

***Interactive comment on* “Variability of aerosol, gaseous pollutants and meteorological characteristics associated with continental, urban and marine air masses at the SW Atlantic coast of Iberia” by J.-M. Diesch et al.**

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Review of the paper entitled "Variability of aerosol, gaseous pollutants and meteorological characteristics associated with continental, urban and marine air masses at the SW Atlantic coast of Iberia" by Diesch et al., ACPD.

This work presents simultaneous aerosol and gas phase pollutant measurements at the station of "El Arenosillo", conducted during a campaign within the DOMINO project. Its main added value comes from the fact that it gathers and combines a large ensem-

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ble of high resolution measurements, both of physical and chemical characteristics of aerosols as well as of gaseous species. Disadvantages rise from the very short duration of the campaign, which cannot probably be representative for the intensity and specific features of pollution transport over the area throughout the year, and the accuracy of the methodology followed to distinguish the different transport patterns. Despite these constrains, to my opinion, this work merits publication to ACP, after strengthening parts of the work that relate to air mass classification. Please find below suggestions for improvement and corrections that should be implemented in the text prior to potential final acceptance.

Author: We are grateful to the reviewer that he or she appreciates the value of our work, which has already proven to be valuable to colleagues in- and outside the DOMINO consortium due to the facts that already several manuscripts have been submitted, some of those are even in press, or are under preparation that use the data presented in this work and cite this manuscript.

1) Abstract, pg 31587, In 6-8: These lines referring to ozone variability are too generic and are possibly valid everywhere in the globe ... so I suggest removing it from the abstract.

Author: Since one of the purposes of this manuscript is to provide an overview over our gas and particle phase measurements during DOMINO for potential users of this detailed data set we decided to keep the discussion of ozone variability within the manuscript. In addition, this discussion shows very clearly the different impacts of diurnal effects and air mass history. To strengthen this further, we specified the ozone characteristics in the abstract as follows: “A significant variability of ozone dependent on the different air mass types was also observed which is additionally strongly determined by the solar radiation”.

2) Pg 31587, In 18-21: The significance of African dust transport during the summer months is true for the Western Mediterranean Basin and not for the whole South Eu-

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rope. For instance, in the Eastern part dust transport is encountered mainly during spring. See Moulin et al., JGR, VOL. 103, NO. D11, PAGES 13,137–144, 1998.

Author: Thank you for this additional information – we removed the related sentences from the text twice but kept it once in the text as a reviewer in an earlier stage of the manuscript review process has requested this.

3) Pg 31589, ln5-7: "Our contribution to ... parameters simultaneously" This sentence needs restructure from the grammatical point of view.

Author: We corrected the sentence as following: "Our contribution to this project is the investigation of the aerosol particle chemistry, composition, formation and transformation processes. Therefore, we measured a large number of atmospheric parameters simultaneously."

4) Section 2.2: At this particular part of the manuscript my main concerns are born. In particular, even though back trajectories are surely one step forward compared to simple wind direction classification for medium to long range transport, it is yet over-appreciated by the authors concerning its validity to distinguish air masses from closer distances. The limitations of this methodology should be clearly stated and at certain points could be empowered by additional proofs. Please consider the following relevant points:

-The authors should go through the HYSPLIT site and relevant publications (e.g. Draxler and Rolph, 2003) and document the models limitations. To my knowledge trajectories below 100 m suffer from high uncertainties. For the current study I would suggest the choice of one back trajectory inside the boundary layer- BL - (e.g. 500 m) and one outside the BL (e.g. 1500 m). That would additionally enable authors to support their air mass classification regarding inside BL transport, possibly from close sources, and free tropospheric transport. Coincidence of the two trajectories would increase the confidence regarding the sector that air masses originate from.

