Reporting on natural events in the EU Member States under Directive 2008/50/EC: years 2008-2009

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Front page picture:
Top left: dust plume off the Sahara desert over the northeast Atlantic Ocean. The Azores are visible at the northwest edge of the dust plume in this SeaWiFS image. The Cape Verde Islands can be seen through the dust near the bottom of the image. Sensor: OrbView-2/SeaWiFS. 1 November 1998. Provided by the SeaWiFS Project, NASA/Goddard Space Flight Center, and ORBIMAGE.
Top right: sea salt aerosol formation. Image kidly provided by www.sxc.hu (image ID: 1340168).
Bottom left: flames raced through the forests northeast of Athens, Greece, on August 22, 2009, when the Moderate Resolution Imaging Spectroradiometer (MODIS) on NASA's Aqua satellite captured this image. Areas where the sensor detected fire on the ground are outlined in red. Provided by the SeaWiFS Project, NASA/Goddard Space Flight Center, and ORBIMAGE.
Bottom right: on July 22, 2001, SeaWIFS imaged a yellowish-brown plume emanating from the Mt. Etna volcano and stretching over 600 kilometers southeastward across the Mediterranean towards Libya. Provided by the SeaWiFS Project, NASA/Goddard Space Flight Center, and ORBIMAGE.

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Summary

Based on the questionnaires reported by Member States to the Commission for the years 2008 and 2009, the reporting on natural events by the Member States under Directive 2008/50/EC was analysed. The contribution of four main natural causes to daily and annual PM_{10} levels and to the exceedances of the corresponding limit values was evaluated: African dust, sea salt aerosols, volcanic dust, and wild-land fires.

Reporting on natural events by the Member States in 2008 and 2009
10 Member States (AT, CY, DE, ES, FR, GB, GR, IT, MT, PT) reported exceedances of the PM_{10} annual and daily limit values due to natural events in 2008, and 8 in 2009 (CY, ES, FR, GB, GR, IT, LV, PT). This information is based on forms 23a and 23b of the questionnaire (see Annex II for the list of forms). In addition, 2 Member States (IS, PL) reported exceedances of the daily limit value due to natural events in form 11h, but did not report those incidents in form 23.

The number of Member States reporting exceedances of the annual limit value due to natural sources was lower than for the daily limit value, with 7 Member States in 2008 (CY, ES, FR, GB, GR, IT, PT) and 4 in 2009 (CY, ES, FR, GR).

The dominant natural event responsible for exceedances of the PM_{10} daily limit value was transport of natural particles from dry regions outside the Member State (standard code G2 in Commission Decision 204/461/EC, see Table S1). The event was named by 70% of the Member States reporting natural events in 2008 (CY, DE, ES, FR, GR, IT, PT) and 50% of the Member States in 2009 (CY, ES, GR and PT). Exceedances due to the more general term “natural source(s) or natural event(s)” (standard code S8 in Commission Decision 2004/461/EC, Table S1) were reported by 30% and 50% of the Member States in 2008 (AT, GB, MT) and 2009 (ES, GB, IT, LV), respectively. This code referred to African dust transport in all cases, and in addition to sea salt aerosols in GB. Finally, other causes were reported by single Member States: wild-land fire inside and outside the Member State (D1 and D2 in Commission Decision 2004/461/EC) and an unspecified cause (H1) by GR in 2008 and 2009, and “embruns marins” (sea salt) by FR in both years.

Mean annual contributions of natural sources to PM_{10} levels ranged from 1-3 $\mu g/m^3$ in IT, FR, PT and GR, to 4-5 $\mu g/m^3$ in ES and GB, to 13 $\mu g/m^3$ in CY in 2008. In 2009, mean contributions were lower in ES (1 $\mu g/m^3$ versus 4 $\mu g/m^3$ in 2008), higher in GR (8 $\mu g/m^3$ versus 3 $\mu g/m^3$ in 2008), and similar in CY (13 $\mu g/m^3$ in both years).

Table S1. Different Tables included in Commission Decision 2004/461/EC referring to natural events: (Top) Table 5, natural events causing limit value exceedances; (Bottom) Table 2, reasons for individual exceedances.

Natural event code	*Description*
A1	*Volcanic eruption inside the Member State*
A2	*Volcanic eruption outside the Member State*
B1	*Seismic activity inside the Member State*
B2	*Seismic activity outside the Member State*
C1	*Geothermal activity inside the Member State*
C2	*Geothermal activity outside the Member State*
D1	*Wild-land fire inside the Member State*
D2	*Wild-land fire outside the Member State*
E1	*High wind event inside the Member State*
E2	*High wind event outside the Member State*
F1	*Atmospheric resuspension inside the Member State*
F2	*Atmospheric resuspension outside the Member State*
G1	*Transport of natural particles from dry regions inside the Member State*
G2	*Transport of natural particles from dry regions outside the Member State*

Reason code	*Description*
S1	*Heavily trafficked urban centre*
S2	*Proximity to a major road*
S3	*Local industry including power production*
S4	*Quarrying or mining activities*
S5	*Domestic heating*
S6	*Accidental emission from industrial source*
S7	*Accidental emission from non-industrial source*
S8	*Natural source(s) or natural event(s)*
S9	*Winter sanding of roads*
S10	*Transport of air pollution originating from sources outside the Member State*
S11	*Local petrol station*
S12	*Parking facility*
S13	*Benzene storage*

The correction by natural sources reduced the number of stations surpassing the 35 exceedances/year threshold in all Member States where this correction was applied,

with the only exception of LV in 2009. In the case of the mean annual values, the correction for natural sources also resulted for all Member States in a reduction of the number of stations exceeding the PM_{10} annual limit value of 40 μg/m^3. Different methodologies were applied by Member States to justify contributions of natural events to PM levels: ES, GB, MT and PT applied the methodology recommended by EC Staff Working Paper 6771/11 (for African dust events), whereas AT, CY, DE and GR applied qualitative methodologies with different degrees of comparability with respect to the methods recommended by Staff Working Paper 6771/11. IT and LV submitted no information on which methodologies were applied.

The data in form 11 indicate that the number of exceedances attributed to natural events is within the average in the cases of CY, ES, GB, GR and PT. The number of natural exceedances of the daily limit value reported by IT was high with respect to the total number of natural events recorded for 2009. The seasonality of the natural exceedances of the PM_{10} daily limit value in all Member States is coincident with the occurrence of African dust outbreaks.

The following recommendations may be provided regarding reporting of natural exceedances of the PM_{10} daily limit value in form 11 for the year 2009:

- Certain Member States reported a reasonable number of natural exceedances of the PM_{10} daily limit value (S8), following the characteristic seasonality. However, it would be advisable to improve the identification of the specific causes, taking into account that these Member States have the necessary tools for this (reporting >1 cause per exceedance should be avoided).
- Certain Member States reported higher number of exceedances than expected. Reporting more than 4 simultaneous causes for each exceedance day should be avoided, as it doesn't seem plausible. These Member States are advised to apply substantial improvements in their reporting strategies for form 11, concerning the correct identification of natural episodes and the discrimination of the causes.
- Member States not reporting the methodologies used to identify natural events are encouraged to do so.

Formal aspects of the reporting procedures

Sea salt aerosols:

It is suggested that sea salt aerosols should be included in future revisions of the EC Staff Working Paper 6771/11 and that a standard code should be created for this entry. This would eliminate the discrepancy existing currently between the texts 2004/461/EC and 6771/11.

Analysis of the types of natural events reported:

- Member States did not always use the codes provided by Commission Decision 2004/461/EC.
- Member States reported natural events using both the general code S8 (Commission Decision 2004/461/EC), specific codes G2, D1, D2 (from Commission Decision 2004/461/EC, Table S1) and unspecified codes (H1, not included in any official document).
- Certain Member States reported both natural and non-natural causes for the same stations and periods.
- Certain Member States applied the same codes for all stations.

Assessment of methodologies applied in 2008 and 2009:

- Methodologies were described in the national languages by some Member States and in English by others.
- Not all Member States provided a description of the methodology used.

- All justification documents were publicly available, but while some were easily found, others were difficult to trace.
- Certain Member States used methodologies for justification of natural events which are not included in Staff Working Paper 6771/11.
- It could be recommended to submit the title of justification document and a brief summary of the methodology in English, in addition to the national language.
- For some Member States reporting exceedances due to the general code S8 ("natural events"), the assessment of the methodology reported for justification allowed for the identification of the type of natural source. However, this was not the case for other Member States, for which the natural causes reported were not specified at all.
- The methodologies for quantification of sea salt contributions presented in Staff Working Paper 6771/11 are somewhat contradictory: whereas the possibility of calculating the sea salt contribution using only sodium or chloride as a tracer is included, the same document discourages the use of chloride as the only tracer given that it is potentially subject to both positive and negative artefacts. It would be advisable to define a clear recommendation regarding this issue.
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Comparison between data reported in form 23 and form 11:

- Codes from two different Tables from Commission Decision 2004/461/EC (Tables 2 and 5, here in Table S1) were used simultaneously for reporting in 2008 and 2009 in forms 23a and 23b. For future reporting, the use of Table 2 is recommended for reporting in form 11h, and Table 5 for reporting in forms 23a and b, according to the Commission Decision 2004/461/EC.
- Certain Member States submitted data in form 11 but did not submit the corresponding information in form 23. This is correct when the 35 exceedances/year threshold for the PM_{10} daily limit value is not surpassed. However, this is not correct when the threshold is exceeded.
- Inconsistencies were detected between both forms regarding the number of stations reporting exceedances of the PM_{10} daily limit value due to natural events.
- Discrepancies were found between causes for exceedances reported by a same station in forms 11 and 23.

1. Introduction

1.1. Legislative background

The Directive 2008/50/EC on ambient air quality and cleaner air for Europe provides Member States with the possibility to subtract the contribution of natural sources under certain conditions before comparing the ambient air pollutant concentrations to the limit values. Guidance on which sources can be regarded as natural in this context and on methods to quantify and subtract the contribution of these sources are given in the Commission Staff Working Paper 6771/11 of 18 February 2011. Before Staff Working Paper 6771/11, the Commission Decision 2004/461/EC established the questionnaire to be used for annual reporting on ambient air quality assessment under Council Directives 96/62/EC and 1999/30/EC and under Directives 2000/69/EC and 2002/3/EC of the European Parliament and of the Council.

In Staff Working Paper 6771/11, six key principles are set out, which the Commission intends to apply when evaluating Member States attributions of exceedances to natural events:

(1) the contributions must not be caused by direct or indirect human activities;
(2) the quantification of the natural contribution must be sufficiently precise;
(3) the quantification of the natural contribution must be consistent with the averaging period of the limit value;
(4) the quantification of the natural sources must be spatially attributed;
(5) the contributions must be demonstrated based on a systematic assessment process;
(6) the quantification of the natural sources must be demonstrated for each pollutant separately.

These key principles clearly lay down the ground for justification of exceedances of the limit values due to natural events. Special attention should be paid to point number (1), which implies that no direct or indirect human activity should be involved in the natural cause. This limitation applies especially to certain types of events (e.g., wild-land fires, re-suspension of dry particles inside the Member State), which may occur with and without human intervention. However, only the events caused strictly by natural causes may be justified. Thus, as an example, in order to justify wildfires as natural events, they must be started by natural causes (e.g., lightning). Human-related causes such as littering of forests which may derive in forest fires should not be justified as natural events. A natural contribution could not be prevented or significantly reduced by human actions.

A non-exhaustive list of sources for which contributions can be eligible for subtraction is explicitly covered in Staff Working Paper 6771/11:

- Transport of natural particles from dry regions
- Sea spray
- Volcanic eruptions & seismic activities
- Wild-land fires.

A second non-exhaustive list describes sources that the Commission does not consider to be eligible for subtraction:

- Primary biological aerosol particles
- Secondary organic biogenic aerosols
- Mineral dust re-suspension

In the Commission Decision 2004/461/EC (Table 5, here Table 1.1), a list of natural events and their standard codes was provided for justification. The events listed in the Commission Decision 2004/461/EC does not coincide exactly with the list of sources (or events) for which contributions can be eligible for subtraction, as stated in Staff Working Paper 6771/11.

Methodologies for identifying and quantifying the contributions that can be subtracted from measured concentrations are described and discussed in the Staff Working Paper 6771/11, for the following natural sources:

- Transport of natural particles from dry regions outside the Member State
- Sea spray
- Volcanic eruptions & seismic activities
- Wild-land fires.

Although atmospheric re-suspension of particles with natural origin from dry regions inside and outside the Member State is explicitly identified in the definition of the contributions from natural sources in the Directive 2008/50/EC and is therefore eligible for subtraction (Table 1.1, code G1), it was not possible so far to identify a method to estimate its contribution quantitatively. It is important to note, however, that this source refers to wind-blown dust, not to the abrasion of the road surface and the degradation of tyres due to friction on hard surfaces (e.g., pavement).

Table 1.1. Natural events causing limit value exceedances: standard codes (Table 5, Commission Decision 2004/461/EC).

Natural event code	*Description*
A1	*Volcanic eruption inside the Member State*
A2	*Volcanic eruption outside the Member State*
B1	*Seismic activity inside the Member State*
B2	*Seismic activity outside the Member State*
C1	*Geothermal activity inside the Member State*
C2	*Geothermal activity outside the Member State*
D1	*Wild-land fire inside the Member State*
D2	*Wild-land fire outside the Member State*
E1	*High wind event inside the Member State*
E2	*High wind event outside the Member State*
F1	*Atmospheric resuspension inside the Member State*
F2	*Atmospheric resuspension outside the Member State*
G1	*Transport of natural particles from dry regions inside the Member State*
G2	*Transport of natural particles from dry regions outside the Member State*

According to the Commission Decision 2004/461/EC, the possibility to subtract contributions derived from natural events before comparing the ambient air pollutant concentrations with the limit values does not mean that pollutants of natural origin are

not adversely affecting health. Member States should therefore always consider all appropriate action to eliminate or reduce excessive exposure to air pollutants.

1.2. Regulated pollutants affected by natural sources

The present working paper aims to assess the exceedances due to natural causes reported by Member States in the years 2008 and 2009. Regarding the pollutants which may be affected by natural sources, the Working Paper refers at all times to "regulated pollutants exceeding the air quality limit values set in the EU ambient air quality legislation", with no specification as to which pollutants are to be considered. Based on the questionnaires reported by Member States to the Commission for 2008 and 2008, the only metric for which natural exceedances were reported was PM_{10}. No natural exceedances of the SO_2 limit values were reported by any Member States, which might have been linked to volcanic eruptions inside or outside the Member States (codes A1 and A2) and/or to geothermal activity inside or outside the Member States (codes C1 and C2).

Therefore, the present working paper will deal only with reporting of natural exceedances of the PM_{10} daily and annual limit values by the Member States. Throughout the text, the terms atmospheric particles, airborne particles, particulate matter and aerosols are used interchangeably. PM_{10} is the size fraction of these particles with aerodynamic diameter <10 microns, and for which limit values were established in the Air Quality Directive 2008/50/EC.

2. Occurrence of natural events in EU

2.1. African dust

2.1.1. Synoptic-scale air mass transport patterns

The determination of the levels of atmospheric particulate matter (PM) is a key parameter in the evaluation of air quality, due to the proven influence of this pollutant on human health, climate and the ecosystems. On a global scale, 50% of the PM mass concentrations are of mineral origin, sea spray being the main contributor to the remaining 50% (IPCC, 2007). Both are predominantly emitted by natural sources. Most of the mineral dust is released to the atmosphere from arid or semiarid areas. The major dust source areas are located in subtropical latitudes of the Northern Hemisphere, and extend from the West coast of North Africa, the Middle East, central and South Asia to China (Prospero et al., 2002). North Africa (Sahara desert and Sahel belt), the Arabian peninsula (Arabian desert) and western Asia (Lot and Thar deserts) are considered the greatest sources of soil dust (León and Legrand, 2003; Prasad and Singh, 2007) able to inject yearly several Tg of dust particles into the atmosphere.

North Africa is considered a typical example of a hot desert area where rain is extremely rare (1 per 100 days registered precipitation during a few hours in Western Sahara, and 2 per 1000 days in the eastern sector, known as the absolute desert, Dubief, 1979). In such environments with extremely low precipitation levels and very high temperatures (60-65 ^{o}C in summer) there are situations very favourable for the massive resuspension of huge quantities of particulate matter. The warming of the surface during the day produces strong vertical thermal turbulences (that can reach altitudes of up to 4000-5000 m in summer, Dubief, 1979) followed by periods of nocturnal stability. Such alternations inhibit the re-deposition of resuspended particles injected into high atmospheric levels (Moulin et al., 1998), increasing aerosol residence

times to weeks or even months, and producing the so-called “dry smog”. Material from this semi-permanent reservoir of dust in resuspension is transported long distances by different mechanisms.

In Europe, African dust may greatly increase ambient PM levels, especially in southern European countries (Bergametti, et al., 1989), where it is a known source causing exceedances of the PM thresholds (Querol et al., 1998; Rodriguez et al., 2001; Viana et al., 2002; Escudero et al., 2005, 2007; Gerasopoulos et al., 2006; Kallos et al., 2006; Kocak et al., 2007; Mitsakou et al., 2008; Pey et al., 2009).

According to a number of studies, different meteorological scenarios can lead to the transport of African dust air masses towards Europe.

For the Western Mediterranean, Rodríguez et al. (2001) and Escudero et al. (2005) defined 3-4 scenarios typically favouring the transport of mineral matter from the Sahara and Sahel deserts. As exemplified in Figure 2.1, the transport may be due to:

A. A low pressure system in the Atlantic (in front of Portugal) and/or over the Morocco area. These scenarios mainly occur in early autumn and spring.
B. A high pressure system (at surface level) over the Iberian Peninsula-Western Mediterranean and/or over the North of Africa. These situations are generally observed in January-March. They give rise to the development of dust plumes over the Atlantic with a well defined convex morphology. After that, the plumes can be transported towards the Iberian Peninsula from the West.
C. A North African anticyclone located above 850 hPa. This is the commonest scenario transporting African dust towards Western Europe, mainly occurring in summer.

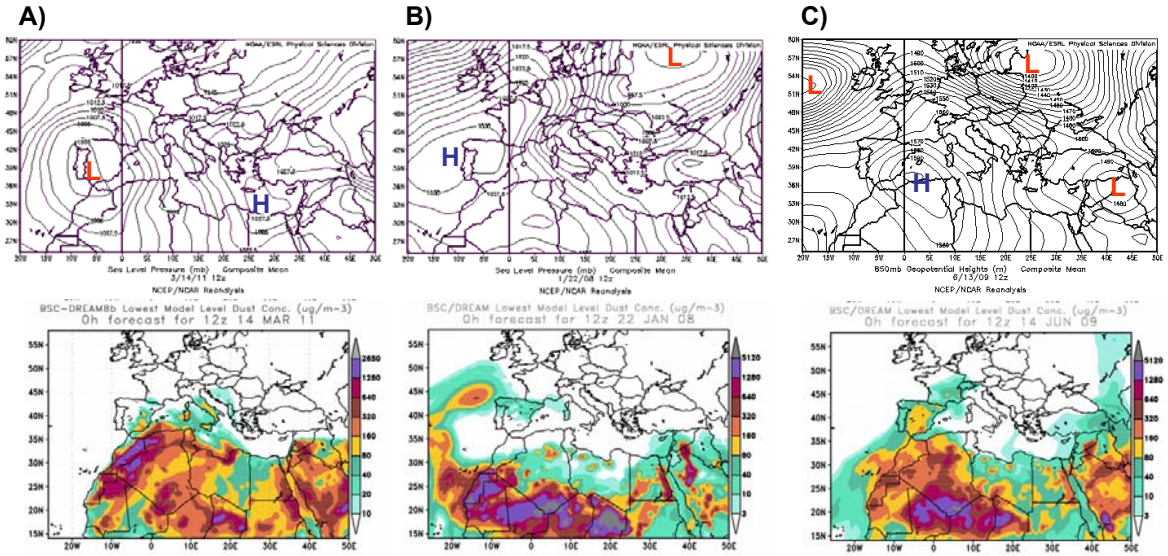

Figure 2.1. Main scenarios giving rise to African dust transport towards the western Mediterranean Basin.

For the Central and Eastern Mediterranean, Nickovic et al. (2001); Kallos et. al (2006 and 2007) and Meloni et al. (2008) identified the two main meteorological situations (Figure 2.2) responsible for the transport of large amounts of mineral dust particles. In spring and early summer, the development of Saharan thermal lows in the South of Atlas takes place under the influence of the strong thermal contrast between the temperature of the cold marine waters and the warm continental surfaces (Moulin et al., 1998). These cyclones travel eastward (**D** scenario) along this thermal gradient and

finally cross the Mediterranean between Libya and Egypt, constituting the main atmospheric scenario responsible for the transport of desert dust over the Eastern Mediterranean Basin (EMB). Severe episodes (**E** scenario) can also be associated with the combination of a deep trough over West Mediterranean and NW Africa and relatively high pressures to the Eastern part of the Mediterranean which is related with the evolution of "**A** scenario" described previously.

