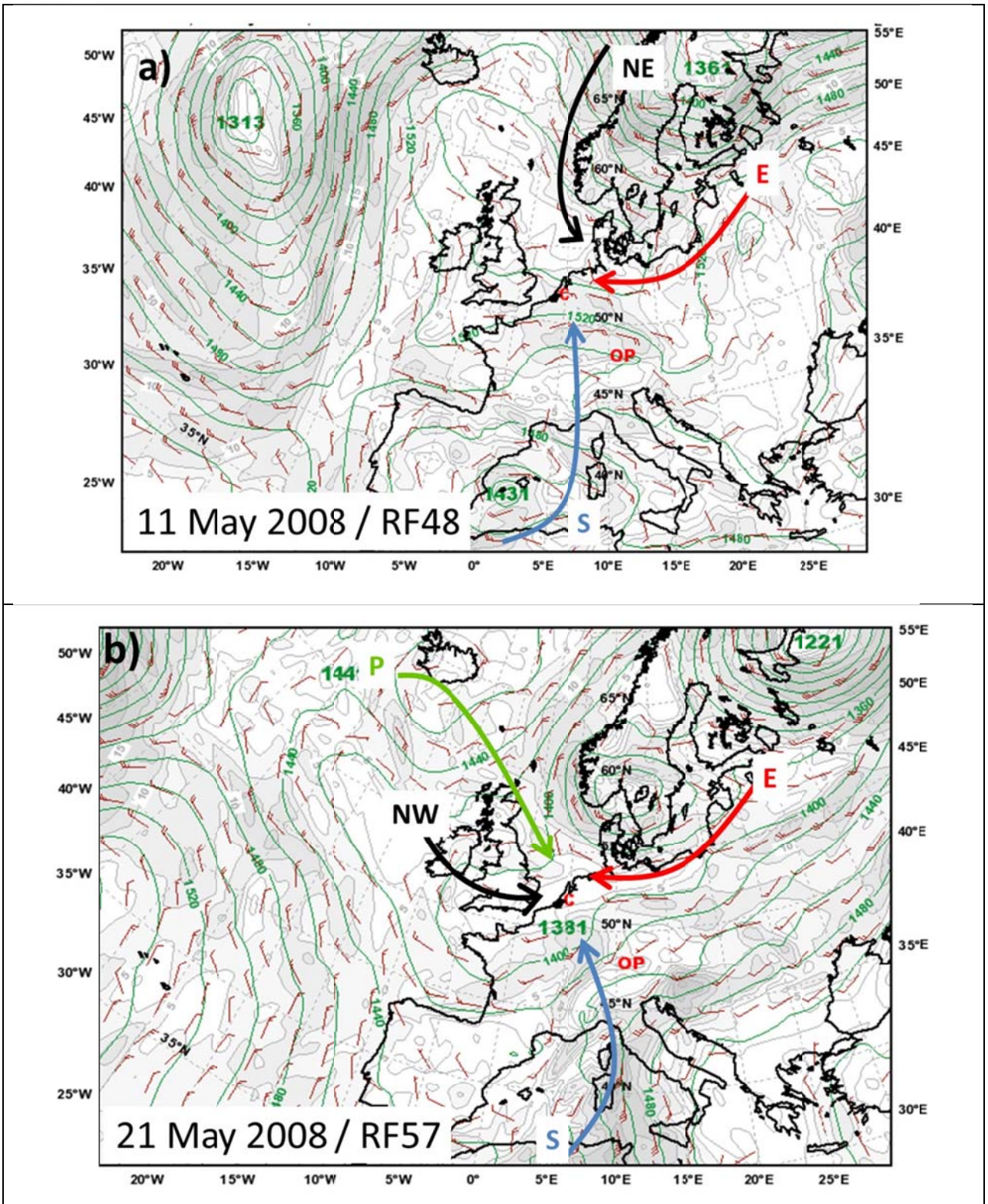

Figure 2 : Geopotential and wind speed at 850hPa for anticyclonic (a) and cyclonic (b) conditions. The arrows correspond to the origin of the air mass trajectories

- p. 9458 l. 26 the free troposphere reaches from the top of the boundary layer up to the tropopause. Were your measurements performed in this whole altitude range? Or are your data rather representative for the lower free troposphere? If yes please make this clear in the text and in the figures as well.

