The authors would like to thank both reviewers for the very constructive and helpful comments which are important to improve the manuscript. In general we have followed most recommendations including improvements on the language. In our opinion, this has resulted in a manuscript that has considerably higher quality. Corresponding to the relatively long list of questions and suggestions for corrections, we have implemented numerous minor changes throughout the manuscript. We sincerely hope that the editor and the reviewers will be satisfied with the improvements and will consider the manuscript to be published in ACP. Furthermore, due to the extensive and very constructive review by both reviewers, then the authors sincerely wish to acknowledge the two anonymous reviewers for their constructive reviews.

Please find below our replies to both the general remarks and the specific comments by both reviewers

#### **Reviewer 2**

#### General comment by reviewer 2:

In this study, the authors investigate the main source of Alternaria fungal spores in Copenhagen. They sampled air with the Hirst spore trap on a bi-hourly basis for a period of 10 years. Data analysis using the HESPLIT back trajectories model. The main conclusion of the authors is that most of the time the source of the spores is local agriculture activity and that long distance transport occur only occasionally. The authors also investigated the emission flux of spores by analysis of samples collected from the exhaust air of a harvesting machine. The results of this study can potentially have a potential for forecasting fungal spore concentrations in the studied area and might have important implication to asthmatic and allergy patients. Yet, there are some crucial points that need to be addressed and they are listed below. At the current stage the paper lacks in several areas, as detailed below. General comments are that standard deviation and error bars should be added to all graphs. Apart of the scientific comments, the level of presentation and the English used in the text are not uniform and must be corrected. Some specific comments are listed below.

#### Reply:

The reviewer does not seem to be familiar with the observational tool: The Hirst pollen and spore trap that is used within the European network on aeroallergens at more than 500 observation locations. The reviewer has raised a number of requests and suggestions that unfortunately cannot all be met, due to technical limitations of the observational methodology.

#### Specific comment by reviewer 2:

Title The main finding in the article is that the source of the spores in Copenhagen is local in most cases. The title does not reflect this concept.

#### Reply:

We agree with the reviewer. The title was chosen as the current belief in our area is that Denmark forms the Northern Boundary of central Europe. However, Encyclopedia Brittanica states that the northern boundary of central Europe is Germany and Poland. We therefore suggest the following title instead:

"Crop harvest in Denmark and Central Europe contribute to the local load of airborne *Alternaria* spore concentrations in Copenhagen"

### Specific comment by reviewer 2:

**Abstract** 

1. P. 14330 line 6 - change "alternaira" to "alternaria".

### Reply:

The correction has been implemented as requested

#### Specific comment by reviewer 2:

2. The authors do not mention which alternaria spores were checked. There are many alternaria species and not all of them cause health effects. The one that mostly effect human health is alternaria alternata and it can be only a small fraction of the entire specie. This should be discussed and weighted in when talking about the total population and the resulting health impacts. Without this, it is impossible to assess the health impacts.

#### Reply:

The observational method in the Danish as well as the European monitoring programme rely on visual identification of pollen and fungal spores at the genus level. This means that this method cannot identify *Alternaria alternata*. However, the threshold of 100 spores/m³ is based on the methods that include both a slit sampler (for growing colonies and subsequent identification) and a Hirst trap for providing atmospheric concentrations (Gravesen, 1979). As such the threshold of 100 spores/m³ includes both allergenic and non-allergenic fungal spores from the genus Alternaria.

In order to clarify the observation method, then we have added the following sentences to the method description on line 24:

"In the monitoring programme, *Alternaria* spores are identified at genus level and counted at 640x magnification on 12 transverse strips for every two hours. The total area of investigation corresponds to 9.75% of the total sample. "

## Specific comment by reviewer 2:

3. The term "clinically relevant" is not a very common term in the literature; please consider changing it throughout the text to concentration that might induce health effects.

We disagree with the reviewer on this particular subject. The term "clinical relevant" has for at least a decade been used in scientific literature (Cecchi et al., 2007). This term is used in connection with observations from the Hirst trap for a specific subset of aeroallergens: Those aeroallergens where there exist clinical based thresholds (e.g pollen from birch and grass and fungal spores from *Alternaria* and *Cladosporium*). So to maintain this tradition, we suggest to keep this particular formulation

#### Specific comment by reviewer 2:

4. The claim that although fields were treated with fungal infections and still fungal spores were found is not clear. Which fungi species were treated? All of them? Maybe some of them are resistant or at least the spores are resistant.

#### Reply:

We fully agree with this comment and have taken the opportunity to rewrite the manuscript as follows.

### Originally submitted version:

Visual inspection of the fields revealed that none of the crops displayed signs of fungal infection. The fields were treated with fungicide and the farmer reported that he had used a recommended application scheme of chemical fungicides and therefore considered the crops to be free of fungal diseases, including *Alternaria*.

#### Re-written version:

Visual inspection of the fields revealed that none of the crops displayed signs of fungal infection. The barley fields had been treated against fungal infections by spraying with pyraclostrobin and tebuconazole on June 15<sup>th</sup>, 2011. The wheat fields had been treated against fungal infections by spraying with propiconazole on April 18<sup>th</sup>, 2011, with pyraclostrobin, epoxiconazole and boscalide on May 25<sup>th</sup>, 2011 and again with epoxiconazole and boscalide on June 16<sup>th</sup>, 2011. All applications were according to manufacturers' and agricultural advisors' recommendations, targeting the fungicide resistance spectra of local fungal pathogens. The fungicides used were neither targeted at, nor are claimed active against Alternaria spp., although it cannot be excluded that the fungicides used in the study fields initially had an inhibitory effect on Alternaria spp. Even though Alternaria triticina has been reported to cause fungal infections in wheat in India (Singh et al. 1998) and Argentina (Perelló and Sisterna, 2005), in Europe the economic effect of Alternaria spp. is considered insignificant in barley (Gannibal 2008) and rare in wheat (Gultyaeva 2008), making their chemical control unnecessary. During maturing and senescence of crop plants, prior to harvest, the earlier applied systemic fungicides will cease to have an effect, which indeed is a regulatory condition for their use. Therefore, as Alternaria spp. are common to the environment, having a role in the decay of organic matter (Kirk et al. 2008), any Alternaria spores found in this study most likely will reflect the normal course of fungal invasion of grain crops during early summer, occurring in most or all fields of central and northern Europe, where moist conditions occur intermittently during the weeks prior to harvest.