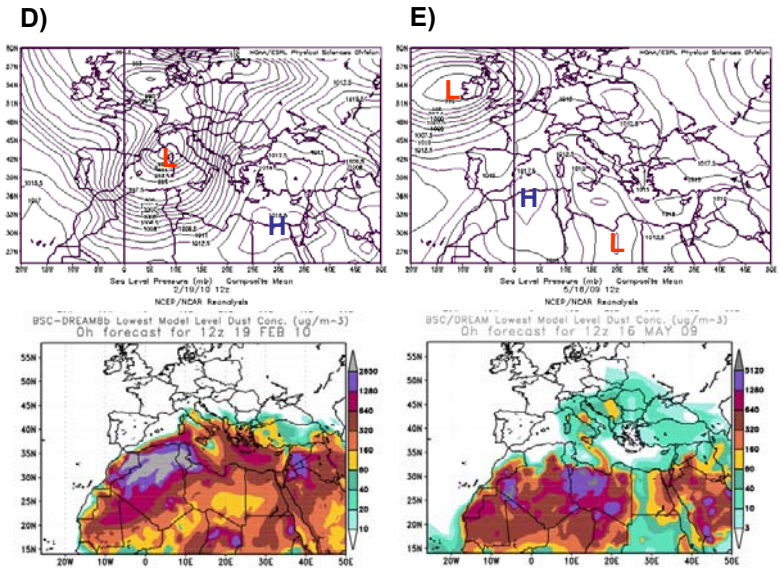

Figure 2.2. Main scenarios giving rise to African dust transport towards the eastern Mediterranean Basin.

A recent paper on African dust in the Mediterranean region (Querol et al., 2009) summarizes the seasonality, occurrence and intensity of African dust episodes over the whole Mediterranean Basin. In that study, a number of PM_{10} data series from monitoring sites across the Mediterranean European region where evaluated. The study revealed clearly increasing PM_{10} trends from the north to the south and from the west to the east of the Basin, almost coincident with the PM_{10} African dust load (Figure 2.3). The authors noted that the PM_{10} background levels were 5-10 $\mu g/m^3$ higher in the EMB when compared with those in the western Mediterranean Basin (WMB). This is due to higher anthropogenic contributions in the latter.

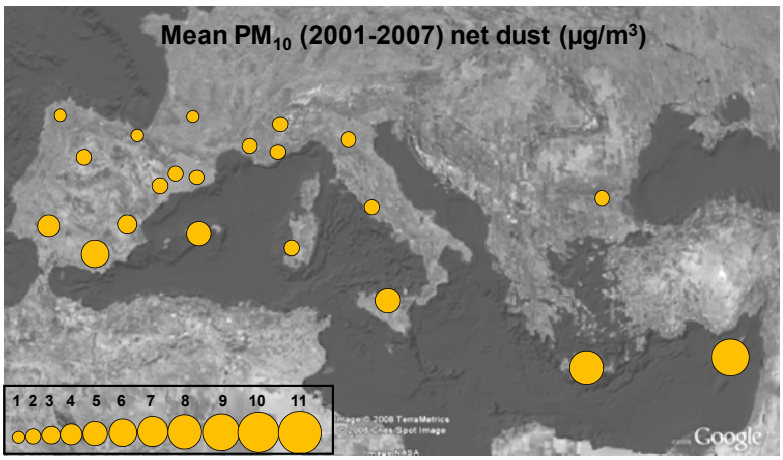

Figure 2.3. Mean annual (2001-2007 average) net African dust contribution to PM_{10} levels across the Mediterranean Basin (adapted from Querol et al., 2009).

Marked PM_{10} seasonal trends were evidenced when comparing eastern and western sites, largely driven by the occurrence of African dust events. Thus, the higher

frequency of dust events in spring-early summer over the eastern causes higher PM levels. Likewise, the summer maximum observed in the western part roughly coincides with the most intense period of African dust outbreaks, although in this region re-circulations of aged air masses significantly increase the background PM_{10} levels (Figure 2.4).

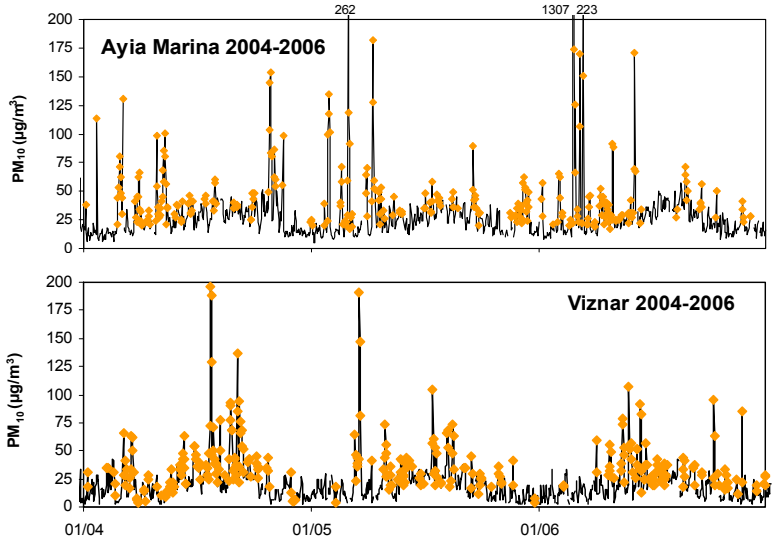

Figure 2.4. Daily PM_{10} levels at Ayia Marina (Cyprus) and Viznar (SE Spain) in the period 2004-2006. Orange diamonds spot African dust episodes (adapted from Querol et al., 2009).

Querol et al. (2009) also found important inter-annual variations in the dust contribution, more evident at the southern sites. These variations were generally associated with the occurrence of extreme dust events. Generally, the years with unusually high dust contributions over the EMB corresponded with anomalously low contributions over the WMB, and vice versa (Figure 2.5).

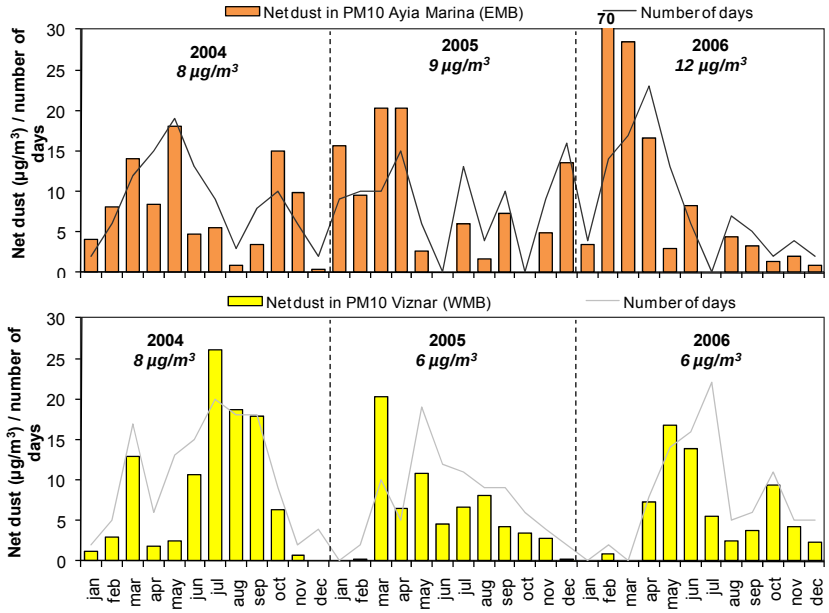

Figure 2.5. Monthly number of African dust episodes and net dust contributions to PM_{10} at Ayia Marina (Cyprus) and Viznar (SE Spain) in the period 2004-2006. (adapted from Querol et al., 2009).

2.1.2. Chemical composition of mineral dust

Globally, the major source areas of mineral dust are the Sahara and Gobi deserts. Aeolian processes in North Africa annually transfer vast amounts of high Ca-, Al-, Si-, K- and Fe-containing mineral particulates westwards to the Atlantic, Caribbean and to America, as well as northwards to the Canary Islands and mainland Europe (e.g. Carlson and Prospero, 1972; Bergametti, et al., 1989; Prospero et al., 1981; Moulin et al., 1998; Karyampudi et al., 1999; Alastuey et al., 2005; Gerasopoulos et al., 2006; Mitsakou et al., 2008; Rodríguez et al., 2011). Most of these particles travel via the Sahara-Sahel Dust Corridor (SSDC: Moreno et al., 2006), the World's largest aeolian dust source (Figure 2.6). However, the regular dust intrusions moving from the African continent will derive from different sources and therefore contain different mixtures of mineral particulate matter. In consequence, the analysis of the chemical composition of mineral dust reflects this mixture of components, and as a result it should be combined with other tools (see section 2.1 below) for the correct identification of African dust transport processes. A certain general characterisation of the chemical composition of mineral dust as a function of source regions may be provided:

i. The Saharan basement massifs: the particulates coming from this source are relatively rich in the more soluble major elements (Na, K, Ca and Mg) and contain abundant Rare Earth Elements (REEs). Much of this particulate material may be viewed as sedimentologically and therefore geochemically immature, having been derived relatively recently from the weathering and erosion of desert exposures of igneous and metamorphic rocks (Evans et al., 2004).
ii. Saharan sedimentary basins: this group of particulates is depleted in primary mafic minerals (silicate minerals rich in magnesium and iron) and therefore have a simple felsic (silicate minerals enriched in lighter elements such as silicon, oxygen, aluminium, sodium, and potassium) composition comprising mostly clay minerals, quartz and diatoms. Such particles typically have a polycyclic history involving repeated fluvial and aeolian transport and sedimentation that can be traced back millions of years (Evans et al., 2004) and so may be regarded as geochemically mature.
iii. Atlantic margin-type particulates: many of these aerosols have a geochemistry strongly influenced by the weathering and erosion of Mesozoic-Cenozoic marine limestones and marls. Such materials are rich in carbonate (and therefore Ca, Mg and Sr), and commonly register the presence of hematite and mafic clays such as palygorskite, and depletion in Ti, Nb, Ta and Rb.
iv. In addition to these sources, regional winds (Harmattan and Monsoon winds) may also largely influence the chemical composition of resuspended soil dust. The Harmattan winds transport basinal diatomaceous dusts westwards, mixing them with particulates from the surrounding basement massifs, and thus enriching them in "hard rock" minerals such as hornblende, and trace elements (Ba, Rb, Zr, Hf, REEs and Th). In contrast, the summer kaolinitic monsoon dust blows into the SSDC from sub-Saharan Africa, and will have a chemistry reflecting deeply chemically weathered terrains, with the more immobile elements (notably Zr, Hf and REEs).

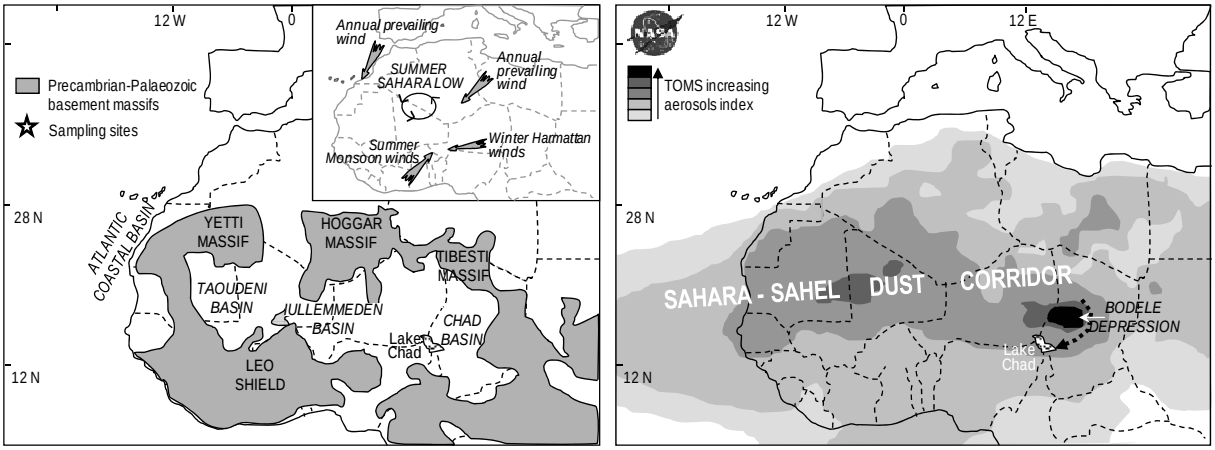

Figure 2.6. Maps of the Sahara-Sahel Dust Corridor showing (left) outcrop of old basement rocks and young sedimentary basins, prevailing winds (inset), and (right) TOMS 13-year (1992-2005) averaged aerosol concentrations, highlighting the dust source hotspots of the Bodélé Depression and Taoudeni Basin (http://toms.gsfc.nasa.gov/aerosols/aerosols_v8.html). Modified from Moreno et al. (2006).

Studies under the Scanning Electron Microscope (SEM) analysing particulate matter (PM) samples from the west and east Mediterranean (including Cyprus, Crete, Israel and Spain) during North African dust episodes have shown east Mediterranean samples as being the most calcareous airborne samples (richest in calcite, with silicates being more common in the finer $PM_{2.5}$ and carbonate in the coarser PM_{10}), whereas the samples from Spain were the most siliceous (Querol et al., 2009, Figure 2.7).

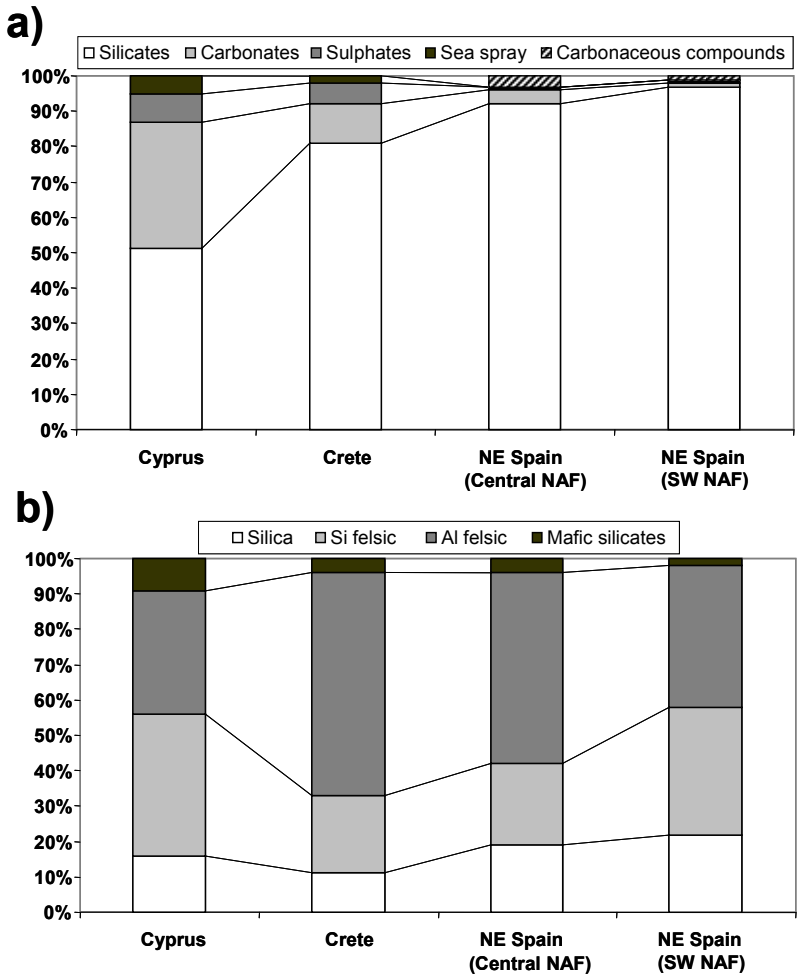

Figure 2.7. a) Scanning Electron Microscopy (SEM-EDX) PM composition (in % number of particles, not mass) for PM_{10} filter samples collected during north African pollution episodes in locations from Cyprus, Crete and NE Spain (the latter including air masses coming from central and SW Africa). b) Idem, considering only silicate compositions. From Querol et al. (2009).

We can conclude therefore that the geochemistry of mineral dust cannot be used as a single tracer of African dust transport, and that it should be combined with other tools available for the proper identification of African dust events. Despite this, subtle geochemical differences may be detected between dusts from different African source areas. Samples derived from Northern, western, central and eastern Sahara (which are most typical of summer intrusions in the Mediterranean, Querol et al., 2009) show an increased proportion of quartz, calcium carbonate and illitic clays, with lower proportions of kaolinitic clays, as compared with samples originating from the southern Sahara and Sahel. The latter, which are more typical of dust intrusions early in the year (February-March) show a composition more consistent with the presence of quartz, kaolinitic clays and iron oxides. In general the chemical composition of PM samples dominated by African dusts will consistently show higher concentrations of typically "crustal" major elements (Si, Al, Ca, Fe, Mg, K) and trace elements (Rb, Sr, Cs, Nb, La and Ce), with such elements being more abundant in PM_{10} than $PM_{2.5}$. The geochemical signature of anthropogenic pollution episodes is characterised by the fact that more anthropogenic metal pollutants compared to crustal material tend to be concentrated in $PM_{2.5}$ rather than PM_{10}. This indicates their fine size and emphases their man-made origin (Allen et al., 2001; Utsunomiya et al., 2004; Birmili et al., 2006).

2.2. Sea salt aerosols

The ocean is an integral part of the climate system. It contains almost 96% of the water in the Earth's biosphere and is the dominant source of water vapour for the atmosphere. Given that oceanic surfaces cover 71% of the Earth's surface and considering the size and potential importance of the air-ocean interface, the ocean chemical composition is surprisingly poorly characterized for certain compounds. For many important chemical species in the atmosphere the role of the ocean remains the greatest uncertainty in the budget (Fowler et al., 2009). The marine aerosol comprises primary and secondary aerosol components. Primary marine aerosol or sea-spray aerosol is a major source of global natural aerosol mass and is important for global climate. The primary aerosol production results from the interaction of wind stress at the ocean surface and results in the mechanical production of sea-spray aerosol (sea spray being the combination of inorganic sea salt and organic matter). The main component of sea salt is the inorganic fraction, consisting of NaCl with traces of Mg and SO_4^{2-}. Sea spray is produced via the bubble-bursting processes typically resulting from whitecap generation, leading the production of film and jet drops, resulting in sea-spray particles in the range of sub-micrometre size up to a few micrometres.

Although the dominant mass fraction of sea-spray aerosol is sea salt, field measurements at Mace Head (Ireland) suggested a significant biogenic primary source of marine organic components (O'Dowd et al., 2004). In particular water insoluble organic fraction in fine marine aerosol collected during periods of phytoplankton bloom in the North Atlantic was observed to be dominant. It was argued that the water-insoluble organic fraction, dominating the organic composition in the fine size fraction, was likely to be derived from bubble-mediated production. Facchini et al. (2008) showed that submicron organic matter was almost entirely water insoluble (WIOM) and consisted of colloids and aggregates exuded by phytoplankton. The authors conclusively confirmed that the WIOM component observed in marine air samples relate to primary aerosol production. The chemical composition of the vast majority of marine secondary organic aerosol (MSOA) still remains to be identified (O'Dowd and De Leeuw 2007). The most relevant MSOA is methanesulphonic acid, which is derived from marine biologically produced dimethyl sulphide. Some dicarboxylic acids have been associated with secondary formation mechanisms (i.e. Kawamura and Sakaguchi, 1999) and recently a new secondary organic aerosol component, produced through the reaction of gaseous amines with sulphuric acid has also been found in marine aerosol (Facchini et al., 2008, Muller et al. 2009).

The interest in sea spray is broad and includes its role in chemical reactions (for coupled nitric acid–sea salt see Sorensen et al. (2005); for coupled sulphate–sea salt cycles (see O'Dowd et al. 1999, 2000) and, in particular, its role in climate change (IPCC 2001). Sea salt is the dominant sub-micrometre scatterer in most ocean regions (e.g. Kleefeld et al. 2002; Bates et al. 2006) and dominates the marine boundary layer particulate mass concentration in remote oceanic regions. Here a significant fraction occurs in the sub-micrometre size range (IPCC 2001) and contributes with about 44% to the global aerosol optical depth.

Among the various components of marine aerosol, sea salt (NaCl) is quantitatively the major contributor to the marine aerosol mass (especially in the super-micrometer fraction) and it is therefore the component to be considered when evaluating the natural contribution of marine aerosol to PM loadings. Sea salts tend to occur as episodic events, in relation to strong winds, and are most relevant for the PM_{10} daily mean limit values. Discounting the sea salt contribution can have a relevant impact in the calculation of the exceedances of the daily limit value for PM_{10}. In coastal regions sea salt may contribute with up to 80% of the annual mean particulate mass, e.g. PM_{10}

(Putaud et al.,2004). By analysing 89 sites in Europe, Manders el al. (2010) found the average concentrations above land ranged between 0.3 and 13 μg/m^3, with maximum concentrations found along the Irish coastline. For example, sea salt was a dominant component of particles larger than 0.8 mm at all sites, with a smaller re-suspension component associated with frequent precipitation events in IE (Ceburnis et al. 2006).

2.3. Volcanic dust

Volcanic activity on Earth is focused within active zones of tectonic plate margins, the frequency and type of events varying with the type of boundary. However, the impact of volcanic ash emissions may have a global impact due to the fact that emissions may be injected into the stratosphere. One of the most evident examples is the eruption of Mt. Pinatubo in Philippines on June 12, 1991. The effects of the eruption were felt worldwide, as it ejected roughly 10 billion metric tonnes of magma, and 20 million tons of SO_2. It injected large amounts of aerosols into the stratosphere, more than any eruption since that of Krakatoa in 1883. Over the months following the eruption, the aerosols formed a global layer of sulphuric acid haze. Global temperatures dropped by about 0.5 °C (0.9 °F), and ozone depletion temporarily increased substantially (Rantucci, 1994).