In our study the free troposphere correspond to the layer over the top of the boundary layer until the maximum of ATR-42 flight altitude (<6000m). This layer will thus correspond to the lower free troposphere. This statement has been revised throughout the manuscript.

- p. 9459 l. 5, again, the given potential temperature cannot be correct.

This has been corrected.

- p. 9459 l. 11, where does this statement come from? You did not provide any proofs that the air mass was loaded with particles in these regions.

Again here, the first author would like to apology for the LATEX compiling issue. This sentence was not supposed to appear here.

- p. 9460 l. 2, it is hard to follow the discussion, because there are so many parameter names. You would help the reader if you would indicate the weather situation (LP/HP) and measurement altitude (BL/FT) directly in the graphs of Fig. 2 (also Fig 3, ⋮ ⋮ :). Moreover, if you use LP as synonym for the cyclonic conditions, please be so consequent to use it everywhere, also in the figure captions, e.g., Fig. 2.

The authors agreed that the number of parameters make the reading a little bit tough. Thus, we modify the figures and all the captions as suggested by the referee.

- p. 9460 l. 6, the E-EUR sector is relatively small, hence the variability of the aerosol in this sector should be “small”, compared to other sectors. But it is0t and this is probably due to the statistics. Your measurements cover more and different meteorological situations; hence there is a larger variability in both the meteorological parameters and the aerosol. And consequently, how representative are the measurements for the NE-EUR sector with only 4% of measurement time in the FT under HP? You should set and name statistical requirements, e.g., at least ten flight hours during three different days, in order to get at least a minimum statistical significance. Otherwise the comparison of different sectors

makes little sense. By the way, did the sectors change their size from flight to flight? This is at least what you suggest when talking about the “variabilities of the sector the air mass is originating from”.

The E-EUR sector is the smaller one but it’s also the sector the most represented in term of percent of time occurring. The referee is right to say that statistically the comparison of different air mass sectors with such a different occurrence time may not be relevant. Thus, we will compare the air mass sectors only when the % time of occurring will be comparable.

The sectors don’t change from flight to flight. This sentence is modified to : ‘Due to the high occurrence time of the E-EUR sector (96$\%$), the measurement period cover different meteorological conditions (wind speed, cloud presence, etc...) and different aerosol source strength (week days, week end days).’

- p. 9460 l. 27, I do not see “generally trimodel” size distributions in Fig. 3 d

The sentence have been modified to: ‘Three over the six size distributions observed in the BL during the whole campaign are trimodal Table 3 , Figure 4), composed of a nucleation mode D_{Nucl}<30 nm, an Aitken mode (30 < D_{Aitken} < 60 nm) and an accumulation mode (100 < D_{Acc} < 175 nm), and bimodal in the FT, composed of an Aitken and an accumulation mode.’

The trimodal size distribution in figure 3d is the black line labeled NE-EUR.

- p. 9462 l. 17, I might have missed it, but why is there no bar for the measurements with polar origin and cyclonic weather conditions (as there are data e.g. in Fig. 2)?

Unfortunately, the AMS was not working properly during these periods. Thus, we could not provide any chemical compositions for the polar air masses.

- p. 9463 l. 12, again, how do your measurements compare to previous aircraft studies, e.g., Morgan et al., ACP, 2009 or Pratt and Prather, JGR, 2010?

A comparison of the measurements shown in this manuscript with the study suggested by the referee as well as other study has been done throughout the manuscript. Here are the sentences added into the manuscript :

‘The aerosol relative and absolute chemical compositions of Group I is similar to the one observed in or around a highly polluted urban area (Mexico city) during the Milagro airborne campaign (DeCarlo et al., 2008). Organics are the major component (about 50%) and nitrates are the second most important component (20%) consistent with previous studies performed in urban area (Europe: Putaud et al., 2004; Toronto: Jeong et al., 2011) as well as in remote area (Germany: Hock et al., 2008; West of UK: Morgan et al., 2009).’