#### Specific comment by reviewer 2:

Introduction

1. P.14331 line 5 – change "provides" to "provide".

### Reply:

We disagree with the reviewer. The correct sentence is as it is written here "The disadvantage of the Hirst trap is that it typically only provides .." and not "The disadvantage of the Hirst trap is that it typically only provide ..". So we have left the sentence as it is.

### Specific comment by reviewer 2:

2. Why does the Hirst trap only provide observation of fungal spores in the genus level?

Isn't it a matter of the analysis? Is it possible to perform PCR for samples collected with the Hirst trap? If so, it would be interesting to investigate which fraction of the alternaria spores are relevant for human health.

#### Reply:

The reason is that the slides from the Hirst trap are visually analysed using a microscope . This methodology provides bi-hourly concentrations but only at the genus level. Another methodology for quantification of atmospheric concentrations is to use the allergens within the fungal spores (or pollen) to identify individual species. However such a methodology requires much larger samples in order to get a signal. This has recently been highlighted by Buters et al (2012). They needed a high volume sampler (800 l) and allergen analysis in order to have enough material for a daily mean value of airborne allergens, which is the highest temperal resolution this method can provide – at a much higher cost. The methodology that uses allergens does not allow for bi-hourly concentrations, which is a requirement in the protocol we have used to analyse the observations. Similarly, the qPCR methods by Burshtein et al (2011) or Lang-Yona et al (2012) have even coarser temporal resolution than the observations by Buters et al (2012). These differences in methods and their advantages/disadvantages are highlighted in our introduction.

We have therefore added following sentence to the methodology after "total sample":

# This area and the flow rate of the fungal spore trap can be used to convert the spore count into bi-hourly concentrations or daily mean concentrations."

#### Specific comment by reviewer 2:

3. P. 14331 line 12 – change "rare" to "rarely".

This change will change the meaning of the sentence. Instead we have modified the sentence so that instead of following: "Despite these advantages, data of fungal spores from Hirst traps are rare compared to pollen data. "

To

"Despite these advantages with data from the Hirst trap, data of fungal spores are rare in comparison to pollen data."

### Specific comment by reviewer 2:

4. P. 14331 line 23-24 – rephrase sentence.

#### Reply:

The sentence has been rephrased from

"Observations from Hirst traps have the last 5-10 years improved knowledge of aeroallergens concerning the temporal distribution and possible source locations to aeroallergens"

To

"Observations from Hirst traps have the last 5-10 years been used to improve knowledge of aeroallergens, especially concerning possible source locations to these aeroallergens"

### Specific comment by reviewer 2:

5. P. 14332 line 4 – please mention that aspergillus and penecilliun are also very common fungal species prevalent in the atmosphere.

## Reply:

We have modified the sentence from

"Fungal spore that are among the most often observed genera are *Cladosporium* and *Alternaria* (Larsen, 1981)"

Into

Fungal spores that are among the most often observed genera are *Aspergillus, Pennicillium, Cladosporium* and *Alternaria* (Lang-Yona et al., 2012; Larsen, 1981).

### Specific comment by reviewer 2:

6. P. 14332 line 7 – add that the specie of alternaria that can threat human health is alternaria alternate.

#### Reply:

As requested, we have modified the sentence from

"Alternaria spores can also threaten human health (Damato and Spieksma, 1995) and cause allergic symptoms in sensitized individuals when the atmospheric concentrations are high (Gravesen, 1979)"

Into

"Alternaria spores of the species Alternaria alternata can also threaten human health(Damato and Spieksma, 1995) and cause allergic symptoms in sensitized individuals when the atmospheric concentrations are high (Gravesen, 1979)"

### Specific comment by reviewer 2:

7. P. 14332 line 13 – the fact that alternaria peaks in the month before leaf fall does not mean that all fungi behave like this. In the study by Burshtein et al. the claim was that the vegetation is a confusing factor for some of the biomarkers and that general fungi peak is during autumn and spring only in the local scale. In the study by Escuredo et al. the claim was concerning the local behavior of the alternaria. This must be calrified in the text.

#### Reply:

We agree with the reviewer, that not all fungi peak before leaf fall. Our study however only concerns *Alternaria*, so a longer discussion on the findings by Burshtein et al (2011) and other fungal spore types are less appropriate. We have therefore removed the reference Buhrstein et al (2011) at this particular place so that the sentence now reads:

"..during the drying and decomposition of above-ground plant tissues (Escuredo et al., 2011; Iglesias et al., 2007)"

#### Specific comment by reviewer 2:

8. P. 14332 line 27 – unify the writing to p. meibomiae.

#### Reply:

The suggested changes has been implemented as requested

## Specific comment by reviewer 2:

9. P. 14333 line 11 – rephrase sentence : : "protocol that has been used in several similar European studies about"...

#### Reply:

As requested, we have modified the sentence from

"We have adapted a protocol that has been used in several similar studies in several European studies on allergenic pollen since 2007"

Into

"We have adapted a protocol that has been used in several similar European studies about allergenic pollen since 2007"

#### Specific comment by reviewer 2:

#### Methodology

1. P. 14333 line 26 – the authors refer to Mandroli et al. 1998, yet there is no such reference in the reference list nor can it be found on the web. The authors must explain how they determined the annual spore index and why it is dimensionless.