The amount and composition of volcanic emissions depend on the thermodynamic conditions in the volcanic edifice (pressure, temperature) and on the magma type. The main compounds emitted include water vapour, ash, CO_2, SO_2, HCl, HF and HBr, and a long list of many other components emitted in lower abundances (von Glasow et al., 2009; Figure 2.8). Amongst the more toxic elements emitted by volcanic eruptions is mercury, with individual volcanoes, such as Mount Etna, emitting levels of this metal that can exceed 500 kg/year (Ferrara et al., 2000). It has been estimated that mercury emissions from this one volcano alone are equivalent to around 5% of all industrial sources in the Mediterranean area (Bagnato et al., 2007).

The sudden eruption of a volcano has the potential to produce transient spikes in PM_{10} levels in EU Member States. Europe has approximately one hundred volcanoes which have been active in the last 10,000 years, of which 30 are in the European Union. In Greece and Italy alone there have been at least 140 eruptions since the 16th century (http://ec.europa.eu/research/leaflets/disasters/en/volcan.html). Within Europe volcanic activity is mostly located in certain islands in the Mediterranean area and on Iceland. Fine volcanic ash from these point sources will normally affect nearby urban areas, but will also be spread widely at high atmospheric altitudes, causing disruption of aircraft flight patterns and ash deposition far from the volcanic source. Emissions of sulphur dioxide (SO_2) from volcanoes will contribute to the formation of secondary particles, further enhancing the PM concentrations. To use Mount Etna as an example, magma degassing from this volcano provides an almost continuous emission of SO_2 (4,000 tons/day) from its volcanic plume (JRC, 2007). In general volcanic emissions cannot be predicted or controlled by human activities, and consequently should be considered as purely natural. At the present time the most active volcanoes in Europe are located in Italy, including Mount Etna (the second-largest active volcano in Europe with major eruptions in the 20th century happening in 1949, 1971, 1981, 1983 and 1991-1993, 2001, 2002-2003, 2006, 2007 and 2008), Mount Vesuvius (located about nine kilometres east of Naples, with eruptions in 1872, 1906, 1929, and 1944), and Stromboli (constantly erupting almost for the last 2,000 years), Greece (where the Santorini island volcano last erupted in 1984), and Iceland (where several volcanoes are highly active including Eyjafjallajoekull which erupted recently, i.e. in 2010).

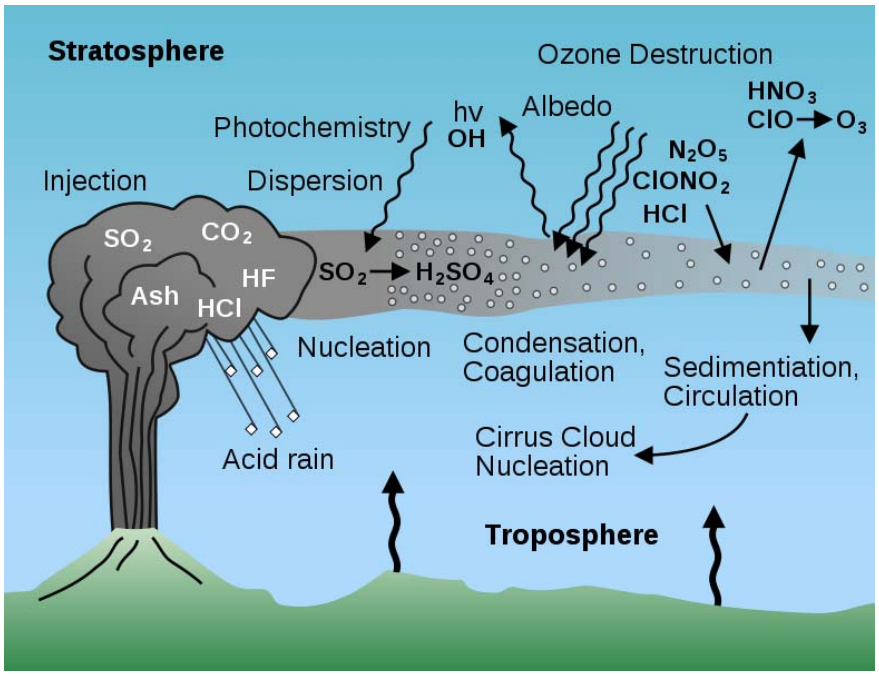

Figure 2.8: Main atmospheric emissions related to volcanic eruptions. Figure in the public domain from the United States Geological Survey.

The most significant documented volcanic air pollution event in Europe was in 1783 when Laki on Iceland erupted, generating SO_2 at a rate 17 times higher than industrial emissions in Europe (1.7 million tons per day during the first 6 weeks of the eruption). In general, volcanic dust emissions are dominated by high aluminium (Al) and silica (Si) contents and characterised by spherical particles derived from high-temperature combustion processes. Volcanic particles are composed of varying proportions of volcanic glass, minerals or crystals and other rock fragments (lithics).

2.4. Wild-land fires

Wild-land fire emissions have a significant effect on atmospheric PM levels, normally during summer time and in forested areas. with recent notable events including widespread uncontrolled combustion in Portugal (2005) and Greece (2007 and 2009). According to the EU Fire Database (http://effis.jrc.ec.europa.eu/fire-history), an average of 95,000 fires occurred annually during the 2000–2005 period in Europe, this resulting in almost 600,000 ha of burnt forestland per year. The emissions from these fires are of special relevance within the Mediterranean countries where summers are drier and hotter, and fire outbreaks are commonly fanned by strong winds. Thus around two-thirds of these fires occur in France, Greece, Italy, Portugal, and Spain where a combined average of half a million hectares of forest land burn every year (Barbosa et al., 2009). A map showing the total area burnt during 2008 in Europe is provided in Figure 2.9. These fires have a pronounced effect on both local and regional air quality. The air quality impacts include both the emission of primary pollutants (PM, CO, NOx) as well as the production of secondary pollutants (O_3, secondary organic aerosol (SOA)) when gaseous pollutants released by fires undergo photochemical processing in the atmosphere (Urbanski et al., 2009).

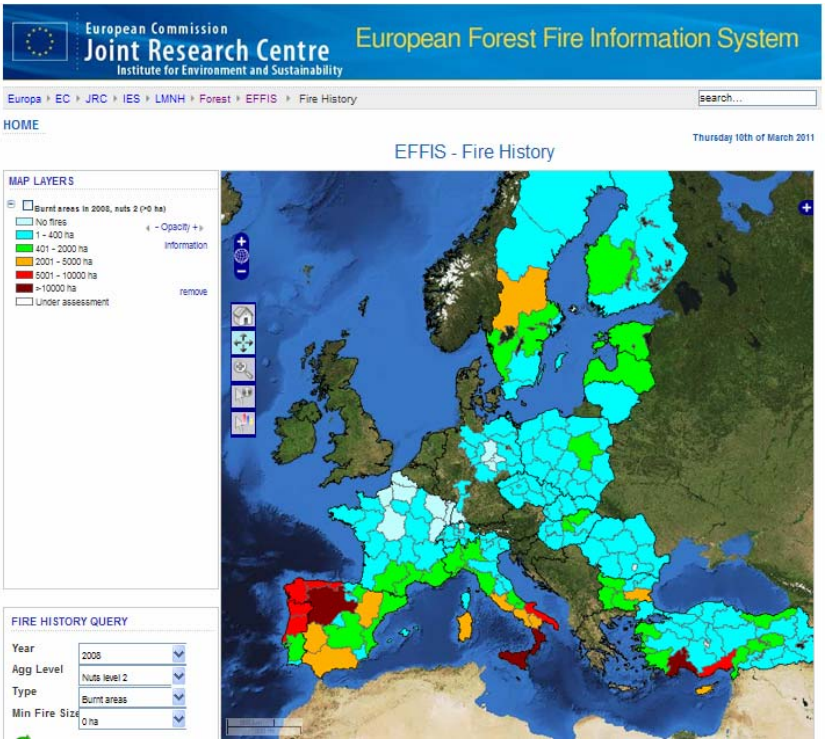

Figure 2.9: Burnt areas during 2008 in Europe. Data from the European Forest data centre (JRC). Available at http://effis.jrc.ec.europa.eu/fire-history.

The main complexity regarding the definition of wildfires lies in the identification of their causes. In order to be considered natural events, wildfires should be strictly caused by natural causes, e.g., lightning. Conversely, human activities causing forest fires may include direct ignition (accidental or on intention) but also littering of forest areas with materials with potential to start a fire (e.g., glass) or the absence of proper forestry strategies (thus resulting in forest areas which are uncared for and prone to suffer burns). The correct identification and further justification of these natural or anthropogenic causes is a key issue with regard to reporting of natural events to the Commission. The Joint Research Centre (JRC) estimates that more than 90% of all Mediterranean fires and 87% of fires in the boreal region of Russia originate from human impacts (Marelli, 2007). Thus as wild-land fires are mainly anthropogenic in origin, they should not be considered as "natural sources of pollution". The assessment of the geographical distribution of forest fires within the Member States may aid in this interpretation, for example, if fires occur in especially humid regions (Figure 2.9), this might suggest anthropogenic influences causing them. Finally, the complexity in the justification of natural exceedances of the limit values due to wildfires increases if the pollution is imported from another country. Thus, according to the document recently published by the Council of the European Union (Staff Working Paper 6771/11), "*if a Member State suffers high PM concentrations due to wild-land fire outside its own country, it may still be appropriate to subtract the contribution from the fire of the total PM levels for compliance purposes. In such situation provisions on transboundary pollution contained in Article 25 of the Directive should also be considered and implemented, especially in case of frequent and reiterated fire episodes".*

3. Identification of natural PM_{10} events: review of existing tools

3.1. African dust

A number of suitable methodologies are currently used to identify the occurrence of dust outbreaks. Generally, a combination of methodologies is the typical way of recognizing these episodes since the use of a single method may lead to wrong interpretations and/or episodes might be overlooked. There are two types of methodologies: those reflecting the qualitative occurrence of these episodes; and those providing a quantitative approach to assess the concentrations of natural dust affecting a given area.

As regards for the qualitative approaches, they include: 1) Interpretation of PM data series together with meteorological information; 2) Atmospheric deposition samples; 3) Aerobiological studies; 4) Radiometric analyses. Among the quantitative methods the most widely used are: 5) Numerical modelling, and statistical analysis; 6) Chemical analysis of PM samples and receptor modelling; finally, 7) Remote sensing studies are qualitative and some may be quantitative.

3.1.1. Qualitative

3.1.1.a) Interpretation of PM data series together with meteorological information

Usually, one of the common methodologies used to identify natural dust outbreaks emerges from the interpretation of the PM data series from a given measurement site. In most cases this interpretation is supported by a number of meteorological tools (including air-mass back-trajectories, meteorological maps, etc.) and/or local parameters measured at the site such as wind direction (from the desert areas), relative humidity (typically decreasing during such events), temperature (generally increasing during these episodes), among others.

Since natural dust outbreaks are usually associated with dramatic increases in PM concentrations, regional background areas (located far away from local sources) are highly recommendable locations to recognize these episodes (Querol et al., 1998; Rodríguez et al., 2001 and 2002; Pérez et al., 2008; Pey et al., 2010). Nevertheless, it is common to find studies based on urban or industrial sites where these episodes can also be identified. In those cases, complementary measurements are needed. For example, in a typical urban area where road traffic emissions are causing most of the PM pollution, peaks of PM are typically concurrent with those of NO_x and CO. This can be similar in industrial environments, where SO_2 increments may be linked to PM peaks. Thus, in such locations the impact of natural dust outbreaks may cause PM peaks not related with those of NO_x, CO or SO_2 emissions (Viana et al., 2002 and 2003; Moreno et al., 2005 and 2006a).

3.1.1.b) Remote sensing

Sun-photometers and other passive remote sensors are able to provide information on the aerosol physical and optical properties, but these column-integrated methodologies do not provide any information on the vertical structure of a PM plume. There is a clear increment in aerosol optical depth at all wavelengths when dust plumes are affecting a given site. The combination of this parameter with the Angström coefficient is generally a useful tool to confirm the existence of dust events. The Angström coefficient is the name of the exponent in the formula that is usually used to describe the dependency of the aerosol optical thickness, or aerosol extinction coefficient on wavelength. Regarding the Angström turbidity coefficients, the arrival of natural dust outbreaks

produces a considerable increase in the Angström coefficient and a dramatic decrease in the Angström exponent, indicating the addition of large particles to the atmospheric column, which is a typical indication that mineral-bearing particulate matter transported from desert regions is involved (Lyamani et al., 2005).

Lidars are increasingly used to characterize the atmosphere, particularly in terms of vertical resolution. Generally, the aerosol backscatter coefficient and extinction coefficient are the most important parameters that can be extracted from Lidar signals. Natural dust plumes are well detected by this technique as shown in a number of studies (Karyampudi et al., 1999; di Sarra et al., 2001; Pérez et al., 2006a; Guerrero-Rascado et al., 2008; Kim et al., 2010). However, lidars are not generally able to characterise the lower 300m of the troposphere, which are the most relevant from of air quality point of view.

The importance of these types of measurements is reflected in the existence of two main global-scale networks on remote sensing: AERONET (NASA/GSFC) and the Asian Lidar Observation Network (NIES). These networks include both aerosol optical depth measurements and vertical section profiles of aerosols. There also exists a regional version (European) of the AERONET network, EARLINET, with the purpose of compiling an aerosol dataset describing vertical, horizontal, and temporal distribution (including its variability on a continental scale). This dataset is also used to validate and improve numerical air quality and climate models that predict the future chemical composition of the atmosphere. The data are thus also important when calculating different scenarios describing possible economic developments, including actions taken to preserve the quality of the environment as well as natural events (which may not be controlled).

Chiapello et al. (1999) used ultraviolet measurements obtained with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) instrument to retrieve information on the distribution of aerosols over oceanic and continental surfaces. They examined the effectiveness of the derived TOMS aerosol index (AI) for the detection of absorbing aerosol in terms of mineral dust aerosol over the North Atlantic Ocean and North Africa by comparing AI measurements with time series of daily aerosol measurements from different locations. They obtained a fairly good response especially with those dust events that yielded daily averages greater than 20 μg m^{-3}.

Satellite data are also used to assess the occurrence of natural dust events (Moulin et al., 1998). SeaWiFS (Sea-viewing Wide Field-of-view Sensor) is providing data on quantitative global ocean bio-optical properties (Chou et al., 2002) but dust plumes are perfectly visible when they are over oceans (http://oceancolor.gsfc.nasa.gov/SeaWiFS/). Similarly, MODIS (Moderate Resolution Imaging Spectroradiometer) is a key instrument aboard the Terra and Aqua satellites, viewing the entire Earth's surface every 1 to 2 days, acquiring data in 36 spectral bands (Remer et al., 2005). These data allow understanding of global dynamics and processes (such as dust plume movements) occurring on the land, in the oceans, and in the lower atmosphere (http://modis.gsfc.nasa.gov/).

3.1.1.c) Atmospheric deposition

When collecting atmospheric deposition, the evidence of dust outbreaks is clear when red rains or red snows are observed. There are several studies worldwide characterizing these types of episodes (Bergametti et al., 1989; Schwikowski et al., 1995; Avila et al., 1998; Kubilay et al., 2000; among others). In addition, other deposition samples are usually collected in dry regions and subsequently studied by microscopy in order to identify dust outbreak impacts. In a recent study performed in

Jordan (Abed et al., 2009) and Lybia (O'Hara et al., 2006), the researchers found that natural dust particles were mainly aluminosilicates (clay minerals, feldspars), quartz, carbonates (calcite and dolomite), with a minor component of phosphates, reflecting the lithology of the North African Sahara. They also study their morphology and they found to be sub-rounded to sub-angular dominated by the 5–20 mm size fraction. Similar findings were reported by Moreno et al. (2006b) and Coz et al. (2009).

3.1.1.d) Aerobiological studies

Long range transport of air masses from desert regions brings not only mineral dust but also other components such as bio-aerosols. A very recent study, performed on the Canary Islands, on the pollen populations according to different air masses origins (Izquierdo et al., 2011) show that during African dust outbreaks some species only typical for desert regions were found. Similar findings were observed by Alastuey et al. (2005) during an intense African dust episode affecting again the Canary Islands. In that case the authors found in the PM samples collected numerous silica skeletons of fresh water diatom from the Melosira genus, present in lakes or ponds of Northern Africa.

3.1.1.e) Radiometric analysis

A number of radiometric elements may be used as tracers of soil dust transport towards a given region. In the case of Canary Islands, recent studies (Hernández et al., 2005 and 2008) focused on the analysis of ^{137}Cs, ^{40}K and ^{7}Be. The authors found a clear parallelism between the African dust inputs and the increments (up to 14 times over habitual levels) in the activity (in μBqm^{-3}) of these radionuclides.

3.1.2. Quantitative

3.1.2.a) Numerical modelling and statistical analysis

Numerical modelling

Modelling of dust plume evolution and concentrations is a worldwide activity. In principle models can be used to quantify the contribution of wind-blown dust for applications in relation to the Air Quality Directive (AQD) if it can be shown that the models fulfil the uncertainty criteria as laid out in the AQD for daily mean PM_{10} concentrations (FAIRMODE, 2011). In practise, however, quantifying daily mean PM_{10} concentrations to the required level of uncertainty (50%) using models only is currently not feasible and their use together with monitoring data, both satellite and ground based, is necessary to reduce the uncertainty. Two examples where models were used to help identifying and quantifying the contribution of wind blown dust from the Sahara, relevant for the AQ Directive, are Mircea et al. (2008) and Mitsakou et al. (2008). Saharan dust forecasts are currently carried out by the University of Athens using the SKIRON forecasting system (Nickovic et al., 2001; Kallos et al., 2005), the Earth Sciences Division of the Barcelona Supercomputing Center (BSC) using the BSC-DREAM8b model (Nickovic et al., 2001; Pérez et al., 2006a;b), the Monterey National Research Laboratories aerosol page and the Tel-Aviv University Weather Research Center. Outputs like concentrations close to the Earth's surface, total atmospheric dust load, dry and wet deposition, and vertical profiles of dust may be obtained from some of these models. A number of, but not all, regional scale air quality models, such as CHIMERE (Vautard et al., 2005) contain also modules that describe windblown dust emissions. Back trajectory modelling may be carried out with a number of models. Models commonly used for such applications including FLEXTRA and FLEXPART

(Stohl et al., 2002 and Stohl 2009) and HYSPLIT (ARL, 2009). Both models have been used for a variety of applications related to the origin of natural emissions.

Similarly, a regional Asian dust forecasting model called ADAM (Asian Dust Aerosol Model) is giving information about dust export from Asian deserts (including Gobi Desert, Loess Plateau, Badain Jaran Desert, Tengger, Mu Us, Hunsandake and Horqin) towards the adjacent areas (Park and In, 2003). Furthermore, there are global models simulating dust production, export, and deposition around the world. These are mainly HYSPLIT (Draxler et al., 2001), currently including substantial improvements (Escudero et al., 2006; Draxler et al., 2010); NAAPS-Naval Research Laboratory (Hogan and Rosmond, 1991; Hogan and Brody, 1991).

Statistical analysis
Recently, Escudero et al. (2007) and Pey (2008) developed a statistical methodology to quantify daily African dust load in PM_{10}. This methodology was developed in collaboration between Spain and Portugal, and it is publicly available at the Spanish (http://www.marm.es). It is the official methodology applied by both EU Member States and recommended by the European Commission (Staff Working Paper 6771/11). African dust events are detected by means of a set of tools including synoptic charts, aerosol maps and back-trajectory analysis. Once the episodic days are identified, this method assumes that during an African dust event, at a given regional background site, PM_{10} may be divided into the regional contribution and the African dust input. Once the PM_{10} regional load is determined, the net African dust input can be obtained by subtraction. As demonstrated by Escudero et al. (2007) and Pey (2008), a very good approach to assess the daily regional background load can be obtained by applying a monthly moving 40^{th} percentile to the PM_{10} time series from a given regional background station (excluding those days coincident with African dust inputs). The 40^{th} percentile is a good representative of the background concentrations during advective conditions. The validity of this methodology was assessed by comparing the estimated net dust load during African dust outbreaks at three regional background sites and the mineral matter determined by chemical speciation of PM_{10} samples.

3.1.2.b) Chemical analysis of PM samples and receptor modelling

Chemical analysis
One of the most precise ways to determine the mineral dust load affecting a given area emerge when performing a chemical analysis of collected samples. Chemical composition may be determined by using a number of different analytical methodologies (real time measurements; total digestion of samples and subsequent analysis by different techniques; X-Ray Diffraction and Particle induced x-ray emission are among the most commonly used methods). In all the cases the required result is given by the bulk of mineral matter encountered in the PM samples. In many cases the mineral dust may not only correspond to long range transport from desert regions but it is a mixture between local, regional and distant sources (Amato et al., 2009). In those cases, the interpretation of elemental ratios (Si/Al; Ca/Fe, etc) may help to relate to natural dust outbreaks. A recent paper (Zhao et al., 2010) reported results for Si/Al ratios from samples collected in northern and southern urban areas of China. The authors observed a clear depletion in Si when natural dust outbreaks occurred. This kind of methodology to identify natural dust outbreaks has been used in the past to document African dust impacts in Illinois-USA (Gatz and Prospero, 1996), in the Virgin Islands (Perry et al., 1997) and in the Amazon Basin and Israel (Formenti et al., 2001). Similar tests have been made by using Ca/Fe ratios in order to link to natural dust occurrences (Gatz and Prospero, 1996; Makra et al., 2002).