'The air masses originating from S sector have the highest mass concentration (29 μg m^{-3}) and the highest refractory material level (5.24 μg m^{-3}) consistent with the highest aerosol volume concentrations in the polluted air masses. In particular, the absolute concentrations of organics reach 18 μg m^{-3} corresponding to values generally observed in different urban areas like Pittsburgh (Zhang et al., 2005), Mexico City (Volkamer et al., 2006) or Zurich (Lanz et al., 2007) and 5 times higher than those reported by Morgan et al. (2009) over the United Kingdom. These high concentrations (total as well as organics) are related to the transport of dust over urban areas (Falkovich et al., 2001). Dust particles are mixed with highly polluted air masses (i.e. high concentration of organic gases) leading to the adsorbtion of gases onto dust particles.'

'The profile of the chemical composition for both meteorological conditions show a decrease of the relative fraction of organics and nitrate with altitude, compensated by an increase of the sulfate fraction. This finding is consistent with previous airborne studies over Europe (Morgan et al., 2009) and over Wyoming (Pratt and Prather, 2010).' The profile of the chemical composition will be provided as supplemental products. Indeed, the main goal of the manuscript was to do an overview of the aerosol properties as a function of the synoptic conditions and of two main layers (LFT and BL). The profiles of chemical concentration do not provide any distinction between the BL and the LFT.

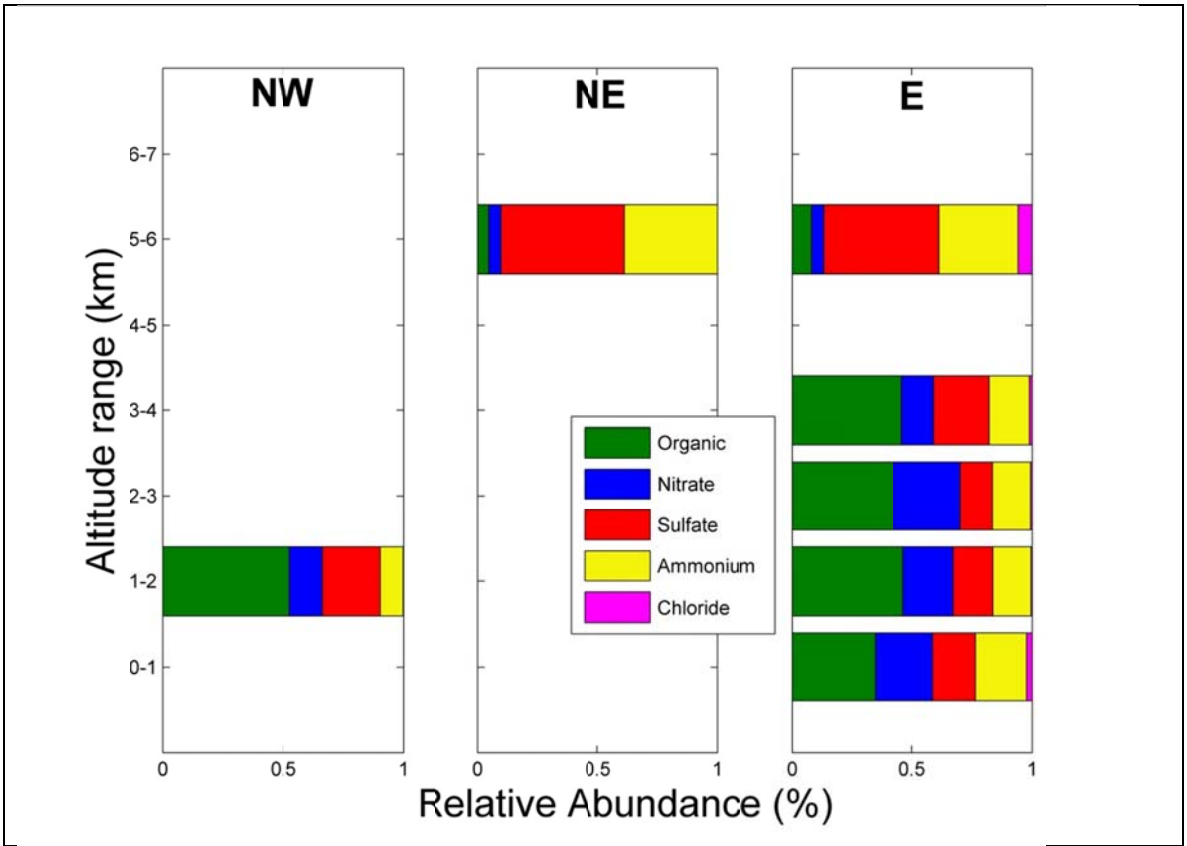

Figure 3 : Profile of the average relative concentrations of non-refractory submicron aerosol as measured by AMS as a function of the air mass origins (NW : North West Europe, NE: North Europe, E : East Europe) during anticyclonic conditions.