### Reply:

The missing reference is "Sampling: Principles and Techniques in Methods in aerobiology by Mandrioli, P., Comtois, P & Levizzani, V. (ed.): 262pp, Tecnoprint S.n.c., Italy". More detailed it is Appendex II, which elaborates on this subject in more detail. This missing references has therefore been implemented as (Mandrioli et al., 1998).

### Specific comment by reviewer 2:

2. Error bars must be added to all the figures.

### Reply:

This request must relate to limited knowledge of the observational methods with the pollen and spore trap of the Hirst design. The observational error from the Hirst trap is known to be related to both counting method (Pedersen and Moseholm, 1993; Sikoparija et al., 2011), environmental variables and the concentration of pollen and spores (Skjøth and Sommer, 2012). Time series of bi-hourly or daily values from this trap (as shown in in figure3, figure 4 and figure 5) are therefore in general never accompanied with error bars in scientific literature. A major reason is that the production of error bars requires additional observations and calculations that are not part of the observational protocol in the European Aeroallergen Network. Although we are strong supporters of error bars, we kindly decline this particular request in order for the figures (figure 3 to figure 5) to be consistent with existing scientific literature. However, Figure 2 can be associated with error bars and we have as requested modified it to include error bars that represent 1 standard deviation. Figure 1 now looks as follows

# Mean diurnual fungal spore concentration (peak days) 2001-2010

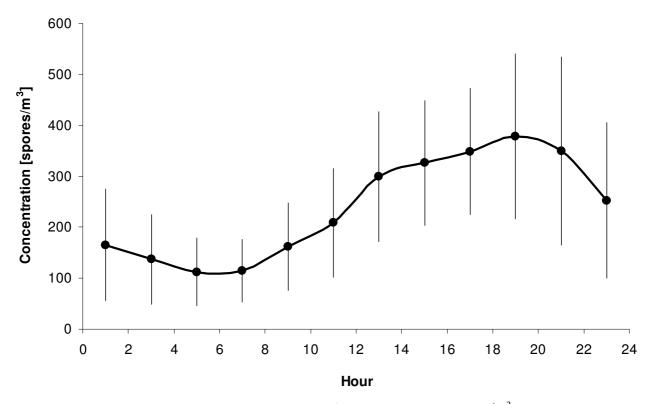


Figure 1. Mean diurnal *Alternaria* spore concentration for days above 100 spores/m³, n=232. The error bar about each mean value corresponds to 1 standard deviation.

## Specific comment by reviewer 2:

3. P.14334 the authors should explain shortly what is the 95% method since it is not clear in the reference they direct to.

## Reply:

We have added the following sentence: "Each of the 10 years are therefore investigated during that period, where the accumulated number of fungal spores are between 2.5% and 97.5% of the total annual catch."

## Specific comment by reviewer 2:

4. P.14334 in the days that are not peak concentrations, what were the values of the spores? Were they significantly lower? Information about the background levels of the spores should be added.

The reviewer got a very good point here. However dealing with background concentrations of fungal spores in a similar way as other air quality components goes against scientific tradition in the analysis of seasonal observations of pollen and fungal spores. This topic was in fact quite well covered by Mandroli et al. (1998) during the discussion of the dimensionless units of pollen and spore indexes (Mandrioli et al., 1998), in particular appendix 2. The main reason is that the length of season will vary from year to year due to weather variables. In many cases, the total load of pollen and spores will not reflect the season length to a large degree. But calculations of mean concentrations, including background loads etc. will be dependent on the season length. To come around this problem and at the same time respecting both scientific tradition and the request by the reviewer, we propose to expand Table 1 with four new columns: a) end of season, b) Total fungal spore catch during season, c) sum of low days, d) sum of high days. With this extension, it becomes more visible, that peak days are always outnumbered by low days and that the major portion of the fungal spore catch is during peak days. The new table is reproduced here:

Table 1. Maximum daily *Alternaria* spore concentrations (spores/m³), day of season start, day of season end and day of maximum spore concentration (days from 1<sup>st</sup> January) and number of days with concentrations above 100 pollen spores/m³ recorded in Copenhagen during 2001-2010. Sum of spores, sum on low days and sum on peak days all correspond to the total accumulated catch during the season (the days that cover 95% of the entire catch) and not the entire year.

Year	Seasonal	Day of	Day of	Day of peak	Peak value	Sum of	Sum, low	Sum, high	Days above
	Spore	season	season	concentration	Spores/m <sup>3</sup>	spores in	days in	days in	100 (high)
	Index	start	end			season	season	season	spores/m <sup>3</sup>
2001	9431	06Jul	20Sep	17Aug	1016	8966	1875	7091	23
2002	7046	05Jul	12Sep	29Jul	567	6686	1877	4809	21
2003	4488	18Jul	18Sep	19Jul	279	4257	1291	2966	17
2004	5651	02Jul	08Sep	06Aug	607	5321	1266	4055	18
2005	8141	21Jun	20Sep	10Aug	468	7565	1600	5965	20
2006	10781	01Jul	21Sep	11Aug	682	10251	2164	8087	27
2007	7813	20Jun	28Aug	17Jul	588	7386	1735	5651	22
2008	5276	26Jun	11Sep	31Aug	313	5006	2208	2798	17
2009	10511	01Jul	15Sep	09Aug	595	9989	1780	8209	31
2010	7519	08Jul	08Sep	03Aug	689	7178	1651	5527	27
Mean	7946	01Jul	12Sep	06Aug	580	7261	1745	5516	23
SD	2239	7 days	7 days	13 days	208	2044	315	1917	5