Nevertheless, in order to minimize the interference of non-long range transport sources the use of measurements at regional background monitoring sites is highly recommended. In those locations, most of the mineral matter can be of an external origin (long-range transport, Marenco et al., 2006; Salvador et al., 2007; Pey et al., 2009).

Receptor modelling
A number of studies have identified and quantified the natural dust contributions by using receptor modelling techniques. To carry out those studies a dataset of chemical composition is needed. The main output of such receptor modelling activities is the distinction between different mineral dust sources; one of them is the contribution from long range transport processes. Examples of the application of receptor modelling with this purpose are Whang et al. (2007); Nicolás et al. (2008); Koçac et al. (2009); Viana et al. (2010)

3.2. Sea salt aerosols

A means to calculate the sea salt contribution is to rely on the average sea water composition. Sea salt aerosol consists mainly of chloride (55.1% by weight) and sodium (30.6% by weight), with a ratio Cl/Na of 1.799 (in ppm). In theory, chloride could be used as a reference ion to calculate the sea salt contribution but the samples can be subjected to chloride depletion due to interaction with HNO_3 or H_2SO_4, or affected by anthropogenically-emitted HCl. Conversely, water-soluble sodium is a conservative tracer, it has a limited number of non-sea salt sources where and when dust re-suspension is not intense (Millero et al, 2004, Manders et al. 2010) and therefore may be used as sea salt aerosol tracer. In the “Guidance on the quantification of the contribution of natural sources under the EU Air Quality Directive 2008/50/EC” Staff Working Paper 6771/11, the Commission recommends the chemical determination of both sodium and chloride at each monitoring station for which sea salt contributions are reported.

In addition to chemical determinations, a number of modelling tools are available to simulate the contribution of sea salt in the lower troposphere (Tsyro et al., 2011). There have been a number of studies that have applied modelling as a primary source of information for assessing the contribution of sea salt to exceedances of the PM_{10} daily and annual limit values (FAIRMODE, 2011). The best respective modelling examples have been carried out in the Netherlands where two separate modelling studies were performed. In the first Van Jaarsveld and Klimov (2011) applied the OPS-ST model and in the second Manders et al. (2009) applied the LOTOS-EUROS model to calculate sea salt contributions using a relatively low model resolution of approximately 6x6 km^2. Generally the modelling studies remain quite uncertain on the temporal scale of one day, with estimated uncertainties in salt concentrations of the factor 2-3. However, the long term average concentrations (over five years) are better represented, with an estimated uncertainty of around 15% (Van Jaarsveld and Klimov, 2011).

In Europe, the regional air quality model LOTOS-EUROS (Schaap et al., 2008) is used to simulate the sea salt distribution across Europe. Marine emissions in the European area were previously also modelled during the NATAIR project (Grice et al., 2008). Whilst the spatial and temporal variability of sea salt is represented well by the models, the absolute concentrations of sea salts remain a challenge. This is due to the low understanding of the balance between sea salt sources and sinks. The available emission parameterizations show considerable variability (O’Dowd and de Leeuw, 2007), with an uncertainty of a factor of 2-3 resulting therefore also in uncertainties in the sea salt concentrations. The uncertainty in emissions is due to the different sea salt

aerosol source functions used in the models. However, the good spatial and reasonable temporal resolution justifies the use of LOTOS-EUROS, with an appropriate scaling factor for the assessment of sea salt concentrations. The low ratio of sea salt in $PM_{2.5}$ and PM_{10} indicates that this model could be improved by further investigating the size dependent processes.

In addition to the LOTOS-EUROS model, a number of other regional scale models also contain emission modules for sea salt (e.g. Unified EMEP model and CHIMERE). However, due to the strong gradient of sea salt concentrations from the coast inland the resolution of these models needs to be 10x10 km^2 or less to capture these gradients adequately. In any case, if such models are applied for sea salt calculations they should be well validated or applied in combination with observations. Forecasts for sea salt are also available, e.g. the Tel-Aviv University Weather Research Center provides sea salt forecasts (http://wind.tau.ac.il/salt-ina/salt.html) for the Mediterranean region using the same model (DREAM) that is applied for wind blown dust forecasts. However, this resolution of this model is too coarse (~35x35 km^2) to capture the strong gradient close to the coast. The contribution of sea salt, dust and biogenic secondary organic aerosol (SOA) to the total PM_{10} mass concentration has been calculated for Europe with the TM5 model (Krol et al., 2005). According to the model sea salt had a small contribution to the continental PM_{10} values; however it can be up to 30% in Ireland, Denmark and coastal regions of Western Europe. A comparison of modelled salt concentrations with measured data from the surface observation network was performed in the framework of AEROCOM (see http://nansen.ipsl.jussieu.fr/cgibin/ AEROCOM/ aerocom/surfobs_annualrs.pl). Over Europe there is a good correlation between modelled and measured values, although TM5 overestimates observed sea salt surface concentrations.

Finally, there are also attempts to model the organic component of sea salt aerosols. Biologically driven oceanic sources of organic carbon (OC) were modelled by Spracklen et al. (2008) with two global chemical transport models. However, the model under- predicted the OC concentrations by a factor of 2. During periods of high biological activity, monthly mean concentrations were under-predicted by a factor of 5-20. The biologically driven OC emissions were found to be comparable in magnitude to the fossil fuel burning OC source and increase the simulated global OC burden by 20%. Fine particulate organic matter (POM) emitted by sea spray processes was also modelled by Vignati et al. (2010) and its influence on the aerosol chemical properties at the global scale was assessed by using the off-line global Chemistry-Transport Model TM5 (Krol et al., 2005). To evaluate the approach, modelled POM concentrations were for the first time compared with the measurements of water insoluble organic carbon (WIOC) carried out in the Northern and Southern hemispheres, while Spracklen et al. (2008), following a different approach, compared the total OC. The estimated marine sources of primary sub-micron POM amounted to more than half the estimated anthropogenic non-biomass burning POM emissions. It was concluded that the marine contribution must be considered in order to correctly predict the primary particulate organic matter emissions and concentrations over the oceans. The contribution of marine organic emissions to the air quality in coastal areas of the western United States was studied using the latest version of the US Environmental Protection Agency (EPA) regional-scale Community Multiscale Air Quality (CMAQv4.7) modelling system. It was found that marine organics can increase the concentration of $PM_{2.5}$ by 0.1–0.3 $\mu g/m^3$ (up to 5%) in some coastal cities (Gantt et al., 2010). The study suggested that marine organic aerosols account for a considerable portion of the aerosol mass over the remote ocean and some near-coastal regions and therefore should be considered in future air quality models.

3.3. Volcanic dust

The identification of natural events related to volcanic eruptions is normally detected by several countries at the same time. Such events can also be easily observed by using satellite data. Contributions to ambient atmospheric PM from volcanoes, although infrequent, can affect air quality data for an extended period of time after an event (sometimes weeks). In addition to the PM, the tracking of gaseous tracers such as SO_2 at air measuring stations throughout the affected Member States is also useful to observe the development of these events in time and space.

Currently, there is no specific tool to quantify the effect of volcanic eruptions in local and regional air quality networks. Considering the lack of such a specific tool, a summary of a series of possible synergistic tools is listed below. These methods are summarised in the Staff Working Paper 6771/11 "Guidance on the quantification of the contribution of natural sources under the EU Air Quality Directive 2008/50/EC". The tools described are based on the comparison of air pollutant concentration levels at the point or in the area under investigation with levels of rural/remote stations in other areas, levels assessed during periods not affected by volcanic eruptions, and analysis of possible plume trajectories, e.g. using satellite data and/or model calculations. It is recommended to take the following steps:

(a) Study of satellite images and back-trajectories in order to try to determine the impact of the event in time and space.
(b) Modelling of the dispersion of plumes from suspected source areas to demonstrate the relationship between high levels of PM_{10} and SO_2 and this natural event (e.g., with MACC service).
(c) Identify peaks in the concentrations of PM_{10} and SO_2 in long time series.
(d) Compile data of a simultaneous time series for regional background sites relevant to the assessed area. The modelled series that is not obtained through direct measurement has to be thoroughly validated and fit for purpose.
(e) Compare the PM_{10} and SO_2 series from those events with high PM_{10} and SO_2 levels and identify a list of coincident high PM_{10} and SO_2 peaks.
(f) Compile a list of volcanic events that occurred during the period when the time series were taken.
(g) Compare the time distribution of these events with that of the coincident high PM_{10} peaks and review information on gaseous tracers for volcanic emissions (SO_2) to confirm the relationship between these events and the PM_{10} peaks in the reference time series.
(h) Use the average of the PM_{10} and SO_2 concentrations registered in the 15 days before and in the 15 days after the episode in the reference time series as the background concentration. The difference between the concentrations measured during the episode and the above mentioned 30 days (episode days excluded) should be considered as contribution of the volcanic eruptions. In case that the duration of the event is significant compared to 30 days, a more elaborate scheme may be necessary to adequately estimate the concentration levels without the volcanic contribution. Other statistical indicators of the levels excluding the natural contribution at the site/area can be used if properly justified.
(i) Any spatial extent of the contribution needs to be explicitly justified through modelling and back trajectories. Spatial representativeness of the measuring station determined on the basis of averaged time series is most probably not adequate for application during a specific event.

3.4. Wild-land fires

The methodology to quantify the contribution from wild-land fires to local and regional air quality is very similar to the one listed above for volcanic eruptions. Member States should address this possible contribution only if evidence is provided regarding the natural origin of the fire, or if the emissions are transported from regions outside the Member State and when provisions of Directive 2008/50/EC related to the transboundary pollution have been applied. In addition if the fire has extended from another Member State any deductions of the common contribution need to be accompanied by the description of measures taken at the short term to eliminate the fire and reduce the exposure of the population. According to the FAIRMODE guidance document (FAIRMODE, 2011), the Staff Working Paper 6771/11 recommends an integrated approach to determining the contribution of wild-land fires that includes the use of validated air quality modelling and back trajectories, combined with satellite and ground based monitoring as well as chemical analysis of particulate matter. When air quality models are used it is important to validate these with observed data, either ground or satellite based, and to use the best estimates possible for the wild-fire emissions. To quantify wild-land fire emissions explicit knowledge of the burned area, burning period, fuel (biomass) characteristics, fire behaviour, fuel consumption, and pollutant specific emission factors are required (Ottmar et al., 2009). Estimates of emissions at the European scale are already available (Barbosa et al., 2009), based on the EU fire database. This database contains data provided each year by individual Member States for each fire event, burned area maps obtained through satellite images, and a map of fuel types. In addition to this emission information some other aspects, such as plume rise of the wild-land fires, are currently uncertain. There are a number of ongoing studies and projects dealing with wild-land fires, particularly in countries such as Portugal, France, Finland and Greece where there can be significant episodic contributions to air pollution due to forest fires, see e.g. Miranda (2004), Hodzic et al. (2007) and Miranda et al. (2008). Some of these studies show the contribution of transboundary pollution from wild-land fire episodes. In the USA a recent collaborative and coordinated effort to model smoke impacts, the BlueSky Smoke Modelling Consortium was established in order to develop and apply real-time smoke modelling to support fire operations and smoke management (Sestak et al., 2002).

4. Reporting of exceedances of the PM_{10} daily and annual limit values due to natural sources in 2008 and 2009

4.1. Member States and stations reporting exceedances due to natural sources

Reporting of PM_{10} limit value exceedances due to natural events is compiled in forms 23a, 23b and 11h of the Questionnaire. Forms 23a and 23b summarise the monitoring stations in each Member State reporting exceedances of the daily (form 23a) and annual (form 23b) limit values due to natural sources, whereas form 11h compiles the specific days on which the exceedances occur. All forms were available for all Member States reporting natural events with the only exception of IT (in 2009), for which Form 23a and 23b were not available. The data presented in this assessment for IT was compiled manually from form 11h.

The analysis of the data in forms 23a and 23b evidenced that 10 Member States (AT, CY, DE, ES, FR, GB, GR, IT, MT, PT) reported exceedances of the PM_{10} daily limit value due to natural sources in 2008, and 8 in 2009 (CY, ES, FR, GB, GR, IT, LV, PT; Figure 4.1 and Table 4.1). The number of Member States reporting exceedances of the annual limit value due to natural sources was always lower than for the daily limit value, with 7 Member States in 2008 (CY, ES, FR, GB, GR, IT, PT) and 4 in 2009 (CY, ES, FR, GR). This could be due to the low impact of the contributions from natural sources when calculating annual mean levels, but also due to the absence of specific tools to quantify these contributions on an annual scale.

Spatially, in 2008 the Member States reporting the influence of natural sources covered the Mediterranean basin (CY, ES, FR, GR, IT, MT) and also central (AT, DE) and western (GB, PT) parts of the EU. Conversely, in 2009 they were mostly centred around the Mediterranean region (CY, ES, FR, GR, IT) and in western Europe (GB, PT). No exceedances due to natural sources were reported in central Europe or in certain Mediterranean States (e.g., MT). In addition, natural exceedances were reported in northern Europe (LV).

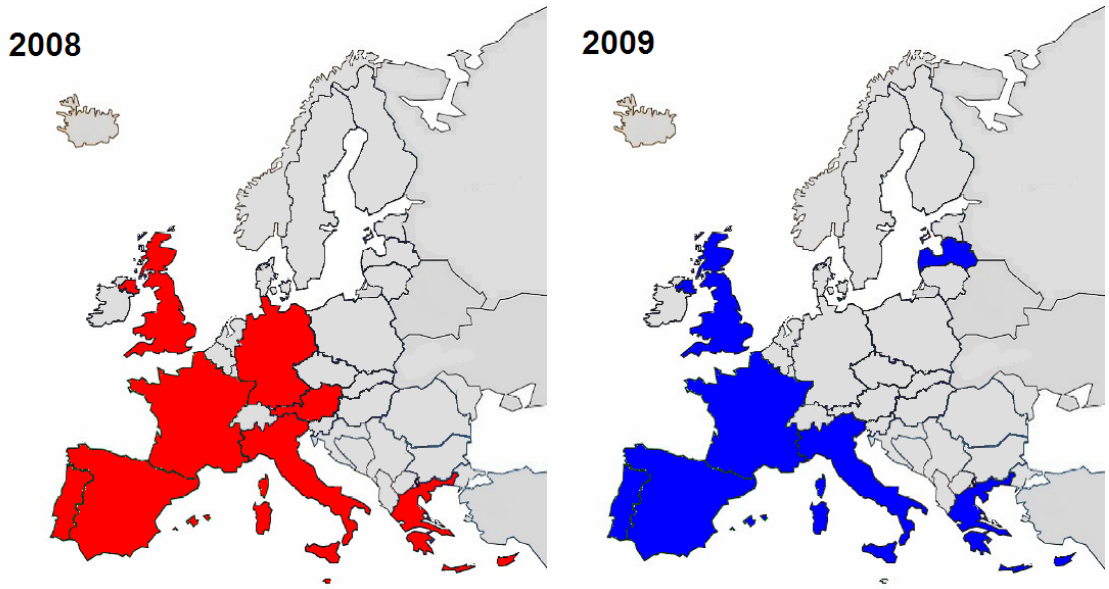

Figure 4.1. Member States reporting exceedances of the PM10 daily limit value due to natural events in 2008 (left, in red) and 2009 (right, in blue).

The Member States reporting natural PM_{10} exceedances for several stations (>30%) were mostly Mediterranean (CY, ES, GR and MT), aside from Latvia. With the exception of Great Britain, Italy and Latvia (Figure 4.2 and Table 4.1), the data evidence a decrease in the number of stations reporting exceedances caused by natural sources from 2008 to 2009. The decrease was especially marked in the case of Spain (123 stations, 30% in 2008 *vs.* 42, 10% in 2009, for exceedances of the daily PM_{10} limit value). Conversely, a marked increase in the number of stations was identified for Italy (4 stations, 1% in 2008 *vs.* 59 stations, 15% in 2009). The reasons behind these different trends are unclear, but they should be at least partly related to the differences in PM levels in the different years. Natural events are only reported when stations surpass the 35 exceedances/year threshold, and therefore in years with high PM_{10} levels it can be expected that larger numbers of stations are included in form 23. However, if this was the only factor affecting the number of stations, then the trend should be similar (either increasing or decreasing) within States in similar geographical regions (e.g., ES and IT). As described above, this was not the case. As an example, mean PM_{10} levels for all stations reporting data did not follow any significant increasing or decreasing trend between 2008 and 2009 in Spain and Italy, with 27±9 µg/m^3 in 2008 and 25±8 µg/m^3 in Spain, and 30±9 µg/m^3 for both years in Italy. Therefore, the general pollution scenario was in this case apparently not the reason for the variability in the number of stations reporting exceedances due to natural events. Thus, there must be other causes, for example the management of stations and/or data flows by the respective Member States.

The types of stations reporting exceedances due to natural events is also different across the Member States (Table 4.2), with e.g. 100% urban sites in France, Great Britain and Latvia, in contrast to 86% in Greece, 63-64% in Italy and Spain, and 33% in Portugal (in 2009, Table 4.2). This variability in types of stations seems to reflect the variability in the distribution between each type of station across Member States, given that no specific trend can be observed. In 2008, the majority of stations reporting PM_{10} exceedances due to natural events were urban and traffic sites in all Member States, whereas in 2009 they were urban sites but classified as both traffic and background.

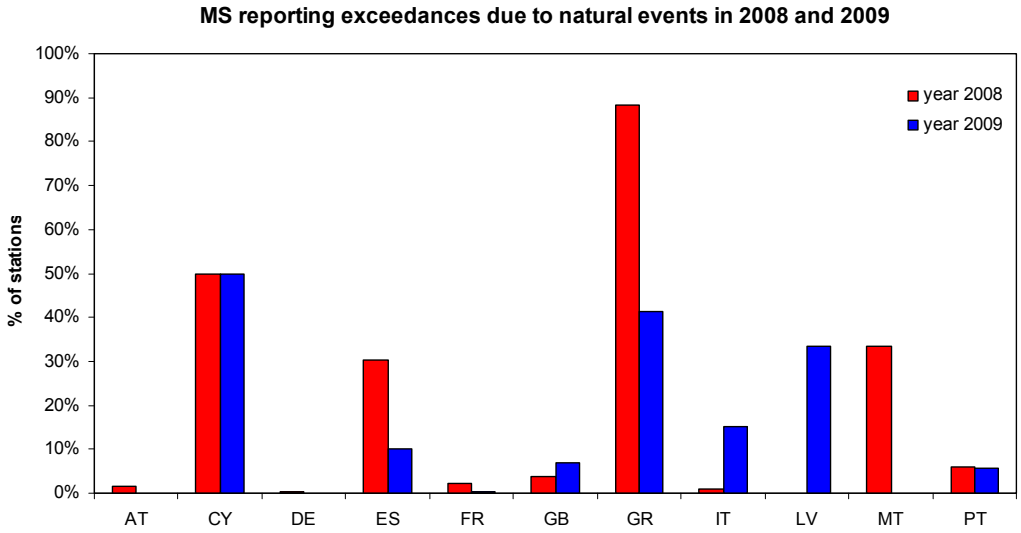

Figure 4.2. Comparison between the proportion of stations reporting PM_{10} exceedances due to natural sources in 2008 and 2009 with respect to the total number of stations in each Member State.

Table 4.1. Member States reporting exceedances of the PM_{10} daily and annual limit values due to natural events in 2008 and 2009. LV: limit value. The values in parenthesis represent the % with respect to the total number of stations reporting PM_{10} data in each Member State. For Italy no data were available regarding the natural contributions to exceedances of the annual limit value in 2009 (form 11h).

MS reporting exceedances due to natural sources							
2008				2009			
Daily LV	Nr. stations	Annual LV	Nr. stations	Daily LV	Nr. Stations	Annual LV	Nr. stations
AT	2 (1%)	CY	1 (50%)	CY	1 (50%)	CY	1 (50%)
CY	1 (50%)	ES	41 (10%)	ES	42 (10%)	ES	12 (3%)
DE	2 (0.5%)	FR	3 (1%)	FR	1 (0.3%)	FR	1 (0.3%)
ES	123 (30%)	GB	1 (1%)	GB	5 (7%)	GR	6 (35%)
FR	8 (2%)	GR	12 (71%)	GR	7 (41%)		
GB	3 (4%)	IT	1 (0.3%)	IT	59 (15%)		
GR	15 (88%)	PT	1 (2%)	LV	3 (33%)		
IT	4 (1%)			PT	3 (6%)		
MT	1 (33%)						
PT	3 (6%)						

Table 4.2. Types of stations reporting PM_{10} exceedances due to natural causes in 2008 and 2009 (as total number of stations and as % of each type of station with respect to the total number of stations reporting exceedances due to natural causes). ND: no data available.

2008												
	Number						%					
MS	Industrial	Traffic	Background	Urban	Suburban	Rural	Industrial	Traffic	Background	Urban	Suburban	Rural
AT	0	1	1	2	0	0	0	50	50	100	0	0
CY	0	1	0	1	0	0	0	100	0	100	0	0
DE	0	2	0	2	0	0	0	100	0	100	0	0
ES	44	42	37	64	52	7	36	34	30	52	42	6
FR	0	6	2	4	2	2	0	75	25	50	25	25
GB	0	2	0	2	0	0	0	100	0	100	0	0
GR	3	9	3	11	4	0	20	60	20	73	27	0
IT	0	4	0	4	0	0	0	100	0	100	0	0
MT	0	1	0	1	0	0	0	100	0	100	0	0
PT	0	2	1	2	1	0	0	67	33	67	33	0

Table 4.2. Continued.