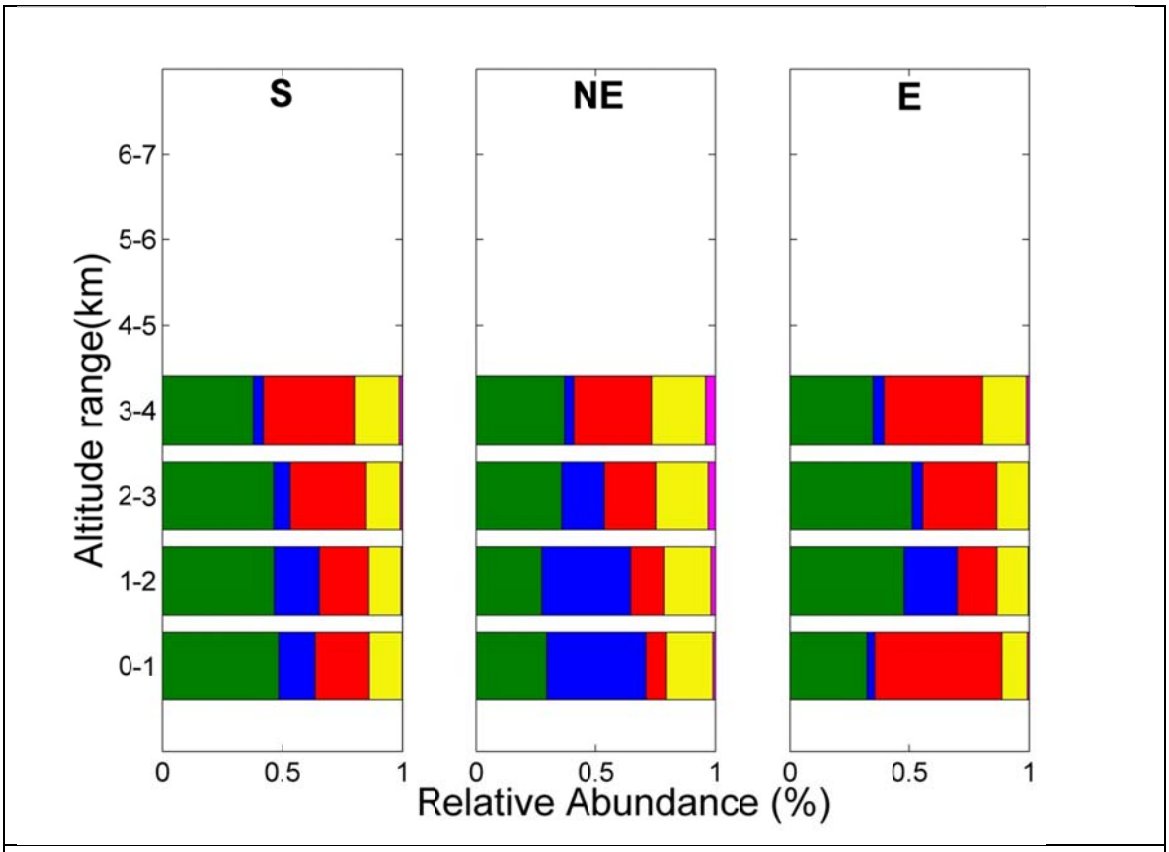

Figure 4: Profile of the average relative concentrations of non-refractory submicron aerosol as measured by AMS as a function of the air mass origins (S: South Europe, NE: North Europe, E : East Europe) during cyclonic conditions.

- p. 9465 l. 7, 50 nm as activation diameter for the rather clean FT might be OK, but in the BL, where are many more and larger particles this value seems to be low, 100 nm seems to me more appropriate. Would 100 nm as minimum activation diameter change your results?

If the activation diameter is set at 100nm in the boundary layer, then the results change significantly (Figure 1). During high pressure conditions, the CCN/N_{100nm} 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_{100nm} ratio is including in 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.