The updated table also requires additional text in the results section, discussion and conclusion. We therefore have changed in the results section from

"The contribution from individual years ranged from 17 days in 2003 and 2008 to 31 days in 2009"

#### Into

"The contribution from individual years ranged from 17 peak days in 2003 (with 70 days) and 2008 (with 60 days) to 31 days in 2009 (with 76 days). The contribution of the high days to the total seasonal load varied from about 55% in 2008 to more than 82% in 2009"

And added to the discussion section, first paragraph after ".. of the day or night (Mahura et al., 2007; Skjøth et al., 2007; Skjøth et al., 2008). ":

into

".. of the day or night (Mahura et al., 2007; Skjøth et al., 2007; Skjøth et al., 2008). The number of peak days are outnumbered by low days (Table 1), but every year the total load during the season has been dominated by the peak days, which contributed up to 82% of the entire *Alternaria* load during the season (Table 1)."

And changed in the conclusion section from

".. that the source to the overall load is mainly local, but with intermittent LDT from more remote agricultural areas. This hypothesis is supported by the analysed data of the 10 year bi-hourly record"

Into

that the source to the overall load is mainly local, but with intermittent LDT from more remote agricultural areas. These LDT episodes contributed to a large degree to the total annual load of *Alternaria* spores. In fact, the peak days dominate the overall *Alternaria* load, although peak days are always outnumbered by low days. The hypothesis is supported by the analysed data of the 10 year bi-hourly record

#### Specific comment by reviewer 2:

5. P. 14335 line 13 – in what conditions were the samples archived until analysis?

#### Reply:

The samples are kept in a protective box in the fields and after return to the lab the samples are prepared by using a mixture that make the fungal spore stand out during counting. Thereafter, the glass slides obtains a cover and the samples are stored in their protective box before counting with microscope. Similar methods are used throughout Europe and it allows for re-inspection of the slides several years back in time.

## Specific comment by reviewer 2:

6. P. 14335 Line 17 – the authors say that they cleaned the pipes with a stream of air; did they assure that this is sufficient? Did they check that there is no contamination left in the pipes? Wouldn't it be preferable to clean the pipes with a solvent that will clear all the spores from previous sampling?

## Reply:

Yes, this was checked using the control samples. They counted zero, although they were only cleaned with air.

## Specific comment by reviewer 2:

7. P.14336 line 13 - delete "for".

### Reply:

The word "for" has been deleted as requested.

## Specific comment by reviewer 2:

8. Regarding the use of the HYSPLIT model – please add the coordinates that you used in the model. The authors need to explain what is the advantage of calculating the back trajectories in altitude of 500m and why was it done in the matrix format.

#### Reply:

We have added the following sentence to section 2.1. "The trap was located on the roof of the Danish Meteorological Institute (55°43'N, 12°34'E) in the centre of Copenhagen at a height of 15m above sea level.".

The 500m altitude is the recommended altitude and default value in the HYSPLIT model. Near all aerobiological studies with trajectory models have used a 500m or near-500m altitude, depending on actual model formulation. To the knowledge of the authors, only the study by Mahura et al (2007) has used the surface as altitude for aerobiological studies. This procedure, however, is against the general recommendation in the use of trajectory models due to a technical limitation. Often there are problems with a correct representation of horizontal and vertical surface winds, which will results in an air mass pattern that often follows the surface and stays there several days back in time, which is highly unrealistic.

The matrix format has the advantage that it provides an estimate of uncertainty of the trajectories. This procedure was used for the first time for aeroallergens over Copenhagen by Skjøth et al (2007). The methodology is quite common in air quality studies and was in 2008 implemented as a standard option in the HYSPLIT model. The reference (Skjøth et al., 2007) has therefore been added to that section.

#### Specific comment by reviewer 2:

Results

1. P. 14337 line 8 - delete "was".

## Reply:

The correction has been implemented as requested

## Specific comment by reviewer 2:

2. Not all alternaria spores affect human health. The clinical threshold the authors stated is 100 spore m-3, why does this value represents the minimal level of the clinical threshold? There is no differentiation

between the alternaria species in this work and it is possible that a very small fraction of the 100 spore m-3 can have health implications.

Please discuss this issue.

### Reply:

Although the subject is very interesting from a clinical and health perspective, we believe, that this discussion is outside the scope of this journal as well as this study. This study is a source-receptor study in biological particles and not an investigation of clinical relevance of the different fractions of Alternaria spores in the air. We therefore suggest not to change this part.

### Specific comment by reviewer 2:

3. P. 14337 lines 17-20 - Did the authors investigate correlation between the peak days and the harvesting times? There might be a simple connection. Moreover, how did the authors calculate the trajectories prior to 2006? (To my best information the HYSPLIT model in the GDAS mode has information only from 2006 till present day and the measurement period is 2001-2010).