2009												
	Number						%					
MS	Industrial	Traffic	Background	Urban	Suburban	Rural	Industrial	Traffic	Background	Urban	Suburban	Rural
CY	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
ES	12	19	11	27	13	2	29	45	26	64	31	5
FR	0	0	1	1	0	0	0	0	100	100	0	0
GB	0	2	0	2	0	0	0	100	0	100	0	0
GR	1	5	1	6	1	0	14	71	14	86	14	0
IT	8	17	32	36	13	8	14	30	56	63	23	14
LV	0	3	0	3	0	0	0	100	0	100	0	0
PT	1	1	1	1	2	0	33	33	33	33	67	0

4.2. PM_{10} daily limit value: assessment of the exceedances due to natural sources

The specific stations and the number of exceedances of the PM_{10} daily limit value due to natural events reported by each of them in 2008 and 2009 are show in Figure 4.3 (in total, 162 stations in 2008 and 121 in 2009). For 2008, 31 Spanish stations are not shown in Figure 4.3 because the exact number of exceedances was not reported (instead, ">35 exceedances" was indicated in form 23a). The number of days per station with exceedances due to natural causes can be considered fairly similar in both years, ranging between 2 and 82 in 2008 and between 1 and 77 in 2009. Only one exception was identified in 2009 (station FR38012) which reported 128 exceedances of the PM_{10} daily limit value due to natural sources (specifically, "embruns marins", not coded in Commission Decision 2004/461/EC). The number of exceedences reported by this station was a clear outlier when compared to the rest of the stations in 2009.

As expected, the highest number of PM_{10} exceedances per station due to natural sources was reported by Mediterranean Member States (Cyprus, Spain, France and Greece, no data available for Italy in 2009, Figure 4.4). With the exception of France in 2009, this is linked to the fact that the cause for exceedances was mainly transport of natural particles from dry regions outside the Member State and natural source(s) or natural event(s) (codes G2 and S8 from Commission Decision 2004/461/EC). This will be further described below. The natural exceedances in France in 2009 were justified as "embruns marins", interpreted as sea spray. Conversely, the lowest numbers of exceedances due to natural sources per station were reported by Austria, Germany and Latvia.

The number of exceedances due to natural sources reported per station and year is subject to a large variability given that it depends on the meteorological characteristics of each year, which rule the dispersion and transport mechanisms of particulate matter emitted during natural events as well as the accumulation of anthropogenically emitted pollutants. Figure 4.4 shows the range of exceedances of the daily limit value due to natural causes as reported per Member State in 2008 and 2009. The results suggest that the broadest ranges of PM_{10} exceedances were reported by Mediterranean Member States (Spain, France, Greece). The number of exceedances in Spain (2008) ranged between 3 and 43, with similar values to those obtained for France (2-47) and Greece (9-67, Table 4.3). In contrast, lower numbers of exceedances (due to African dust, as will be described below) were reported by Portugal (6-20 in 2008). Thus, the number of exceedances reported per station was relatively similar in the different Mediterranean regions, with an increasing trend from the Western to the Eastern Mediterranean basin. This trend coincides with the results published by various authors (Moulin et al., 1989; Querol et al., 2009) and with the characteristic atmospheric transport patterns of African dust in Mediterranean regions.

Finally, distance of a Member State from the African deserts is also a factor to be taken into consideration, probably with the exception of Portugal, a country which is in close proximity to the African deserts but usually with a lower impact of African dust due to the fact that dust transport patterns are preferentially east-bound. Of course, the range of exceedances due to natural causes depends on the number of stations reporting data; i.e., only one station reported natural exceedances on Cyprus (82 cases in 2008) and also on Malta (21 cases). However, the number of exceedances reported seems to be of the same order of magnitude as those reported by other Mediterranean Member States, even if the number is slightly higher in the case of Cyprus. On the other hand, the number of exceedances due to natural causes reported by the only station in France in 2009 (128, Table 4.3) is significantly higher than anyone reported by other countries in any stations. The station reporting 128 natural exceedances of the PM_{10} limit value in 2009 is located on the isle of La Réunion.

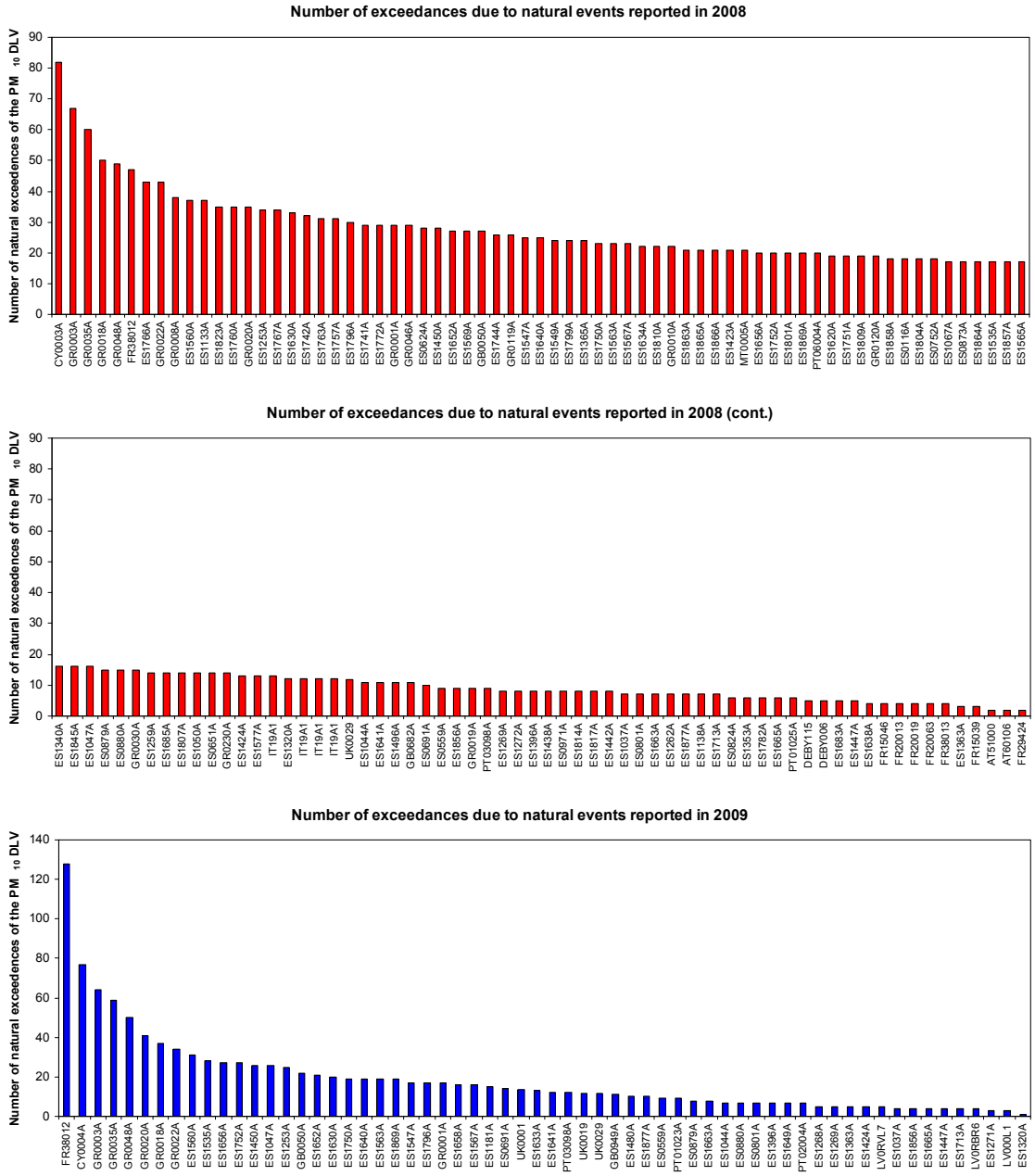

Figure 4.3. Stations and number of PM_{10} exceedances of the daily limit value due to natural events reported in 2008 and 2009. No data reported for IT in 2009.

In non-Mediterranean regions, the number of exceedances of the daily limit values due to natural causes per station was lower than in the Mediterranean Member States..

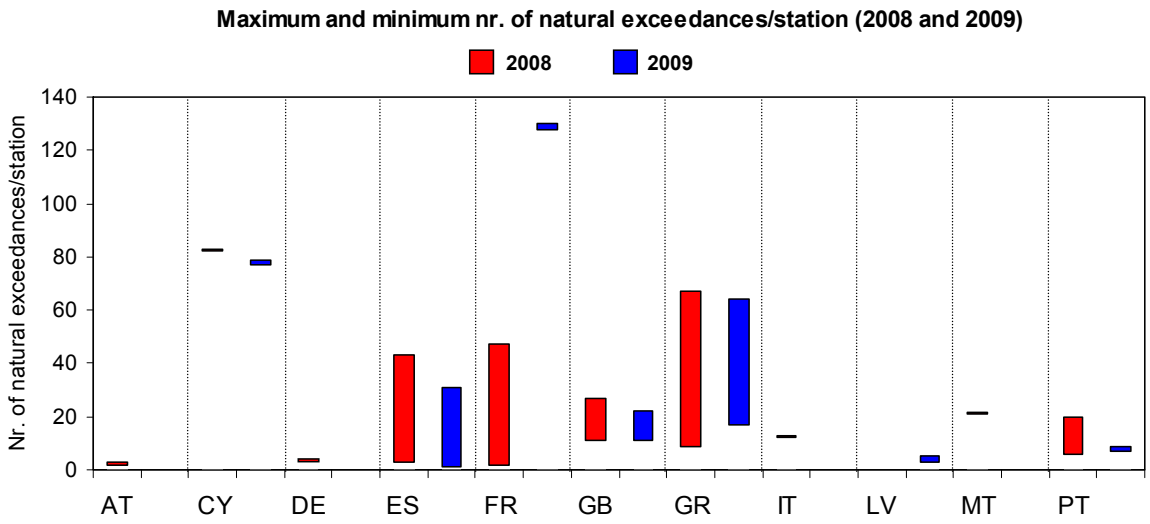

Figure 4.4. Range of PM_{10} exceedances of the daily limit value due to natural causes reported per Member State in 2008 and 2009.

The proportion of stations reporting different numbers of PM_{10} exceedances due to natural sources was also analysed (Figure 4.5), with the aim to detect in how far Member States report different degrees of influence of natural events on PM_{10} concentrations. To this end, the number of exceedances reported per station and year were classified in 5 ranges: <20 ; 20-40; 40-60; 60-80; and >80 exceedances/station.

Table 4.3. Range of exceedances of the PM_{10} daily limit value due to natural sources as reported per Member State in 2008 and 2009.

Nr. of natural exceedances of the daily limit value per MS				
	2008		**2009**	
	Min.	Max.	Min.	Max.
AT	2	2		
CY	82	82	77	77
DE	5	5		
ES	3	43	1	31
FR	2	47	128	128
GB	11	27	11	22
GR	9	67	17	64
IT	12	13	0	0
LV			3	5
MT	21	21		
PT	6	20	7	12

On average and based on this classification, 4 different patterns were observed:

(a) Member States in which the majority of stations reported between 0 and 60 exceedances/station of the PM_{10} daily limit value due to natural sources, and with a decreasing number of stations reporting from 0-20 to 40-60 exceedances/year. One example of this pattern, which was the most frequent among the Member States, was represented by Spain (Figure 4.5).

(b) Member States reporting >60 exceedances/station, consecutively for both years 2008 and 2009. Cyprus was the only Member State showing this pattern, although it must be noted that only one station reported exceedances of the PM_{10} daily limit value.

(c) Member States with larger numbers of stations reporting between 20-60 exceedances/station than 0-40 exceedances/station. Greece was a clear example of this pattern, with slightly different patterns in 2008 compared to 2009 but always with a prevalence of 20-60 exceedances/station.
(d) Member States for which discrepancies seem to occur between 2008 and 2009. This is the example of France, which showed different patterns in 2008 and 2009, and which included the only outlier described above regarding the number of exceedances (128, maximum number of exceedances due to natural sources reported in both years of study).

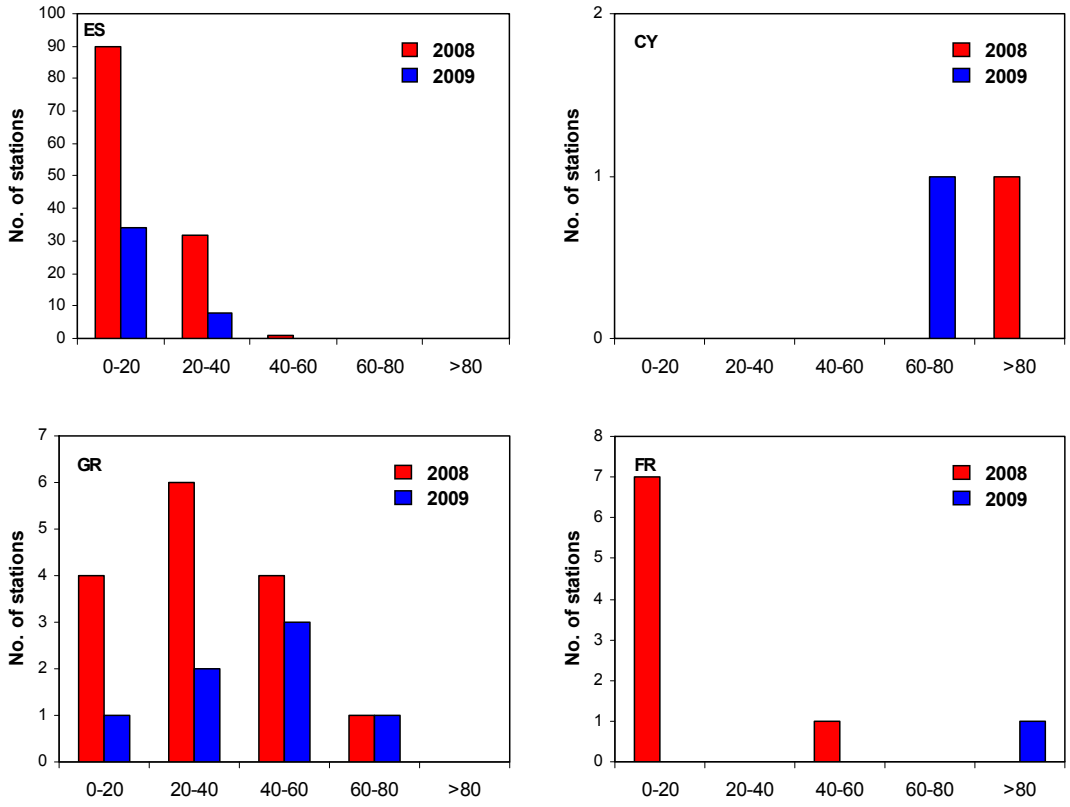

Figure 4.5. Number of exceedances of the PM_{10} daily limit value per station and Member State, classified in 5 ranges: <20 exceedances/station, 20-40 exceedances/station, 40-60 exceedances/station, 60-80 exceedances/station, and >80 exceedances/station.

This analysis evidences that, for the majority of Member States reporting exceedances due to natural events in 2008 and 2009, it was relatively unusual to detect >40 exceedances per station and year. Thus, based on the data from the years 2008 and 2009, the proportion within a given Member State of stations reporting >40 exceedances per station and year (over the total number of stations reporting exceedances in that Member State) was considered to be a good indicator of the correct or incorrect classification of exceedances as having a natural origin.. It is of course essential to consider that the total number of exceedances (both due to natural and anthropogenic sources) is subject to large variations as a function of changing meteorological and pollution situations during a year. Furthermore, the distance from the Member States to the African deserts is also a factor to be taken into consideration (probably with the exception of PT, as described above). In addition, given Member States may be affected by specific natural sources of exceptional occurrence (e.g., wildfires in Greece in 2007 and 2008, see Table 4.7.a below, or in Moscow in 2010) which may have a large impact on the number of exceedances of the daily limit value.

Table 4.4. Proportion of stations reporting >40 exceedances of the PM_{10} daily limit value due to natural sources per station and year in 2008 and 2009, in the different Member States.

% of stations reporting >40 exceedances/year		
	2008	**2009**
AT	0	
CY	100	100
DE	0	
ES	1	0
FR	13	100
GB	0	0
GR	33	57
IT	0	
LV		0
MT	0	
PT	0	0

As shown in Table 4.4, the proportion of stations reporting >40 natural PM_{10} exceedances/year in the different Member States was generally lower than 15% in 2008 and 2009. The only exception was detected for GR, with 33% in 2008 and 57% in 2009. As stated above, this was to the impact of wildfires registered in summer 2008 in the surroundings of Athens (see Table 4.7 and Figure 4.6 below). The cases of CY and FR (in 2009) are not comparable, as only data from 1 station were reported.

4.3. PM_{10} daily limit value: natural sources reported for justification of exceedances

The Commission Decision 2004/461/EC, of 29 April 2004, laying down a questionnaire to be used for annual reporting on ambient air quality assessment under Council Directives 96/62/EC and 1999/30/EC and under Directives 2000/69/EC and 2002/3/EC of the European Parliament and of the Council, established a list of reasons that can be used to explain individual exceedances of the daily and annual limit values (Table 4.5). These reasons include a general term for "natural source(s) or natural event(s)", coded as "S8". More specifically, the possible natural events causing limit value exceedances for PM_{10} are described in Table 1.1.

Table 4.5. Reasons for individual exceedences: standard codes (Table 2, Commission Decision 2004/461/EC).

Reason code	*Description*
S1	*Heavily trafficked urban centre*
S2	*Proximity to a major road*
S3	*Local industry including power production*
S4	*Quarrying or mining activities*
S5	*Domestic heating*
S6	*Accidental emission from industrial source*
S7	*Accidental emission from non-industrial source*
S8	*Natural source(s) or natural event(s)*
S9	*Winter sanding of roads*
S10	*Transport of air pollution originating from sources outside the Member State*
S11	*Local petrol station*
S12	*Parking facility*
S13	*Benzene storage*

It is interesting to note that contributions from sea-salt aerosols are not included in Table 4.5 nor in Table 1.1 (Tables 2 and 5 in Commission Decision 2004/461/EC). However, sea spray is included as a cause eligible for subtraction in Staff Working paper 6771/11. The absence of a standard code to report sea spray as a natural cause results in the fact that certain Member States (e.g., France) reported PM_{10} exceedances in 2008 and 2009 due to the impact of sea-salt aerosols using references which are not coded in Table 1.1 (e.g., “embruns marins”, as shown in Tables 4.6 and 4.7). **Thus, it is suggested that this source should explicitly be included in future evaluations and that a respective standard code should be created. This would eliminate the discrepancy existing currently between the wording given in Commission Decision 2004/461/EC and Staff Working Paper 6771/11. Member States have the option to add new codes to describe the reasons for individual exceedances, in Form 12 of the questionnaire. However, this is not possible for the natural event code (Table 5 in the questionnaire). Thus, it is strongly recommended to bring the codes for reasons for exceedance (Table 4.5) and the list of natural event codes (Table 1.1, Table 5 in the questionnaire) in line with the Staff Working Paper 6771/11.**

From a formal perspective, the analysis of the types of natural events reported by Member States in 2008 and 2009 evidenced the following issues:

- Member States did not always use the codes provided by Commission Decision 2004/461/EC: France reported exceedances due to “embruns marins”, Germany by “Saharastaub”, Malta provided the codes “S8a” and “S8b”, and Greece provided codes “H1” and “H43”. None of these terms are specified in Commission Decision 2004/461/EC and none of these codes can be found in form 12.

- Member States reported natural events using both the general code S8 (Table 4.5) and specific codes G2, H1, D1, D2 (Table 1.1): while certain Member States

specified the causes of the natural events, others used the more general term "Natural source(s) or natural event(s)".

- Certain Member States reported both natural and non-natural causes: this was the case for Italy in 2009, which provided the codes "S1; S5; S3; S8" for all sites.

- Member States applied the same codes for all stations: with the exception of France (2008) and Spain (2009), all Member States reported the same codes for all their stations (Figure 4.6). For example, in 2009, all stations on Cyprus were affected by G2, all Spanish stations by G2 (except for one station reporting S8), all French stations by "embruns marins", all British stations by S8, all Greek stations by H1;G2;D1, all Latvian stations by S8, all Polish stations by G2, and all Italian stations by S1; S5; S3; S8. The same was observed for all the days reported for each station, as will be discussed below. The fact that all stations within a given Member State are affected exactly by the same natural sources and during all days reported seems, at least, unlikely.

The causes reported by the different Member States are summarised in Tables 4.6a and b, and Figures 4.7a and b. The dominant natural cause contributing to exceedances of the PM_{10} limit values was transport of natural particles from dry regions outside the Member State (G2). This cause was given by 70% of the Member States reporting natural events in 2008 (CY, DE, ES, FR, GR, IT, PT) and 50% of the Member States in 2009 (CY, ES, GR and PT). Exceedances due to the more general term "natural source(s) or natural event(s)" (S8) were reported by 30% and 50% of the Member States in 2008 (AT, GB, MT) and 2009 (ES, GB, IT, LV), respectively. The analysis of the methodologies submitted for reporting of natural events (section 4.6 below) suggests that this code referred to African dust transport in all cases, and in addition to sea salt aerosols in Great Britain. Finally, other causes were reported by single Member States: wild-land fire inside and outside the Member State (D1 and D2), an unspecified cause (H1) by Great Britain in 2008 and 2009, and "embruns marins" by France in both years.