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Author: Extensive and systematic investigation of the local meteorology at the measurement site and broad sensitivity studies of HYSPLIT back trajectories for this site and for the time interval of the DOMINO campaign (at 10-1000 m arrival height) (J.A. Adame Carnero, EGU 2010, manuscript in preparation) have shown that for the time interval of the DOMINO campaign atmospheric transport in the measurement area was dominantly driven by the synoptic situation and not by mesoscale processes (J.A. Adame Carnero, Atmos. Chem. Phys. Discuss., interactive comment, 11, C14767-C14769, 2012) and that variation of trajectory arrival height did not result in significant differences in trajectory pathways. The conclusion of these extensive studies is that, even though HYSPLIT has general limitations for the calculation of local transport processes and at low heights, the trajectories used in our work to separate source regions actually reflect atmospheric transport for that time pretty reliably. Therefore, after going through the HYSPLIT site and relevant publications the following lines were implemented in the manuscript: “HYSPLIT is intended for transport processes on larger spatial scales due to its relatively low grid resolution. Especially for lower trajectory altitudes the model suffers from severe limitations. Therefore, under the influence of mesoscale processes, HYSPLIT is not able to reproduce local meteorology adequately. However, during the DOMINO campaign synoptic conditions dominated also regional transport which can therefore be reproduced with sufficient accuracy down to the lower boundary layer by HYSPLIT calculations as was shown by thorough sensitivity analyses and comparison to measurement data (personal communication, J. A. Adame Carnero).”

-What is for sure is that back trajectories cannot identify air masses coming from a narrow domain like Huelva, since their spatial uncertainty exceeds by far the extend of the city. In this case, trajectories can give just a first indication that should be additionally certified in two ways: one is the wind direction from the meteo station provided that there are no physical blocks in between El Arenosillo and Huelva (distance 20km), and the other is for these cases possibly influenced by Huelva pollution, to run forward trajectories with Huelva as starting point and at various altitudes (mostly inside the BL)

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and check whether indeed El Arenosillo is among the receptor points. In all cases combination of methods and statistical support would increase the level of confidence concerning air mass origins.

Author: Both suggestions to increase the level of confidence concerning air mass origins were further examined. There are no significant blocks between El Arenosillo and Huelva so the air transport is not redirected by local topography in this area between the sources and the station. Secondly, forward trajectories were calculated starting at Huelva at different heights. We found that many of these trajectories pass the measurement station. In addition, the agreement of locally measured wind directions with trajectory arrival directions – as presented in the manuscript – further increases the level of confidence concerning the categorization of air masses. For this reason and due to J. A. Adame's extensive and systematic investigation of the local meteorology at the measurement site and broad sensitivity studies of HYSPLIT back trajectories for this site during the DOMINO campaign we are confident that the classification made on basis of the back trajectories in this work actually reflects atmospheric transport for that time. In addition, the results presented in our manuscript show reasonable differences for the different source categories, also indicating that the separation into these categories is valid. Further evidence for this fact is provided by a further manuscript also submitted to ACP. In this paper a combination of back trajectories and local meteorology was used to separate different types of nucleation events. The data clearly show that the separation works well and that reasonable and significant differences are observed in the different types of nucleation events.

-Another issue is how authors have attributed back trajectories to a certain angle direction in order to compare it with local wind direction fields. That should be clarified since good agreement between the two methods actually strengthens their conclusions.

Author: Yes indeed. We have written in the text more clearly how we compared the two methods: "Fig. 2 shows a map including back trajectories classified into 6 air mass categories based on air mass origins and pathways. The resulting air mass

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categories with the corresponding angles of the limiting trajectory arrival directions as well as the associated measurement time in percent of the entire measurement period are: “Seville” (65°-82°, 6%) - “Continental” (340°-65°, 15%) - “Portugal+Huelva” (310°-328°, 3%) - “Marine+Huelva” (285°-310°, 18%) - “Portugal+Marine” (265°-285°, 15%) - “Marine” (140°-265°, 6%).” And: “In Fig. 3 locally measured wind direction data were compared for both classification methods. The three different wind direction ranges of the “Continental” (340°-110°), “Urban” (285°-330°) and “Marine” (140°-265°) sectors are shown as grey shaded areas. Red colored box plots reflect the local wind directions measured during the arrival times of the back trajectories associated with the various air mass categories.”