#### Reply:

The three episodes (Figure 3, Figure 4 and Figure 5) were all analysed using the GDAS archive. Additionally all episodes that were listed in Table 2 were also analysed (but not shown). This extended period used the reanalysis data set instead, as this is the only data set that covers the entire observation period. We have therefore modified page 14341, line 5-9 from

"Finally, then the small fraction of peak days with a different diurnal pattern than the overall pattern have been analysed with respect to air mass transport. In all cases, the air masses came from more remote areas that are also dominated by land cover types containing potential sources to *Alternaria* spores. Such episodes were identified almost every year during the study period (Table 2)"

Into

"Finally, the small fraction of peak days, which show a diurnal pattern that differs from the overall pattern, have been analysed with respect to air mass transport (using HYSPLIT and the reanalysis meteorological data set). In all cases, the air masses came from more remote areas that are also dominated by land cover types containing potential sources to *Alternaria* spores. Such episodes were identified almost every year during the study period (Table 2) "

#### Specific comment by reviewer 2:

4. Please change "Scania" to "Sweden" throughout the text.

We understand the reviewers request for a harmonized way of writing the geographical names. However we must decline this suggestion. When discussing Sweden, it is very important to distinguish between Scania and rest of Sweden. Soil type and land use in Scania is very different compared to the rest of Sweden. Most of Scania is covered by very intensely used agricultural land while the rest of Sweden is mainly covered by forest. So changing from Scania to Sweden will change the meaning of the sentence and the statements will no longer be correct.

## Specific comment by reviewer 2:

5. P. 14337 line 25 - delete "an".

### Reply:

The correction has been implemented as requested

### Specific comment by reviewer 2:

6. P. 14338 line 1 - delete "of".

#### Reply:

The correction has been implemented as requested

### Specific comment by reviewer 2:

7. P. 14338 line 5 - change "at" to "as".

## Reply:

The correction has been implemented as requested

### Specific comment by reviewer 2:

8. P. 14338 line 25 - delete "to".

## Reply:

The correction has been implemented as requested

## Specific comment by reviewer 2:

9. P. 14338 line 26 – rephrase to: "The weather in the study region and period had a high pressure: ::"

### Reply:

The sentence has been modified from

"The weather in the study region had during the study period a high pressure ridge extending from Iceland (1029 hPa) over Scandinavia (~1020 hPa) to northern Germany and central Poland (1022-1023 hPa), which in the beginning of the period pushed air masses from the North towards Copenhagen."

Into

"During the study period, the weather in the study region had a high pressure ridge extending from Iceland (1029 hPa) over Scandinavia (~1020 hPa) to northern Germany and central Poland (1022-1023 hPa). This caused air masses to be pushed from the North towards Copenhagen."

### Specific comment by reviewer 2:

10. P. 14339 line 8 – why weren't the harvesting possibilities checked? If there is a way to retrieve this information it should be done to make the claim (that the harvest causes high spores concentration) stronger.

#### Reply:

We agree with the reviewer here that it would be a good idea to verify harvesting possibilities and check if the farmers have been harvesting. Unfortunately this is not possible, as harvesting dates are in general not recorded. In fact it is a general problem for environmental science to obtain reliable information about agricultural management (activity data) on either national scale or European scale (Skjøth et al., 2004; Skjøth et al., 2008a). Here Denmark is in a unique position with the largest and most detailed agricultural registers, which are accessible to researchers (Gyldenkærne et al., 2005) for agricultural modelling. This has enabled large scale modelling of agricultural management (Skjøth et al., 2011). Due to this, the Danish approach has been selected for implementation in the EMEP and EMEP4UK air quality models within the large integrated FP7 project ECLAIRE (http://www.eclaire-fp7.eu/) during 2011-15. Despite a number of improvements and access to some of the most detailed data bases, it is still not possible to obtain the requested information for this kind of study. So handling of harvesting dates is indeed a subject for future research within environmental science including reactive nitrogen and if possible also bioaerosols such as Alternaria.

### Specific comment by reviewer 2:

11. Please add information about the local wind speed and direction for sections 3.3.1-3.3.3.

Wind speed and wind direction generally changes with height, especially within the first 300-500 metres; this is called the Ekman layer. The exact distribution of wind speed with height depends on many parameters such as the overall weather situation and local surface roughness. For a complete description see chapter 16.5 by Seinfeld and Pandis (Seinfeld and Pandis, 1998). As a result, these surface variables are not reliable indicators of the path taken by fungal spores or possible source areas. This is the main reason why atmospheric models such as HYSPLIT, FLEXTRA or FLEXPART (e.g. Stohl, 1998; Stohl and Seibert, 1998) are used in this kind of studies. We therefore believe that increasing this study with wind speed and direction near the surface is not pertinent to this study. We therefore suggest not to implement this correction.

#### Specific comment by reviewer 2:

12. P. 14340 line 4 – correct to "in the evening of the 16th".

#### Reply:

The correction has been implemented as requested

### Specific comment by reviewer 2:

13. P. 14340 lines 8-11 – rephrase sentence.

#### Reply:

The sentence has been rephrased from

"This caused in the beginning of the period that air masses from the East and the Baltic states were pushed towards Denmark, arriving at Copenhagen from the North West passing northern over parts of Scania."

To "

"This caused air masses from the East and the Baltic states to be pushed towards Denmark in the beginning of the period. These air masses arrived at Copenhagen from the North West passing over northern parts of Scania."

## Specific comment by reviewer 2:

14. P. 14340 line 17 - If heavy rain was recorded for this period over Denmark, how do the authors explain the extreme high concentrations? Wouldn't they expect sedimentation of the spores by the rain to the ground before they reach the sampling site? Are they released from the ground? Explain and provide supporting information.

Heavy summer rain in this part of Europe rarely lasts more than a few hours. In fact this is to some degree the definition of summer showers that affects these latitudes. The areas that are affected by showers will in general not be harvested. But most of the day over these areas will still be dry, which will limit rain out. So from a process based view, a shower will stop the emission process (the harvesting) for some time but once the shower stops, the advection of spores from more dry areas will immediately start again. This is one more reason why, it is very relevant to study these mechanisms with atmospheric models. Here a basic requirement is, that the atmospheric models has access to estimates of emission areas, which we have produced in this manuscript.