When comparing the natural causes given by the Member States in the two years analysed, differences were observed which were linked to the expected variability of natural events but also to the reporting schemes. The main difference was observed regarding cause G2 (transport of natural particles from dry regions outside the Member State), which was listed by 7 and 4 Member States in 2008 and 2009, respectively (Figure 4.6 and Tables 4.6a and b). This source refers mainly to transport of African dust towards Europe, and it is therefore probable, due to the nature of these outbreaks, that Member States such as Germany or France registered the influence of those events in 2008 but did not in 2009. However, it seems unlikely that Mediterranean Member States such as Italy or Malta did not register African dust outbreaks in 2009 (see Figure 4.7b), given that other Member States in the region (Spain, Greece, Cyprus) did report exceedances due to African dust. Conversely, in the case of Italy no exceedances due to the more general cause S8 (natural source(s) or natural event(s)) were reported in 2008, whereas this code was used for reporting in 2009. Therefore, it seems probable that the absence of G2 codes in Italy in 2009 results from a discrepancy in the reporting scheme between 2008 and 2009, whereby African dust events (possibly among other natural sources) were reported as G2 in 2008 and as S8 in 2009.

One other discrepancy was observed for Greece, given that codes G2 and D2 (mineral dust and wild-land fires) were reported in 2008 and 2009, whereas an additional cause (H1, not codified in Commission Decision 2004/461/EC) was provided in 2009 for all stations.

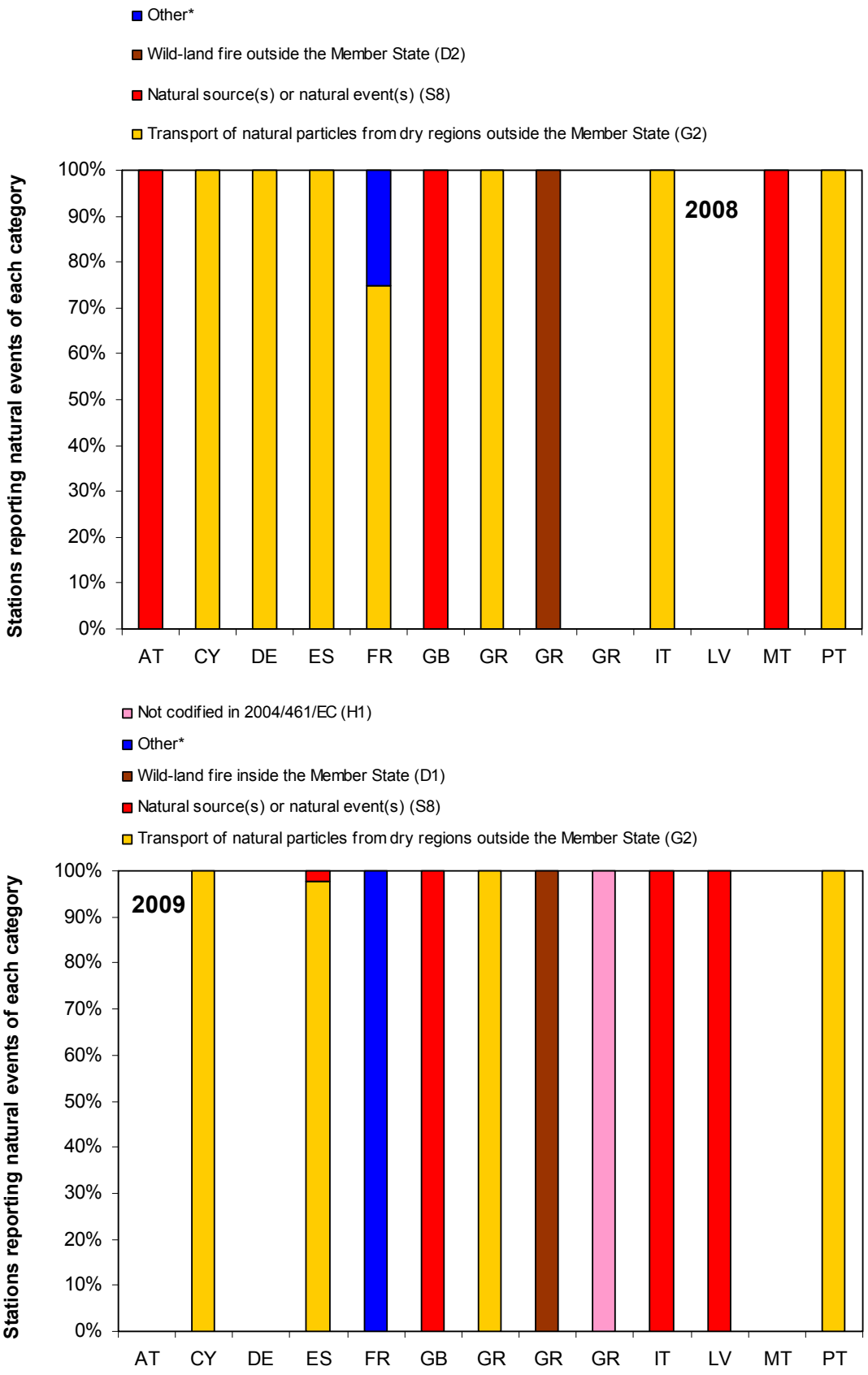

Figure 4.6. Proportion of stations reporting different types of natural causes for exceedances of the PM_{10} limit values in the Member States, in 2008 and 2009. Greece is listed 3 times given that >1 natural causes were reported for all stations. Other*: “embruns marins” (sea salt).

Table 4.6a. Natural causes reported for exceedances of the PM_{10} limit values by Member States in 2008. Nr. Stat.: number of stations reporting exceedances due to natural events.

Natural causes reported by Member States in 2008					
MS	**Nr. Stat.**	**G2**	**S8**	**D2**	**Other**
AT	2	0	2	0	0
CY	1	1	0	0	0
DE	2	2	0	0	0
ES	123	123	0	0	0
FR	8	6	0	0	2
GB	3	0	3	0	0
GR	15	15	0	15	0
IT	4	4	0	0	0
MT	1	0	1	0	0
PT	3	3	0	0	0

G2: Transport of natural particles from dry regions outside the Member State
S8: Natural source(s) or natural event(s)
D2: Wild-land fire outside the Member State
Other: “embruns marins”

Table 4.6b. Natural causes reported for exceedances of the PM_{10} limit values by Member States in 2009. Nr. Stat.: number of stations reporting exceedances due to natural events.

Natural causes reported by Member States in 2009						
MS	**Nr. Stat.**	**G2**	**S8**	**H1**	**D1**	**Other**
CY	1	1	0	0	0	0
ES	42	41	1	0	0	0
FR	1	0	0	0	0	1
GB	5	0	5	0	0	0
GR	7	7	0	7	7	0
IT	59	0	59	0	0	0
LV	3	0	3	0	0	0
PT	3	3	0	0	0	0

G2: Transport of natural particles from dry regions outside the Member State
S8: Natural source(s) or natural event(s)
H1: Not codified in Commission Decision 2004/461/EC
D1: Wild-land fire inside the Member State
Other: “embruns marins”

Regarding the remaining natural causes, the following were consistently reported by Member States in both years: sea-salt aerosols (“embruns marins”) by France, natural events (S8) by GB, and wild-land fires (with contributions from inside and outside the Member State, D1 and D2) by Greece. Because of the common nature of sea-salt contributions, it is somewhat surprising that this type of event was only reported by two Member States (FR and GB, see section 4.6). **This can possibly be explained by the fact that this source of PM is not included in Table 1.1, and therefore it was not listed by other Member States which may also be affected by it.**

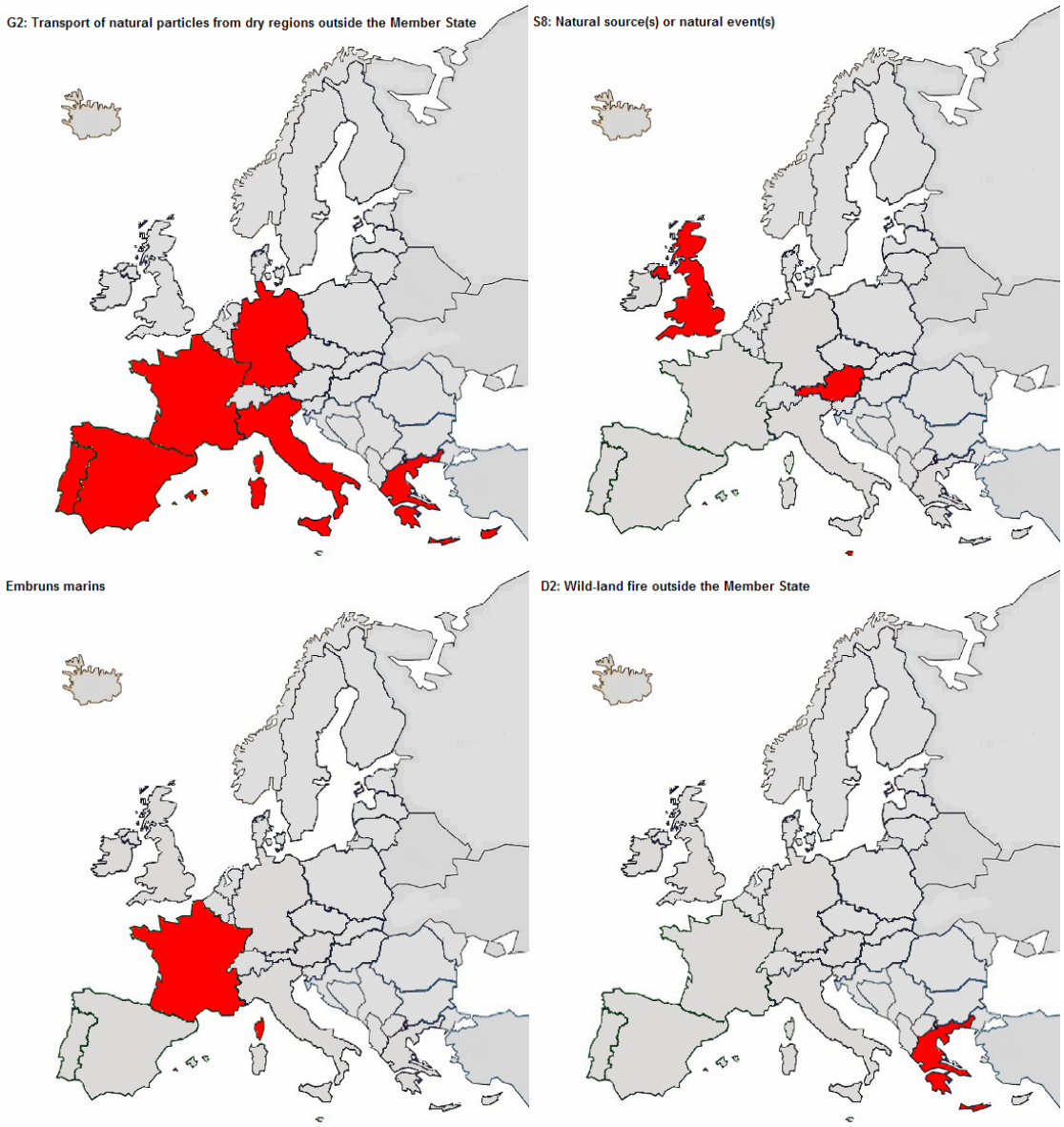

Figure 4.7a. Member States reporting exceedances of the PM_{10} limit value due to natural causes in 2008: Transport of natural particles from dry regions outside the Member State (G2); Natural source(s) or natural event(s) (S8); Wild-land fire outside the Member State (D2); Other ("embruns marins").

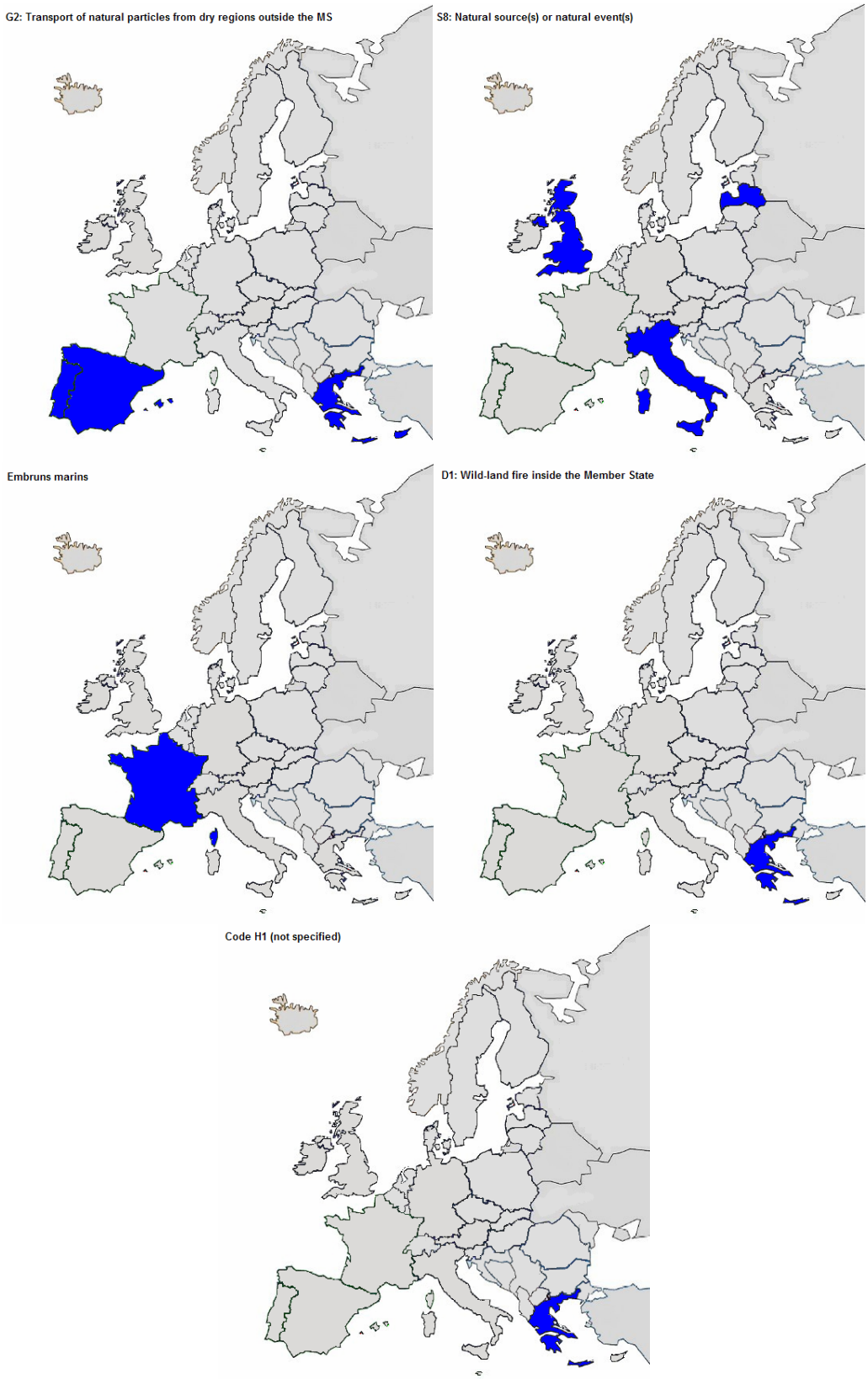

Figure 4.7b. Member States reporting exceedances of the PM_{10} limit value due to natural causes in 2009: Transport of natural particles from dry regions outside the Member State (G2); Natural source(s) or natural event(s) (S8); Wild-land fire inside the Member State (D1); Other ("embruns marins"); Code not specified in Commission Decision 2004/461/EC (H1).

4.4. PM_{10} annual limit value: assessment of the exceedances due to natural sources

Member States also reported natural contributions to annual PM_{10} levels, even though the number of States was lower than in the case of the daily limit value, as described above. In addition, the stations reporting natural contributions to the PM_{10} annual limit value exceedances in each Member State were different in 2008 than in 2009. Thus, the comparison between the two years is hampered. Table 4.7 summarises the number of Member States, the natural causes reported and the quantification of natural contributions to annual PM_{10} concentrations in 2008 and 2009.

According to the results submitted in forms 23b, in 2008 mean annual contributions of natural sources to PM_{10} levels ranged from 1-3 µg/m^3 in Italy, France, Portugal and Greece, to 4-5 µg/m^3 in Spain and Great Britain, to 13 µg/m^3 on Cyprus. In 2009, mean contributions were lower in Spain (1 µg/m^3 *vs*. 4 µg/m^3 in 2008), higher in Greece (8 µg/m^3 *vs*. 3 µg/m^3 in 2008), and similar on Cyprus (13 µg/m^3 in both years). The case of France is not comparable, given that African dust was the natural cause reported in 2008 and sea spray in 2009.

Table 4.7. Member States reporting natural contributions to PM_{10} annual concentrations in 2008 and 2009.

2008					
MS	**Nr. Stations**	**Natural event code(s)**	**Annual mean natural contribution (µg/m^3)**	**Max. annual mean natural contribution (µg/m^3)**	**Min. annual mean natural contribution (µg/m^3)**
CY	1	G2	13	13	13
ES	41	G2	4	10	2
FR	3	G2, embruns marins, S8	1	1	0.5
GB	1	S8	5	5	5
GR	12	G2;D2	3	7	1
IT	1	G2	1	1	1
PT	1	G2	2	2	2

2009					
MS	**Nr. Stations**	**Natural event code(s)**	**Annual mean natural contribution (µg/m^3)**	**Max. annual mean natural contribution (µg/m^3)**	**Min. annual mean natural contribution (µg/m^3)**
CY	1	G2	13	13	13
ES	12	G2	1	4	0.5
FR	1	Embruns marins	17	17	17
GR	6	H1;G2;D1	8	9	6

With the exception of Great Britain, Greece and France (in 2009), the natural contributions reported were linked to the transport of African dust to the Member States. Based on this assumption, the mean and maximum annual natural contributions estimated by the Member States (Figure 4.8) were compared with the results published by Querol et al. (2009) in a study in which mean contributions of African dust were estimated across the Mediterranean basin (Figure 4.9). This comparison shows that all Member States report concentrations which fall within the ranges expected (CY: 11 µg/m^3; ES: 1-4 µg/m^3; FR: 2-3 µg/m^3; GR: 6-7 µg/m^3; IT: 2-3 µg/m^3; PT: 1 µg/m^3), and therefore validate the estimations provided by the Member States. There is only one exception for Spain, where the range is higher than the one published by Querol et al (2009). This is due to the fact that the results in Table 4.7 include the Canary Islands, with very high African dust contributions (Viana et al., 2002;

Rodríguez et al. 2011), whereas the ranges described by Querol et al. (2009) refer only to the Mediterranean basin.

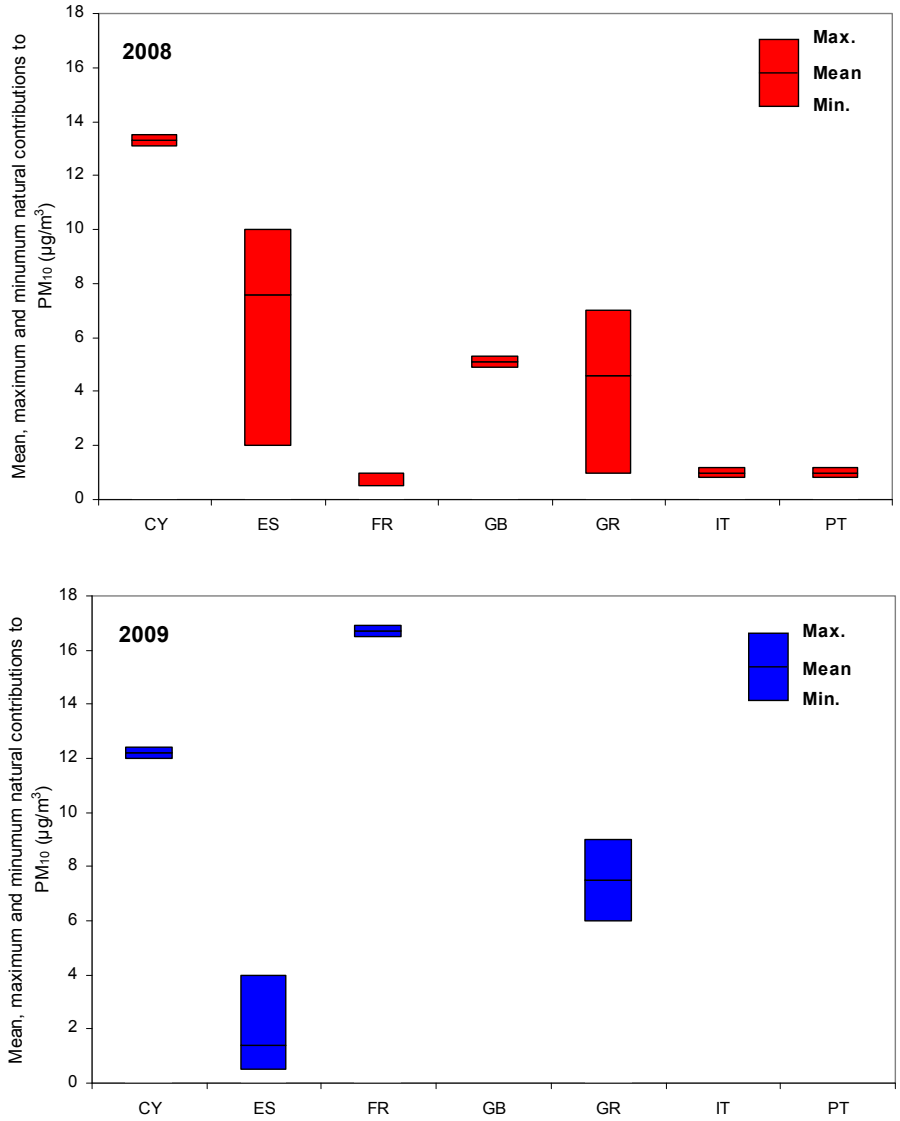

Figure 4.8. Mean, maximum and minimum natural contributions to annual PM_{10} levels (µg/m^3) in 2008 and 2009.