-In Fig. 4 and on 28/11/08 there seems to be an interesting case of an event during which all parameters peak. This event is not classified into any of the existing classes. Is it a case that falls between other classes and the authors cannot distinguish? Is it a case of stagnant conditions? In the first case an attempt to classify it a posteriori based on its "pollution" characteristics would be interesting, while in the latter case the addition of the "stagnant conditions" class would probably be appropriate.

Author: As shown in Fig. 4, this event occurred when there was an air mass change between “Seville” and “Marine”. Trajectories for this time period show a complex curved path and an additional trend, moving further to more westerly directions from hour to hour and therefore were not classified into one of the categories. However, the trajectories indicate that they pass Huelva before reaching the measurement site, likely causing the high concentrations. This was also implemented in the manuscript as follows: “As also shown in Fig. 4, the highest concentrations observed on Nov 28, 2011 were not considered in the study due to an air mass change occurring during this time. Trajectories for this time period show a complex path and a trend, moving further to the West from hour to hour. Therefore, they were not classified into one of the categories.”

5) Pg 31600, ln 13: Some of the CPC error bars are missing in Fig 6 thus the discussion

cannot be easily followed. In lines 18-20, I do not understand the argument why error bars are not presented in the graph.

Author: For a better understanding and as suggested, we added all error bars in Fig. 6 and removed the sentence. New particle formation events that occurred during the campaign lead to a large variation in particle number concentrations. This feature is explained and figured in the manuscript recently submitted to ACP. On the other hand, large number concentration variability is not an abnormal feature as due to different sources within a certain air mass source category the number concentrations can differ strongly. E.g., in the marine category generally low concentrations due to wide absence of sources were measured. However, when ships passed along the coast, extremely high particle number concentrations can be observed which result in a large particle number concentration variability.

6) Pg 31600, ln 20-23: The comparison between the different classes is not clear.

Author: Sorry for the misunderstanding. We reformulated the passage as follows: “Despite the strong variations in number concentrations, a general trend is clearly visible and major differences occur between the group of “Continental”, “Marine+Huelva” and “Portugal+Marine” air masses compared to the group of “Seville”, “Portugal+Huelva” and “Marine” air masses: The ratios of particle number to particle mass concentration bar heights in Fig. 6 differ significantly between these two groups of air masses.”

7) Pg 31601, ln 16-19, and caption in Fig 7: The authors present size distributions from two different instruments based, as they also mention, on different techniques, thus providing results that are not comparable both by means of diameters but also on the absolute amplitude of the observed aerosol modes. The discrepancies in the overlapped area is not due to the fact that the instruments reach their limits, as they say in Fig. 7 caption, but due to this correction which also affects the measured quantity since there is a change in the integration interval of $DN/D\log(D_p)$. I would suggest they use their full chemical data set and current relevant literature to infer on the optical

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properties of aerosols in that range, proceed to diameter type homogenization, based on well documented assumptions if necessary, and then readjust OPC data to FMPS, in order to provide a continuous reliable distribution. Another option would be to discuss each mode separately and not mix between number, surface and volume distributions.

Author: Very likely, the optical instrument reaches its limits in the first one or two smallest channels. Our first priority would also have been the diameter type homogenization before submitting the manuscript to ACPD. However, we do not have the full chemical information (we cannot measure refractory species like sea salt and for example the black carbon size information, especially we do not have any composition information for the upper sub-micrometer and the super-micrometer size range) needed for the OPC conversion. Therefore, we cannot execute the relevant calculations. As suggested by the reviewer, we decided to separate Figure 7 in (a, on the left) showing the FMPS size distribution and (b, on the right) showing the OPC size distribution for the particle number, surface and volume distributions and discuss each mode separately.