### Specific comment by reviewer 2:

#### Discussion

1. P.14340 line 21 - It is a bit difficult to conclude that the 232 peak days have a strong diurnal pattern without standard deviation data. What about the day prior to the peak and the day that follows? The authors should investigate this relation as well and provide this data. In the "special cases" the authors claim that they did not see a pattern of the peaks change during the day. What happens to the pattern in regular days when the spore concentration is below 100 spore m-3?

### Reply:

We have modified Figure 1 so that includes an error estimate of 1 standard deviation. With respect to low days and typical pattern, such days are often excluded in the analysis of samples that contain hourly values of pollen and spores (Emberlin and Norris-Hill, 1991; Norris-Hill and Emberlin, 1991; Skjøth et al., 2009). The main reason is that the observed values on low days are heavily influenced by rainout in the atmosphere and exactly when the rains arrive at the observational site. Emberlin and Norris-Hill (1991) therefore recommend that analysis in relation to daily patterns focus on dry days and days with high values. We have also investigated extended periods around the peak days. In fact there are a number of peak days that follow each other. On other days, a peak day can be followed by very low numbers. Our investigation points out that the low numbers are associated with a change in meteorological conditions such as winds arriving from the North or the arrival of large scale precipitation events. This suggests, that some patterns in the *Alternaria* load can be simulated with atmospheric models once the emission pattern (location and temporal variation) has been correctly described

We have there added additional text to the conclusion by changing following from

"... forecasting. Furthermore, development of emission models and inventories makes it possible to use source-based models such as DEHM (Brandt et al., 2012), SILAM (Sofiev et al., 2006) and COSMO-ART (Zink et al., 2012) for improved understanding of aeroallergens and ultimately better information to the public."

Into

"..forecasting. The episodes that we analysed in detail showed that it is possible to both have peak days that follow each other and that the change from low to high load of *Alternaria* is related to both a change in weather and potential source area. Such patterns can be simulated with atmospheric transport models. The development of emission models and inventories therefore makes it possible to use source-based models such as (Brandt et al., 2012), SILAM (Sofiev et al., 2006) and COSMO-ART (Zink et al., 2012) for improved understanding of aeroallergens and ultimately better information to the public."

### Specific comment by reviewer 2:

2. P.14340 line 26 - One out of 23 peak days in a year peak, in average, is not enough to conclude about the pattern of the diurnal cycle, please recalculate for all peak days.

#### Reply:

The reviewer must have misunderstood the message, which we believe is due to the wording of our sentence. The mean diurnal cycle (Figure 2) is based on all peak days, which is also written in the legend. The daily pattern of all these 232 were investigated individually and compared with the mean pattern. Only 16 episodes had a daily pattern that were markedly different compared to the mean. In fact, there were only 1 episode in 2001 and no episode in 2002, which the reviewer clearly has seen.

To make this clear, we have rephrased line 26 in page 14340 from

"In fact only a small fraction of the peak days has a diurnal pattern that deviates from the overall pattern (Table 2)"

Into

Here we have investigated all 232 high days individually. Only 16 (Table 2) of the 232 high days has a diurnal pattern that deviates from the overall pattern (Figure 2).

#### Specific comment by reviewer 2:

3. P. 14341 line 5 – delete "then". The authors should display the information about the peak days with a different diurnal pattern that was analyzed with respect to air mass transport e.g. back trajectory map, meteorology etc.

### Reply:

The correction with respect to "then" has been implemented as requested.

As written earlier in this reply, all 16 days in Table 2 have been analysed with the HYSPLIT model using the renanalysis meteorological data set. This information has also been added to the manuscript. Additionally, the 3 chosen episodes have also been analysed using the GDAS meteorological data set. GDAS has higher spatial and temporal resolution than the reanalysis data. There was however not much difference in the two different calculations using different meteorological data sets (GDAS and reanalysis) and this will not

change the overall conclusions at all. We believe that it is out of scope of the manuscript to start to compare results from HYSPLYT calculations that originate from these two data sets.

## Specific comment by reviewer 2:

4. P. 14341 line 8 - change "to" to "of".

### Reply:

The correction has been implemented as requested.

#### Specific comment by reviewer 2:

5. P. 14341 line 12 - delete "in".

#### Reply:

The correction with respect to "in" has been implemented as requested and the sentence has been modified slightly so that it now reads

"Additionally, it was shown that even if a region such as eastern Denmark and southern Sweden had obtained very large amounts of rain making harvest very difficult, more remote regions, e.g. Poland, could have contributed with large amounts of *Alternaria* spores (Fig. 5)."

### Specific comment by reviewer 2:

6. P. 14341 line 13 - delete "with".

#### Reply:

After the modification which is written above, we believe that "with" is still needed in the sentence

#### Specific comment by reviewer 2:

7. P. 14341 line 13-15 – the author's claim that the air mass for these days could have arrived from Poland, because it rained heavily in Denmark at the same time. The trajectories calculated for these days are not consistent enough to conclude that there was a LDT event.

### Reply:

We agree that the trajectories in Fig5b are not that consistent with respect to potential source areas. But we disagree with the reviewer that the trajectories are not that consistent with respect to LDT. The reason is that during the last 100km towards Copenhagen the air masses do in fact have a similar pathway – over

water. These are areas without sources. So the source to the high amount of Alternaria spores must originate from another country before the air masses passed arrived to the sea. This supports LDT but does not suggest exactly the source area.