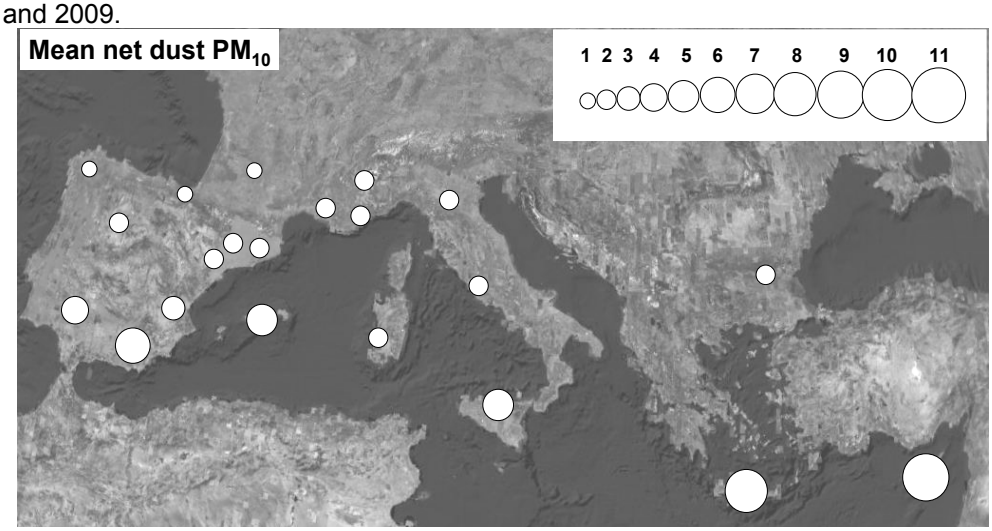

Figure 4.9: Mean annual African dust contribution to PM_{10} levels in the Mediterranean basin (Querol et al., 2009).

4.5. Effect of the correction by natural sources on the compliance with PM_{10} daily/annual limit values

4.5.1. Exceedances of the PM_{10} daily limit value

Table 4.8 and Figure 4.10 summarise the number of stations reporting exceedances of the PM_{10} daily limit value in 2008 and 2009, before and after subtraction of the exceedances due to natural sources. According to the data reported by the Member States, the correction by natural sources reduced the number of stations surpassing the 35 exceedances/year threshold in all Member States where this correction was applied, with the only exception of Latvia. In this Member State, the correction reduced the number of exceedances at each station, but none of them went below the 35 exceedances/year threshold. In the remaining Member States, the number of stations not surpassing the 35 exceedances/year threshold because of the correction ranged from 1 in Austria, Cyprus, Germany, France, Great Britain (only in 2008), Italy (only in 2008) and Malta, to 2-5 in Great Britain, Greece, and Portugal, and 18-51 in Spain. Proportionally, the percentage of stations not surpassing the 35 exceedances/year threshold after correction was maximal on Cyprus and Malta (100%), as opposed to 41-43% in Spain, 33-80% in Great Britain and Portugal, 27-71% in Greece, and 25% in Italy. In the case of Italy, correction due to natural events was not requested (given that form 23 was not submitted). The proportion of stations not surpassing the 35 exceedances/year threshold due to the correction was relatively stable in both years on Cyprus (100%) and in Spain (41-43%). In the remaining Member States, the effect of the correction was more variable.

Table 4.8. Number of stations reporting exceedances of the PM_{10} daily limit value (DLV) before and after subtraction of natural contributions. No data were available in Form 23 for IT in 2009. ND: no data available or reported.

Number of stations reporting exceedances of the PM_{10} DLV				
	2008		**2009**	
	Before correction	**After correction**	**Before correction**	**After correction**
AT	2	1	ND	ND
CY	1	0	1	0
DE	2	1	ND	ND
ES	123	72	42	24
FR	8	7	1	0
GB	3	2	5	1
GR	15	11	7	2
IT	4	3	59	ND
LV	ND	ND	3	3
MT	1	0	ND	ND
PT	3	0	3	2

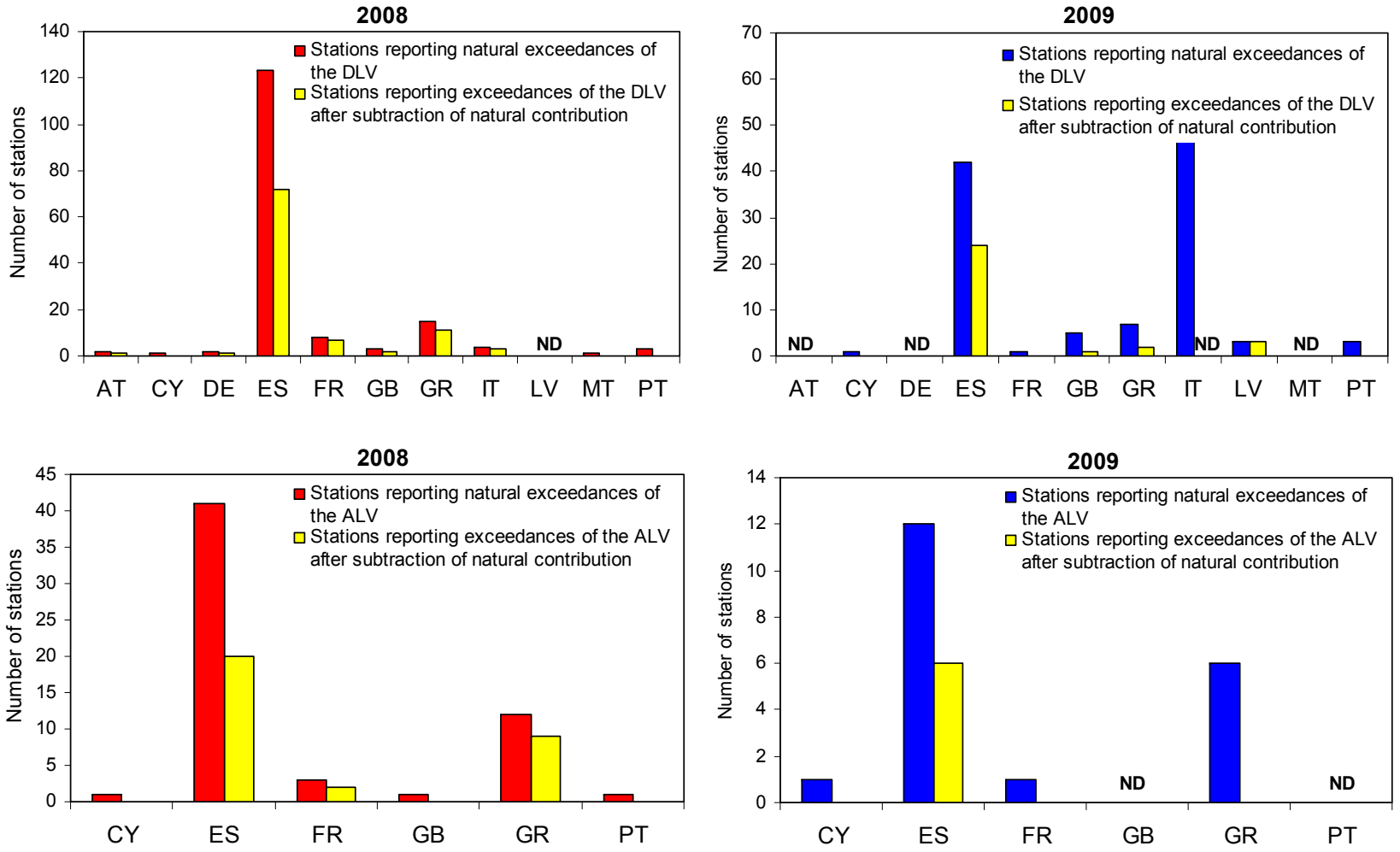

Figure 4.10. Number of stations exceeding the PM_{10} daily limit value (DLV, top) and annual limit value (ALV, bottom) in 2008 and 2009 before and after subtraction of natural contributions. ND: no data reported.

4.5.2. Exceedances of the PM_{10} annual limit value

Table 4.9 and Figure 4.10 summarise the number of stations reporting the exceedance of the PM_{10} annual limit value in 2008 and 2009, before and after subtraction of the mean annual natural contribution to PM_{10} levels. As described above, the number of Member States reporting annual contributions to PM_{10} levels of natural sources is much lower than in the case of the daily contributions. The subtraction of the mean annual natural contribution resulted in a reduction of the number of stations exceeding the PM_{10} annual limit value of 40 µg/m^3 in all Member States. The decrease in the number of stations ranged from 1 station on Cyprus, in France, Great Britain and Portugal, to 3-6 in Greece and 6-21 in Spain. In relative terms, this would mean a 100% reduction in the case of Cyprus, Great Britain, Portugal and France and Greece in 2009, and down to 50% in Spain, 33% in France (2008) and 25% in Greece (2009). After the correction, the following Member States still exceeded the PM_{10} annual limit value: Spain, France and Greece in 2008, and Spain in 2009.

Table 4.9. Number of stations reporting exceedances of the PM_{10} annual limit value (ALV) before and after subtraction of natural contributions.

Number of stations reporting exceedances of the PM_{10} ALV				
	2008		**2009**	
	Before correction	**After correction**	**Before correction**	**After correction**
CY	1	0	1	0
ES	41	20	12	6
FR	3	2	1	0
GB	1	0	ND	ND
GR	12	9	6	0
PT	1	0	ND	ND

4.6. Assessment of methodologies applied in 2008-2009

Different methodologies were applied by the different Member States to justify the natural exceedances of the daily and annual PM_{10} limit values reported in forms 23a and 23b. Table 4.10 transcribes the text submitted by the Member States for this justification in 2008 and 2009. From a formal point of view, the following issues may be highlighted:

- Methodologies were described in the national languages by some Member States and in English by others. The language heterogeneity poses an initial limitation to the assessment of the methodologies used. **So, it could be recommended to submit the title of justification document and a brief summary of the methodology in English, in addition to the national language.**
- Not all Member States provided a description of the methodology used, or they provided codes which were not found in Commission Decision 2004/461/EC (see examples of IT, CY in 2008 and LV in 2009).
- The majority of Member States referred to one document for justification, with the exception of Great Britain in 2008 and 2009 (referring to 1 additional document for Gibraltar) and Germany in 2008.
- All justification documents were publicly available, but while some were easily found (for example, in the cases of GB and DE), others were difficult to trace.

- Certain Member States used methodologies for justification of natural events which are not included in Staff Working Paper 6771/11. However, it is essential to consider that the justifications under assessment in the present document were submitted in 2008 and 2009, whereas Staff Working Paper 6771/11 was published in 2011.
- For Member States reporting exceedances due to the general code S8 (“natural events”), the assessment of the methodology reported for justification allowed for the identification of the type of natural source (for example, sea salt aerosols in GB and African dust in MT). However, this was not the case for other Member States (see LV), for which the natural causes reported were unspecified.

The methodologies reported by each of the Member States in 2008 and 2009 are described and assessed in Annex I:

Table 4.10. Transcription of the text submitted by Member States for the justification of exceedances of the PM_{10} annual and daily limit values due to natural sources. Years 2008 and 2009.

2008	Reference to justification
AT	Ferntransport von Saharastaub 2008.doc; Bruckmann_2008.pdf
CY	S14
DE	1. An outbreak of Saharan dust causing high PM10 levels north of the Alps, veröffentlicht in Gefahrstoffe- Reinhaltung der Luft - Ausgabe 11/12-2008
	2. Mitarbeitermagazin des DWD November/Dezember 2008, Seite 14
ES	ESTUDIO Y EVALUACION DE LA CONTAMINACIÓN ATMOSFÉRICA POR MATERIAL PARTICULADO EN ESPAÑA. Informe episodios naturales 2008
FR	analyse_PM2008_RA
GB	1. Reference A, listed in Form 20, method is also described in UK PM10 time extension application http://ec.europa.eu/environment/air/quality/legislation/time_extensions.htm
	2. African dust events - Report published on Gibraltar air quality website: http://www.gibraltarairquality.gi/documents/Gib_natural_quantification_2008_v1.pdf
GR	μοντέλο μεταφοράς σκόνης SKIRON, δορυφορικές εικόνες TOMS και SEAWIFS
IT	
MT	Justification report on natural PM10
PT	Annexed report

2009	Reference to justification
CY	Backtrajectories analysis for 2009 exceedance days has been uploaded together with this questionnaire.
ES	ESTUDIO Y EVALUACION DE LA CONTAMINACIÓN ATMOSFÉRICA POR MATERIAL PARTICULADO EN ESPAÑA. Informe episodios naturales 2009
FR	Corrélation avec la houle
GB	1. Reference A, listed in Form 20, method is also described in UK PM10 time extension application http://ec.europa.eu/environment/air/quality/legislation/time_extensions.htm
	2. African dust events - Report published on Gibraltar air quality website: http://www.gibraltarairquality.gi/documents/Gib_natural_quantification_2009_v2.pdf
GR	μοντέλο μεταφοράς σκόνης SKIRON, δορυφορικές εικόνες TOMS και SEAWIFS, θαλάσσιο αλάτι
IT	
LV	5%
MT	Justification report on the contribution of natural events to the PM10 daily limit value
PT	Annexed report

4.7. Assessment of the reporting strategies of the Member States for specific days exceeding the PM_{10} daily limit value

4.7.1. Comparison between data reported by Member States in form 23 and form 11

Forms 11 and 23 in the reporting questionnaire compile data regarding the exceedances of the PM_{10} limit values. Whereas forms 11 list specific data regarding exceedances of the PM_{10} daily (stations, date, daily level, reason) and annual (station, annual level, reason) limit values, form 23 compiles information about exceedances due to natural causes. Four main issues were detected when comparing the data reported by Member States in both forms:

1) Certain Member States submitted data in form 11 indicating natural events as reason for the exceedances but did not submit the corresponding information in form 23. This is correct when the 35 exceedances/year threshold for the PM_{10} daily limit value is not surpassed (e.g., Iceland in 2009), but not otherwise (e.g., Italy in 2009). **The absence of data in form 23 is unusual and may indicate an inconsistency in the reporting strategy of the respective Member State**.

2) For 2009, the number of stations reporting exceedances of the PM_{10} daily limit value due to natural events (S8), according to form 11 (Table 4.11, in brackets), was compared with the number of stations surpassing the 35 exceedances/year threshold due to natural causes (Table 4.1). On an initial evaluation, inconsistencies were detected between both Tables. The number of stations in Table 4.11 should be equal or larger than in Table 4.1, given that in Table 4.1 only stations surpassing 35 annual exceedances are included, whereas Table 4.11 summarises the total number of stations reporting natural causes (with and without surpassing the 35 exceedances/year threshold). However, in the cases of Spain and Great Britain, the numbers of stations in Table 4.1 (form 23a, 42 and 5, respectively) are larger than in Table 4.11 (form 11h, 34 and 1, respectively). It must be noted that form 23 is the form with legal validity for reporting exceedances due to natural events. It is recommended that **this inconsistency will be addressed by respective Member States for future reporting.**

3) Other inconsistencies regarding different standard codes in forms 11 and 23 or wrong station codes should be also addressed in the reporting strategies by MS.

4.7.2. Assessment of the stations and specific days reported as natural exceedances of the PM_{10} daily limit value

In this section the monthly and annual number of exceedances of the PM_{10} daily limit value reported by each Member State in form 11h was analysed (Table 4.11), for exceedances caused by natural events and coded as S8. Codes G2 (corresponding to "Transport of natural particles from dry regions outside the Member State") are not included due to the fact that codes described in Table 4.5 were only used for reporting in form 11h, whereas codes described in Table 1.1 were used in form 23. Therefore, for the assessment of the specific exceedance days it is not possible to identify the specific natural cause behind the exceedance. **It is interesting to note that, as described in the previous sections both codes from Tables 1.1 and 4.5 were used for reporting in 2008 and 2009 in forms 23a and 23b. For future reporting, it is recommended to follow requirements in the Commission Decision and use table 4.5 and table 1.1 for reporting in forms 11h and 23a and b, respectively.**

The aim of this analysis is to assess the specific days which were reported as impacted by natural sources and to evaluate the geographical extent of this impact. The present analysis focuses mostly on African dust due to the fact that it is the only natural source reported for 2008 and 2009 with an impact on PM levels on a large geographical scale. This not the case for sea salt contributions (which may or may not be comparable in neighbouring countries, e.g. PT and ES) or forest fires (only reported by GR for 2008 and 2009). Thus, the present analysis focuses on the exceedances reported for 2009 in form 11h.

Table 4.11. Monthly number of total S8 exceedances in 2009 reported by each country. Normalised annual mean number of exceedances per station and country. (In parenthesis the total number of stations reporting exceedances due to natural sources). IS: Iceland.

No. of exceedances (S8)	**CY (1)**	**ES (34)**	**FR (1)**	**GB (1)**	**GR (7)**	**IT (62)**	**IS (2)**	**LV (3)**	**PT (10)**
January	0	22	18	0	83	1130	0	0	0
February	15	57	13	0	53	700	1	0	22
March	15	132	15	3	45	484	1	0	54
April	8	41	9	0	42	95	0	2	1
May	5	135	7	1	32	70	6	3	21
June	1	188	11	3	21	5	0	1	21
July	3	150	9	7	37	24	0	2	0
August	2	131	8	3	16	17	1	0	24
September	4	42	8	2	28	147	0	2	28
October	16	77	16	3	34	436	0	2	16
November	18	199	12	5	84	752	1	0	4
December	25	19	1	0	59	779	3	0	0
Total	**112**	**1193**	**127**	**27**	**534**	**4639**	**13**	**12**	**191**
Average exc./station	**112**	**35**	**127**	**27**	**76**	**75**	**7**	**4**	**19**
% Average exc. /station	**31%**	**10%**	**35%**	**7%**	**21%**	**21%**	**2%**	**1%**	**5%**

Results evidence a high number of stations reporting natural exceedances in Italy (62) and Spain (34). In order to assess the number of natural exceedances excluding the influence of the number of monitoring stations per Member State, the mean number of exceedances per station was calculated. Based on these data, it is evident that, in 2009, natural events (mostly African dust episodes) caused and/or contributed to exceed the PM_{10} daily limit value on Cyprus on around 30% of the days; 10% in Spain, 35% in France, 7% in Great Britain; 21% in Greece and Italy; and 5% in Portugal. When comparing these results to the data on African dust events in the Mediterranean published by Querol et al., (2009), the number of exceedances attributed to natural events (S8) appears to be within the average in the case of all countries named above but Italy and France. The **results indicate that the number of exceedances of the daily limit value caused by natural sources as attributed by Italy was high with respect to the total number of natural events that occurred in 2009 (Matassoni et al., 2009)**. The case of FR (only one station reporting) is not comparable given that the reported reason for exceedences was sea salt contributions (reported in form 23 and not in form 11h).

According to the synthesis of results published by Querol et al. (2009), a first analysis of the data was carried out distinguishing Member States located in the Western Mediterranean and those in the Central-Eastern Mediterranean regions. As stated in Querol et al. (2009), in the Western Mediterranean African dust outbreaks occur mainly in summer, whereas in the Eastern Mediterranean the major occurrence is recorded in

winter/spring. In addition, years with high occurrence of dust events in the Western Mediterranean generally coincide with a low frequency of dust events in the Eastern Mediterranean, and vice versa. From these conclusions, the highest number of exceendeces of the PM_{10} daily limit value due to African dust was expected to follow this pattern in the Western and Eastern Mediterranean Member States. As shown in Figure 4.11 the exceedances reported by Spain, Great Britain and Portugal were highest in spring-summer, and lowest in wintertime (with a secondary maximum in November in Great Britain). In contrast, the Central and Eastern Mediterranean States (CY, GR, IT) reported most of the natural exceedances of the PM_{10} daily limit value between October and April, with few exceedance days during the summer months. **Thus, the seasonality of the natural exceedances of the PM_{10} daily limit value is coincident with the occurrence of African dust outbreaks, and thus confirms the correct justification of this type of exceedances by the Member States.** The seasonality observed for France refers to sea salt contributions, and therefore should not be compared to the one described for African dust outbreaks. The higher incidence of sea salt contributions in autumn and winter seems plausible for France.