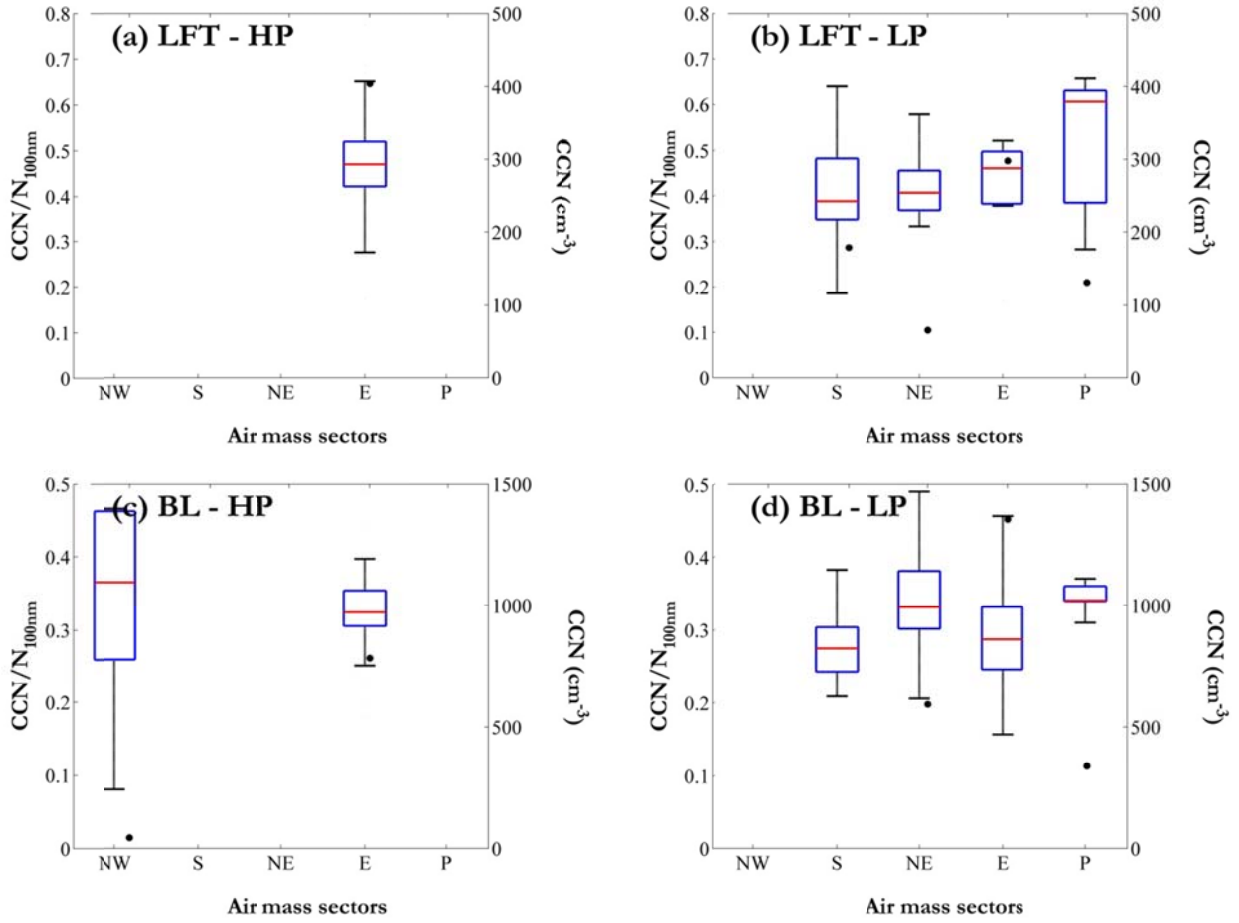

Figure 5 : CCN/N_{100} ratios as a function of the air mass origin origin (NW : North West Europe, S: South Europe, NE: North Europe, E : East Europe, P : Polar) in the lower free troposphere during anticyclonic (HP, a) and cyclonic (LP, b) conditions as well as in the boundary layer during anticyclonic (HP, c) and cyclonic (LP, d) conditions.

- p. 9465 l. 15, I wonder about the rather constant CCN/N50 ratio in the BL. The particle size distributions (Fig. 3) are different, hence I would assume that the CCN/N50 ratios also differ. And it is unlikely that the effect of the size distribution is balanced by the chemical composition plus dynamics to result in the same ratio for so many cases.