### Specific comment by reviewer 2:

8. P. 14341 lines 18-19 – rephrase sentence.

#### Reply:

As requested, we have rephrased the sentences from line 16-19 from

A number of source receptor studies on aeroallergens have recently linked measured concentrations from the Hirst traps with both local sources and intermittent long distance transport from regions with high source densities. Common for all of these studies are is that they are concerned with pollen such as *Betula* 

#### Into

Recently a number of source receptor studies on aeroallergens have been carried out by combining measured concentrations from the Hirst traps with trajectory calculations. These studies have identified both local sources and intermittent long distance transport from regions with high source densities. Common for all of these source-receptor studies is that they focus on pollen such as *Betula* 

#### Specific comment by reviewer 2:

9. P. 14341 line 28-29 – the author's claim that the fields were treated with fungicides and that no fungi infection was seen in the fields, yet samples collected from the harvesting machine showed high emissions of fungal spores. Did the authors check if the harvesting machine itself was not the source of the spores? In page 14335 line 16 the authors mention collection of negative controls from the field, yet the results are not shown. These are the background levels before harvest and it is not clear from the text what was done exactly and if the controls collected randomly in the field showed fungal spores concentrations.

#### Reply:

No, this was not checked initially, but was not deemed necessary as the harvesting machine was brand new and was being used for the first time when the first measurement was taken. In addition, when the farmer let the machine run idle (the machine not advancing, i.e. no grain being harvested, but the motors running at normal speed), we were unable to find spores in the exhaust of the machine.

### Specific comment by reviewer 2:

10. P. 14342 lines 20-24 - Although fungi flourish after the rain, it does not happen instantly and since the ground is wet, it is predictable that uplifting of particles will be minimal.

### Reply:

We agree with the reviewer. However, uplifting by weather is not an issue here as we study the release of fungal spores that are within the crop and is released during harvesting process by mechanical force.

### Specific comment by reviewer 2:

11. P. 14344 line 4 – correct to "Friesen et al., (2001)".

## Reply:

The correction has been implemented as requested.

### Specific comment by reviewer 2:

12. P. 14344 lines 9-10 – it is not acceptable to reference Wikipedia, please find the original reference for this information.

## Reply:

Reviewer it considers these references not needed. We have therefore removed them entirely.

### Specific comment by reviewer 2:

13. P. 14344 lines 21-23 - r2 = 0.67 is not a very strong correlation; please show this data and try to explain why is the correlation is not stronger if the 2 factors are dependent.

### Reply:

As requested, we have removed the word "highly". The data reviewer 2 requests are in fact already present in two columns of the original Table 1. To make this more clear, we have changed the words "not shown" into "column 2 and 10 in Table 1"

### Specific comment by reviewer 2:

#### Conclusions

The authors should rewrite the conclusions after all corrections are made.

#### Reply:

After we have modified the manuscript and clarified how to interpret trajectory calculations and identify LDT, we do not feel that this part needs to be rewritten to a large degree. We have added a few sentences

(see above) in relation to the extended Table 1. However, there is one particular point that reviewer 2 has identified. It cannot be said for sure, if the source to Alternaria spores that were found in the exhaust from the harvesting machine came from the fields or from the harvesting machine itself

#### Specific comment by reviewer 2:

Overall changes in the English and presentation levels are needed.

#### Reply:

We have modified the manuscript with respect to both presentation of levels and the written English. We are would here like to thank then very constructive review by two reviewers

#### REFERENCES

Brandt, J., Silver, J. D., Frohn, L. M., Geels, C., Gross, A., Hansen, A. B., Hansen, K. M., Hedegaard, G. B., Skjøth, C. A., Villadsen, H., Zare, A., and Christensen, J. H., 2012, An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport of air pollution: Atmos. Environ., **53**, 156-176.

Burshtein, N., Lang-Yona, N., and Rudich, Y., 2011, Ergosterol, arabitol and mannitol as tracers for biogenic aerosols in the eastern Mediterranean: Atmos. Chem. Phys., **11**, 829-839.

Buters, J. T. M., Thibaudon, M., Smith, M., Kennedy, R., Rantio-Lehtimäki, A., Albertini, R., Reese, G., Weber, B., Galan, C., Brandao, R., Antunes, C. M., Jäger, S., Berger, U., Celenk, S., Grewling, L., Jackowiak, B., Sauliene, I., Weichenmeier, I., Pusch, G., Sarioglu, H., Ueffing, M., Behrendt, H., Prank, M., Sofiev, M., and Cecchi, L., 2012, Release of Bet v 1 from birch pollen from 5 European countries. Results from the HIALINE study: Atmos. Environ., **55**, 496-505.

Cecchi, L., Malaspina, T., Albertini, R., Zanca, M., Ridolo, E., Usberti, I., Morabito, M., Dall' Aglio, P., and Orlandini, S., 2007, The contribution of long-distance transport to the presence of Ambrosia pollen in central northern Italy: Aerobiologia, **23**, 145-151.

Damato, G. and Spieksma, F. T. M., 1995, Aerobiologic and Clinical Aspects of Mold Allergy in Europe: Allergy, **50**, 870-877.

Emberlin, J. and Norris-Hill, J., 1991, Spatial Variation of Pollen Deposition in North London: Grana, **30**, 190-195.

Escuredo, O., Seijo, M., Fernández-González, M., and Iglesias, I., 2011, Effects of meteorological factors on the levels of *Alternaria* spores on a potato crop: Int. J. Biometeorol., **55**, 243-252.

Gannibal Ph.B. (A.A.Jaczewski Laboratory of Mycology and Phytopathology, All-Russian Institute of Plant Protection, St. Petersburg, Russia). 2008. Alternaria alternata (Fr.) Keissl., Cladosporium herbarum (Pers.) Link., Epicoccum purpurascens Ehrenb., Botrytis cinerea Pers. - Black Ear of wheat. In Afonin, A.N.; S.L. Greene; N.I. Dzyubenko, A.N. Frolov (eds.). 2008. Interactive Agricultural Ecological Atlas of Russia and Neighboring Countries. Economic Plants and their Diseases, Pests and Weeds[Online]. Available at: <a href="http://www.agroatlas.ru/en/content/diseases/Hordei/Hordei Alternaria alternata/">http://www.agroatlas.ru/en/content/diseases/Hordei/Hordei Alternaria alternata/</a>

Gravesen, S., 1979, Fungi as a Cause of Allergic Disease: Allergy, 34, 135-154.