8) Pg 31602, 1st paragraph: In the discussion of number concentrations, I would like to mention that nucleation is also found for the Portugal-Marine case. Additionally, if Huelva is the reason why Portugal-Huelva distribution shows the maximum number concentration at 30 nm then why isn't this also the case for Marine-Huelva? What is the role of pollution from Portugal?

Author: For both, "Continental" and "Marine+Huelva" categories we clearly identified new particle formation events. Therefore, we were able to subtract the particle number concentrations originating from these events from the total size distribution as shown as dotted lines in Fig. 7. For the "Portugal+Marine" category we are not able to identify nucleation events unambiguously, but we also cannot exclude that they occurred. As mentioned in a second manuscript which deals with the new particle formation events observed during the DOMINO campaign and which was recently submitted to ACP, we found nucleation occurred over the continent and emissions from Huelva superimpose the new particle formation events. To make this clearer we added in the text: "For the

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“Portugal+Marine” category we also measured a mode around 10 nm but we can neither identify nor exclude unambiguously new particle formation events for this source category.”

9) Section 3.4.3: Authors base their discussion and interpretation of particulate organics diurnal patterns on the mean diurnal course. Is this for the whole period? Is the pattern the same when different air masses are encountered? The high error bars indicate that during different days much different patterns might be observed.

Author: The interpretation of particulate organics is based on the whole campaign dataset as also mentioned in the paper. It is obvious, that day to day and air mass to air mass differences also cause differences in the concentrations of the organic aerosol types and therefore large relative standard deviations. We have investigated diurnal patterns also for each of the categories separately. However, this did not lead to additional conclusions and is not possible to discuss in the framework of this publication.

10) Section 3.5: The whole analysis and discussion on ozone behavior seems to be detached from the rest of the document. Moreover, the analysis depth does not comply with the respective analysis for aerosols, suffering in many point from generalizations and lack of interpretation depth. I do not see how this section can add something to the paper and I suggest it is removed. In all cases, the paper is way too long to follow undistracted.

Author: This manuscript represents an overview of our gas and particle phase measurements during the DOMINO campaign and discusses the variability of these parameters dependent on air mass origin. In the analysis of the gas phase species we found a clear dependence of ozone regarding the air mass categories and the solar radiation which can be nicely presented in Fig. 10. In addition, the presentation of the gas phase data are also important for other DOMINO campaign members and colleagues outside this consortium, which cited the paper in their manuscripts presented in ACP or other

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journals. However, as suggested by the reviewer, we shortened the manuscript in the “Discussion & Summary” section as detailed below.

11) Section 3.6: What is here meant by "relative" standard deviation"? Is the fact that inner-category variability is larger than the inter-category an admission of biased classification methodology? I wonder whether these two standard deviations are comparable, since the conclusions potentially drawn could have severe effect on the validity of the presented results.

Author: Sorry for the misunderstanding – in both cases, for the inner- and inter-category variability we used the relative standard deviation (i.e. the standard deviation of the absolute values divided by the average). The separation in the different categories was done with an independent method (HYSPLIT) and is therefore not biased for this reason. “Although the inner-category variability is often larger than the inter-category variability, which is calculated over the whole set of source categories, there are significant differences between individual categories for individual parameters.” – as added in the “Discussion&Summary” section. As there are significant differences for several parameters, we think that the separation in the different categories is valid.

12) I would strongly urge the authors to shorten Section 4 by maintaining only major findings, possibly better under discrete bullets.

Author: We have shortened section 4 by completely cutting the comparison of our results with those obtained from different measurement campaigns. Therefore, we were able to remove Table 4. Now, the manuscript is one page shorter and the results were discussed and summarized in the same detail as before.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 31585, 2011.

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