As the referee is right and as we changed the figures (see comment above), this sentence will not appear anymore in the manuscript.

- p. 9468 l. 1, sorry, I didnot get the message of this paragraph about the Angström exponent and the asymmetry parameter, Iom too tired

As this section of the manuscript did not improve the understanding of the manuscript, the authors decided to remove definitely this part.

- p. 9468 l. 21, the “Conclusions” are only descriptive, what and which values were measured. What is missing is if there is something new in the data, something unusual, a new observation, or a new conclusion. Please invest some more time into the data analysis.

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. Especially the measurements performed in the FT troposphere are rare in this location and of great interest for climate modelers. 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.

The conclusion has been corrected:

“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 predominent 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. "

- p. 9461 l. 7, how do the measured size distributions and fit parameter compare to the ones presented by e.g., Petzold et al., 2002? What is similar, what is different and if so, why?

The size distributions shown in Pertzold et al. (2002) are coming from PCASP and FSSP measurements. The measurement principle and the location of these instruments are totally different from the SMPS. Indeed, 1) the PCASP and FSSP are usually located on the wings while the SMPS is usually in the cabin; 2) the PCASP and FSSP are reporting optical equivalent diameters while the SMPS are reporting mobility diameter. Moreover, the diameter range of the SMPS is limited to 10-700 nm while the diameter range of the association of the PCASP and the FSSP is starting at 100 nm up to 20 µm. All these instrumental differences make the comparison harder. The mean geometrical diameter comparison, in the common diameter range, of this study and Pertzold et al. show strong differences. Indeed, the mean geometrical diameters observed by Pertzold et al. in the boundary layer are larger than 180 nm while they are not larger than 164 nm in our work. The same tendency is observed in the free troposphere where the Pertzold et al. mean geometrical diameters are larger than 200 nm while in this work they are lower than 175 nm. The similarities between these SD are that the concentration is generally (except particular case) larger in the boundary layer than in the free troposphere and the mean geometrical diameter is larger in the free troposphere than in the boundary layer.

Unfortunately, the authors didn't find similar measurements of SMPS size distribution (SD) measured onboard a research aircraft over Europe. Thus, we can only compare the measurements performed in the boundary layer with those measured at ground base stations. Birmili et al (2001) is using a similar air

mass classification than what is done in this study. By comparing the SD, we observed similarities on the size distribution shapes: trimodal size distributions (with an Aged nucleation, Aitken, Accumulation mode), on the concentration magnitudes and also on the width of the lognormal except for the aged nucleation mode. Indeed, we observed sharper mode than Birmili et al. (2001). This difference may be due to a source of precursors which was further or different wind conditions that transport freshly emitted particles faster or slower. Part of this discussion has been inserted in the manuscript.

		Aged nucleation mode	Aitken mode	Accumulation mode
D (nm)	Birmili et al (2001)	13-20	45-88	150-250
	This work	23-28	33-95	112-164
Concentration (cm-3)	Birmili et al (2001)	980-4000	1400 - 9100	320-2400
	This work	186 - 3540	327 - 5900	138-1905
Sigma	Birmili et al (2001)	1.49 - 1.75	1.47 - 1.86	1.44 – 1.65
	This work	1.16 - 1.34	1.35 - 1.88	1.49 - 1.71

- p. 9480 Tab 2., the “0” in the N500 particle concentration in the free troposphere should be 0.1 or 0.01 or whatever, but there are at least some particles. And please use a space between the particle diameter and the respective unit, e.g., “10 nm”.

As we are now using the N_{100} for the CCN/CN ratio, the authors decided to show the N_{100} instead of N_{500}.

- Fig. 2, 5, and 7: the provision of 0% and 100% percentiles might be misleading because how sure can you be that there are no outliers in your data? Wouldn’t it be better to provide the 5% or 10% (95% or 90%) percentiles?

All the box plots presented in this manuscript are now plot with lower and upper limits corresponding to 25% and 75% percentiles, bottom and top whiskers the 5% and 95%.

All the small technical comments of the referee were taking into account in the manuscript. One more time the first author would like to apology for the latex compiling issues that causes a lot of incomplete sentences.