Gultyaeva E.I. (A.A.Jaczewski Laboratory of Mycology and Phytopathology, All-Russian Institute of Plant Protection, St. Petersburg, Russia). 2008. Alternaria alternata (Fr.) Keissl., Cladosporium herbarum (Pers.) Link., Epicoccum purpurascens Ehrenb., Botrytis cinerea Pers. - Black Ear of wheat. In Afonin, A.N.; S.L. Greene; N.I. Dzyubenko, A.N. Frolov (eds.). 2008. Interactive Agricultural Ecological Atlas of Russia and Neighboring Countries. Economic Plants and their Diseases, Pests and Weeds[Online]. Available at: http://www.agroatlas.ru/en/content/diseases/Tritici/Tritici Alternaria alternata/

Iglesias, I., Rodríguez-Rajo, F., and Méndez, J., 2007, Evaluation of the different *Alternaria* prediction models on a potato crop in A Limia (NW of Spain): Aerobiologia, **23**, 27-34.

Kirk PM, Cannon PF, Minter DW, Stalpers JA. (2008). *Dictionary of the Fungi. 10th ed.* Wallingford: CABI. p. 22.

Lang-Yona, N., Dannemiller, K., Yamamoto, N., Burshtein, N., Peccia, J., Yarden, O., and Rudich, Y., 2012, Annual distribution of allergenic fungal spores in atmospheric particulate matter in the Eastern Mediterranean; a comparative study between ergosterol and quantitative PCR analysis: Atmos. Chem. Phys., **12**, 2681-2690.

Larsen, L. S., 1981, A 3-Year-Survey of Micro-Fungi in the Air of Copenhagen 1977-79: Allergy, 36, 15-22.

Mahura, A., Korsholm, U., Baklanov, A., and Rasmussen, A., 2007, Elevated birch pollen episodes in Denmark: contributions from remote sources: Aerobiologia, **23**, 171-179.

Mandrioli, P., Comtois, P., and Levizzani, V., 1998, Methods in Aerobiology: Pitagora Editrice, Bologna, ISBN 88-371-1043-X, 262pp.

Norris-Hill, J. and Emberlin, J., 1991, Diurnal-Variation of Pollen Concentration in the Air of North-Central London: Grana, **30**, 229-234.

Pedersen, B. V. and Moseholm, L., 1993, Precision of the daily pollen count. Identifying sources of variation using variance component models: Aerobiologia, **9**, 15-26.

Perelló, A.E. and M.N. Sisterna. 2005. Leaf blight of wheat caused by Alternaria triticina in Argentina. New Disease Reports 11, 24-28.

Seinfeld, J. H. and Pandis, S. N., 1998, Atmospheric Chemistry and Physics: John Wiley & Sons Inc, New York, USA.

Sikoparija, B., Pejak-Sikoparija, T., Radisic, P., Smith, M., and Soldevilla, C. G., 2011, The effect of changes to the method of estimating the pollen count from aerobiological samples: Journal of Environmental Monitoring, **13**, 384-390.

Singh, R.V., A.K. Singh, and S.P. Singh. Distribution of pathogens causing foliar blight of wheat in India and neighboring countries. In: E. Duveiller, Dubin, H.J., Reeves, J., McNab, A. (eds.). 1998. Helminthosporium Blights of Wheat: Spot Blotch and Tan Spot. Mexico, D.F.: CIMMYT, pp. 59-62.

Skjøth, C. A., Smith, M., Brandt, J., and Emberlin, J., 2009, Are the birch trees in Southern England a source of Betula pollen for North London?: Int. J. Biometeorol., **53**, 75-86.

Skjøth, C. A. and Sommer, J., 2012, Two extended pollen and fungal spore calendars for Denmark, their representativeness and cross-reactivity between pollen and food products: Submitted to Aerobiologia Feb 2012, under review.

Skjøth, C. A., Sommer, J., Brandt, J., Hvidberg, M., Geels, C., Hansen, K., Hertel, O., Frohn, L., and Christensen, J., 2008, Copenhagen — a significant source of birch ( *Betula* ) pollen?: Int. J. Biometeorol., **52**, 453-462.

Skjøth, C. A., Sommer, J., Stach, A., Smith, M., and Brandt, J., 2007, The long range transport of birch (*Betula*) pollen from Poland and Germany causes significant pre-season concentrations in Denmark: Clinical and Experimental Allergy, **37**, 1204-1212.

Sofiev, M., Siljamo, P., Valkama, I., Ilvonen, M., and Kukkonen, J., 2006, A dispersion modelling system SILAM and its evaluation against ETEX data: Atmos. Environ., **40**, 674-685.

Stohl, A., 1998, Computation, accuracy and applications of trajectories - A review and bibliography: Atmos. Environ., **32**, 947-966.

Stohl, A. and Seibert, P., 1998, Accuracy of trajectories as determined from the conservation of meteorological tracers: Quarterly Journal of the Royal Meteorological Society, **124**, 1465-1484.

Zink, K., Vogel, H., Vogel, B., Magyar, D., and Kottmeier, C., 2012, Modeling the dispersion of *Ambrosia artemisiifolia* L. pollen with the model system COSMO-ART: International Journal of Biometeorology, in press, doi 10. 1007/s00484-011-0468-8, 1-12.