The analysis of the natural and anthropogenic causes reported in form 11h for the exceedances of the PM_{10} daily limit value (Table 4.12) evidenced that most of the Member States reported several causes for each exceedance day. It is possible that certain natural events (S8) coincide in time with events causing high anthropogenic emissions (S_x in Table 4.5), so that the sum of contributions causes finally the exceedance. Only France, Island and Portugal reported one single cause (S8) for the majority of the natural exceedance days. On the contrary, Cyprus, Great Britain and Italy reported 3 or more simultaneous causes for each exceedance day. Spain and Greece reported most of the exceedence days as the sum of a natural event (S8) and the influence of traffic emissions. In the case of Spain, air quality network managers currently receive the net African dust load for each day affected by African dust air masses, and therefore have at their disposal all the tools necessary to properly address the justification of natural exceedances of the PM_{10} daily limit value. The case of Greece is similar, even though the net African dust loads are estimated by means of other tools (modelling). **Thus, reporting of exceedances of the PM_{10} daily limit value due to African dust in form 11h should be improved by certain Member States.**

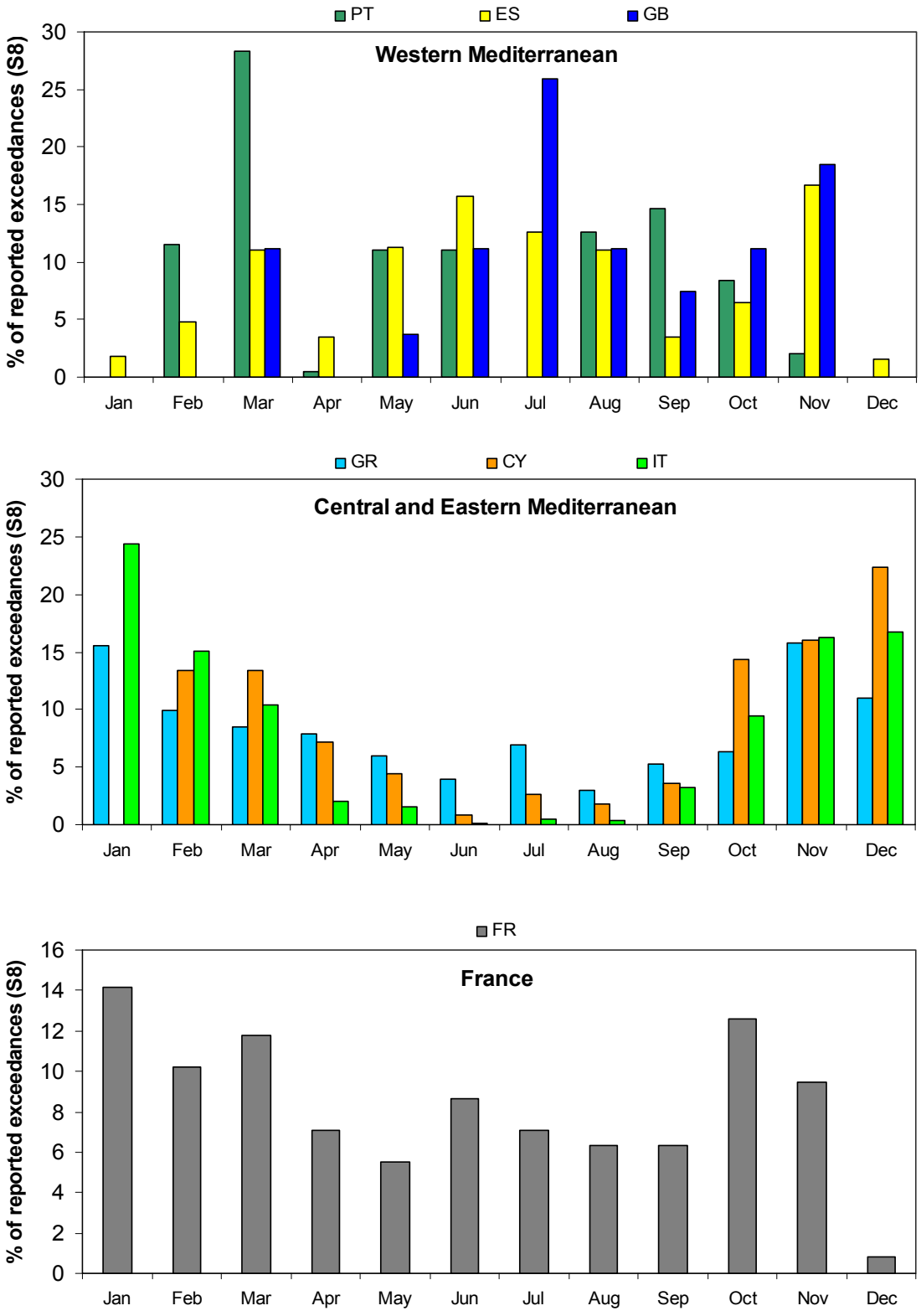

Figure 4.11. Monthly frequency of exceedances of the PM_{10} daily limit value due to natural sources (S8). Top: Western Mediterranean Member States (including Gibraltar, Great Britain); middle: Central and Eastern Mediterranean Member States; bottom: FR.

Table 4.12. Number of exceedances reported by each country and their justification by codified causes.

Causes	CY	ES	FR	GR	GB	IT	IS	LV	PT
S8	0	76	**127**	0	0	0	**11**	0	**165**
S1;S8	0	479	0	534	0	0	0	10	0
S2;S8	0	304	0	0	0	0	0	0	0
S3;S8	0	76	0	0	0	0	0	0	0
S4;S8	0	39	0	0	0	0	0	0	0
S5;S8	0	0	0	0	0	0	0	0	3
S8;S10	0	0	0	0	0	0	2	0	0
S1;S2;S8	0	43	0	0	0	0	0	0	10
S1;S5;S8	0	0	0	0	0	3	0	0	0
S1;S8;S9	0	0	0	0	0	0	0	2	0
S2;S3;S8	0	0	0	0	27	0	0	0	0
S4;S6;S8	0	44	0	0	0	0	0	0	0
≥4 causes	**112**	132	0	0	0	**4636**	0	0	13
Total	112	1193	127	534	27	4639	13	12	191

To summarise, the following recommendations may be provided regarding reporting of natural exceedances of the PM_{10} daily limit value in form 11 for the year 2009:

- The number of exceedances of the daily limit value due to natural causes reported seemed reasonable for Iceland, Portugal, Spain, Greece, Cyprus and Great Britain, whereas larger numbers of exceedances than expected were reported by France and Italy.
- For several Member States, it is advised to improve the identification of the specific causes. Reporting >1 natural cause per exceedance as a general practice should be avoided.

5. Summarising conclusions

Based on the questionnaires reported by Member States to the Commission for the years 2008 and 2009, the reporting on natural events by the Member States under Directive 2008/50/EC was analysed. The contribution of four main natural causes to daily and annual PM_{10} levels was evaluated:

- African dust
- Sea salt aerosols
- Volcanic dust
- Wild-land fires

The result of this evaluation is summarised in the following:

Member States reporting exceedances due to natural sources (form 23)
10 Member States (AT, CY, DE, ES, FR, GB, GR, IT, MT, PT) reported exceedances of the PM_{10} daily limit value due to natural sources in 2008, and 8 in 2009 (CY, ES, FR, GB, GR, IT, LV, PT). The number of Member States reporting exceedances of the annual limit value due to natural sources was always lower than for the daily limit value, with 7 Member States in 2008 (CY, ES, FR, GB, GR, IT, PT) and 4 in 2009 (CY, ES, FR, GR). This could be due to the low impact of the contributions from natural sources on annual mean levels, but also to the absence of specific tools to quantify these contributions on an annual scale.

Number of exceedances of PM_{10} daily limit value due to natural sources
The highest number of PM_{10} exceedances per station due to natural sources was reported by Mediterranean Member States (CY, ES, FR and GR, no data available for IT in 2009). With the exception of FR in 2009, this is linked to the fact that the cause for exceedances was mainly transport of natural particles from dry regions outside the Member State. The natural exceedances in FR in 2009 were justified as sea spray. Conversely, and for the same reason, the lowest numbers of natural exceedances per station were reported by AT, DE and LV. The number of natural exceedances reported per station and year is subject to a large variability. This is mainly due to year-to-year changes in meteorology controlling natural event as well as anthropogenic air pollutant emissions and their dispersion in Europe.

Natural sources reported for justification of exceedances of the PM_{10} daily limit value
The dominant natural cause of exceedances of the PM_{10} daily limit value reported by the different Member States was transport of natural particles from dry regions outside the Member State (G2), named by 70% of the Member States reporting natural events in 2008 (CY, DE, ES, FR, GR, IT, PT) and 50% of the Member States in 2009 (CY, ES, GR and PT). Exceedances due to the more general term “natural source(s) or natural event(s)” (S8) were reported by 30% and 50% of the Member States in 2008 (AT, GB, MT) and 2009 (ES, GB, IT, LV), respectively. The analysis of the methodologies submitted for reporting of natural events suggests that this code referred to African dust transport in all cases, and in addition to sea salt aerosols in GB. Finally, other causes were reported by single Member States: wild-land fire inside and outside the Member State (D1 and D2) and an unspecified cause (H1) by GR in 2008 and 2009, and “embruns marins” (sea salt) by FR in both years.

Because of the nature of sea-salt contributions, it is somewhat surprising that this type of event was only reported by two Member States (FR and GB). This may only be explained by the fact that this source of PM is not included in Table 5 of Staff Working Paper 6771/11, and therefore it was not listed by other Member States which might have been affected by sea salt events. It is therefore recommended to include sea salt aerosols in a revised version of the Staff Working Paper 6771/11,

Assessment of the exceedances of the PM_{10} annual limit value due to natural sources

Mean annual contributions of natural sources to PM_{10} levels ranged from 1-3 µg/m^3 in IT, FR, PT and GR, to 4-5 µg/m^3 in ES, GB and to 13 µg/m^3 in CY in 2008. In 2009, mean contributions were lower in ES (1 µg/m^3 versus 4 µg/m^3 in 2008), higher in GR (8 µg/m^3 versus 3 µg/m^3 in 2008), and similar in CY (13 µg/m^3 in both years). The case of FR is not comparable, given that African dust was the natural cause reported for 2008 and sea spray for 2009. With the exception of GB, GR and FR (in 2009), the reported natural contributions were linked to the transport of African dust to the Member States.

Effect of the correction by natural sources on the compliance with limit values

PM_{10} daily limit value

The correction by natural sources reduced the number of stations surpassing the 35 exceedances/year threshold in all Member States where this correction was applied, with the only exception of LV (in 2009). In the remaining Member States, the number of stations not surpassing the 35 exceedances/year threshold in 2008 and 2009 because of the correction ranged from 1 in AT, CY, DE, FR, GB (only in 2008), IT (only in 2008) and MT, to 3-5 in GB, GR, IT, LV and PT, and 18-51 in ES. Proportionally, the percentage of stations not surpassing the 35 exceedances/year threshold after correction for natural event contributions was highest in CY, MT and PT (100%), compared to 41-43% in ES, 33-80% in GB, 27-71% in GR, and 25% in IT. In the case of IT, correction due to natural events was not requested (due to the fact that form 23 was not submitted).

PM_{10} annual limit value

In all Member States with respective reporting, the subtraction of the mean annual natural contribution resulted in a reduction of the number of stations exceeding the PM_{10} annual limit value of 40 µg/m^3. The decrease in the number of stations ranged from 1 station in CY, FR, GB and PT, to 3-6 in GR and 6-21 in ES. In relative terms, this implies a 100% reduction in the case of CY, GB and PT, and down to 50% in ES, 33% in FR and 25% in GR. After the correction, the following Member States still exceeded the PM_{10} annual limit value: ES, FR and GR in 2008, and ES in 2009.

Assessment of methodologies applied in 2008-2009

Different methodologies were applied by Member States to justify natural contributions to PM levels:

- African dust: ES, GB and PT applied the methodology recommended by Staff Working Paper 6771/11, and MT applied a prior version of this method, which is mostly comparable to it. AT, CY, DE and GR applied qualitative methodologies with different degrees of comparability with respect to the method recommended by the Staff Working Paper 6771/11.
- Sea salt aerosols: no Member States applied the methodology recommended by Staff Working Paper 6771/11. GB applied a modelling approach, whereas no specific details were provided for justification by FR.
- Wild-land fires: this natural source was reported by GR, but satellite images were the only tools listed for justification.
- No methodologies were submitted by IT or LV for justification of African dust contributions or the unspecified cause S8 (natural events), respectively.

Assessment of stations and specific days reported as natural exceedances (form 11)

In 2009, natural events (mostly African dust episodes) caused and/or contributed to exceed the PM_{10} daily limit value in CY on around 30% of the days; 10% in ES, 35% in FR, 7% in GB; 21% in GR and IT; and 5% in PT. When comparing these results to the data on African dust events in the Mediterranean published by Querol et al., (2009), the

number of exceedances attributed to natural events appears to be within the average in the cases of CY, ES, GB, GR and PT. In addition, the seasonality of the natural exceedances of the PM_{10} daily limit value is coincident with the occurrence of African dust outbreaks. This thus confirms that the justification for this type of contribution to exceedances given by Member States is reasonable. The number of natural exceedances of the daily limit value attributed by IT was high with respect to the total number of natural events that occurred in 2009.

The following recommendations can be given regarding the reporting of natural exceedances of the PM_{10} daily limit value in form 11 for the year 2009:

- The number of exceedances of the daily limit value due to natural causes reported seemed reasonable for Iceland, Portugal, Spain, Greece, Cyprus and Great Britain, whereas larger numbers of exceedances than expected were reported by France and Italy.
- For several Member States, it is advised to improve the identification of the specific causes. Reporting >1 natural cause per exceedance as a general practice should be avoided..

From a formal perspective, the following issues were detected when analysing the replies to the questionnaire by Member States in 2008 and 2009:

Sea salt aerosols:
It is suggested that sea salt aerosols should be included in future revisions of the EC Staff Working Paper 6771/11 and that a standard code should be created for such an entry. This would eliminate the discrepancy that currently exists between the respective texts 2004/461/EC and 6771/11. In Form 12 of the questionnaire Member States have the option to add new codes to describe the reasons for individual exceedances. However, this is not possible for the Natural event code (Table 5 in Commission Decision 2004/461/EC). Thus, it is recommended to unify the codes for reasons for exceedance and the list of natural event codes.

Analysis of the types of natural events reported:
- Member States did not always use the codes provided by Commission Decision 2004/461/EC: FR reported exceedances due to “embruns marins”, DE by “Saharastaub”, MT provided codes “S8a” and “S8b”, and GR provided codes “H1” and “H43”. None of these terms are specified in Commission Decision 2004/461/EC.
- Member States reported natural events using both the general code S8 (Table 1.1) and specific codes G2, H1, D1, D2 (Table 4.5): while certain Member States specified the causes of the natural events, others use the more general term “Natural source(s) or natural event(s)”.
- Certain Member States reported both natural and non-natural causes: this was the case of IT in 2009, which provided the code “S1; S5; S3; S8” for all sites.
- Member States applied the same codes for all stations: with the exception of FR (2008) and ES (2009), all Member States reported the same codes for all their stations (Figure 4.6). For example, in 2009, all CY stations were affected by G2, all ES stations by G2 (except for one station reporting S8), all FR stations by “embruns marins”, all UK stations by S8, all GR stations by H1;G2;D1, all LV stations by S8, all PT stations by G2, and all IT stations by S1; S5; S3; S8. The same was observed for all the days reported by each station, as will be seen below. The fact that all stations within a given Member State are affected exactly by the same natural sources and during all days reported seems to be unlikely.

Assessment of methodologies applied in 2008-2009:

- Methodologies were described in the national languages by some Member States and in English by others. The language heterogeneity poses an initial limitation to the assessment of the methodologies used. This issue might be solved by submitting the title of justification document and a brief summary of the methodology in English, in addition to the national language.
- Not all Member States provided a description of the methodology used, or they provided codes which cannot be found in Commission Decision 2004/461/EC (see examples of IT, CY in 2008 and LV in 2009).
- The majority of Member States referred to one document for justification, with the exception of GB in 2008 and 2009 (referring to one additional document for Gibraltar) and DE in 2008.
- All justification documents were publicly available, but while some were easily found (for example, in the cases of GB and DE), others were difficult to trace.
- Certain Member States used methodologies for justification of natural events which are not included in Staff Working Paper 6771/11. However, it is essential to consider that the justifications under assessment in the present document were submitted in 2008 and 2009, whereas Staff Working Paper 6771/11 was published in 2011.
- For Member States reporting exceedances due to the general code S8 ("natural events"), the assessment of the methodology reported for justification allowed for the identification of the type of natural source (for example, sea salt aerosols in GB and African dust in MT). However, this was not the case for other Member States (see LV), for which the natural causes reported were unspecified.
- The methodologies for quantification of sea salt contributions presented in Staff Working Paper 6771/11 ate somewhat contradictory: whereas the possibility of calculating the sea salt contribution using only sodium or chloride as a tracer is included, the same document discourages the use of chloride as the only tracer given that it is potentially subject to both positive and negative artefacts. It would be advisable to define a clear recommendation regarding this issue.

Comparison between data reported in form 23 and form 11:

- Codes from Tables 2 and 5 from Commission Decision 2004/461/EC were used simultaneously for reporting for 2008 and 2009 in forms 23a and 23b. For future reporting, it is recommended that the Commission clarifies which types of codes (from Tables 2 or 5) should be used for reporting in forms 11h and 23a and b.
- Certain Member States submitted data in form 11 but did not submit the corresponding information in form 23. This is correct when the 35 exceedances/year threshold for the PM_{10} daily limit value is not surpassed (e.g., IS in 2009), but not otherwise (e.g., IT).
- The number of stations reporting exceedances of the PM_{10} daily limit value due to natural events (S8), according to form 11, was compared with the number of stations surpassing the 35 exceedances/year threshold due to natural causes (form 23). On an initial evaluation, inconsistencies were detected between both forms. The number of stations in form 11 should be equal or larger than in form 23, given that in form 23 only stations surpassing 35 annual exceedances are included, whereas form 11 summarises the total number of stations reporting natural causes (with and without surpassing the 35 exceedances/year threshold). However, this was not the case for all Member States.

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Abbreviations

Geographical name	Country code*
Kingdom of Belgium	BE
Bulgaria	BG
Czech Republic	CZ
Denmark	DK
Germany	DE
Estonia	EE
Ireland	IE
Iceland	IS
Greece	EL
Spain	ES
France	FR
Italy	IT
Cyprus	CY
Latvia	LV
Lithuania	LT
Luxembourg	LU
Hungary	HU
Malta	MT
Netherlands	NL
Austria	AT
Poland	PL
Portugal	PT
Romania	RO
Slovenia	SI
Slovakia	SK
Republic of Finland	FIN
Sweden	SE
United Kingdom	UK

*Source: http://publications.europa.eu

Annex I: Assessment of the methodologies applied by the Member States for justification of exceedances due to natural causes in 2008-2009

(AT) Austria 2008

Description of the methodology provided by the Member State

"Ferntransport von Saharastaub 2008.doc; Bruckmann_2008.pdf"

"A pronounced episode of long range transport of Saharan dust occurred from May 27 to June 1 2008. Elevated PM_{10} concentrations were observed as the particle cloud travelled from Southern France via parts of Italy, Switzerland and Austria to North-West Germany. The main transport of Saharan dust occurred aloft, leading to an uneven distribution of elevated PM_{10} concentrations at the ground. While most of Austria was affected near ground level, elevated PM_{10} burdens in Switzerland were only measured at two alpine stations. Chemical analyses and comparison of $PM_{2.5}/PM_{10}$ ratios revealed a change in the PM_{10} composition during the transport. While the high PM_{10} levels in the alpine region were mainly due to Saharan dust, visible in the high amount of PM_{10} and almost no change in $PM_{2.5}$, the PM_{10} maxima in North-West Germany were mainly caused by secondary aerosol and a smaller but still significant share of dust particles. This is confirmed by maxima in the $PM_{2.5}$ fraction occurring at the same time as the PM_{10} maxima and a high amount of ammonium nitrate and ammonium sulphate identified by chemical speciation. The exceedances of the European PM_{10} daily limit value during the episode can thus predominantly be assigned to natural sources in the Alpine region and in Bavaria, but only to a smaller part in North-Western Germany."

Text in English and German.

The summary coincides with the justification submitted by Germany (see below).

The method is qualitative, but not quantitative (allowing for the identification of events, but not for quantification of contributions in terms of PM mass).

Assessment of the methodology:

A combination of tools including the analysis of time series of PM_{10} levels, satellite imagery, back-trajectories, grain size distribution and chemical analyses was applied. The description of the event seems valid, as well as the methodology. Its limitation lies in the fact that this methodology is only applicable to specific episodes, and would not be optimal for broad-scale application in national or regional air quality networks due to its complexity. The justification provided is based on similar tools to the procedure recommended by Staff Working Paper 6771/11 (which was not available in 2008). Therefore the results are comparable, even though the procedure described in Staff Working Paper 6771/11 is recommended for future justification of natural events.

(CY) Cyprus 2008 and 2009

Description of the methodology provided by the Member State

"S14" (2008)

"Backtrajectories analysis for 2009 exceedance days has been uploaded together with this questionnaire".

Text in English.

The method is qualitative, but not quantitative (allowing for the identification of events, but not for quantification of contributions in terms of PM mass).

Assessment of the methodology:

For 2008, no justification was found to correspond to the code S14. Therefore, it is recommended that a methodology is provided for the justification of natural events. Given that the natural sources reported were mineral dust from outside the Member State (G2), it is recommended that the methodology described in Staff Working Paper 6771/11 be used for future justification of natural events.

Regarding the 2009 justification, back-trajectory analysis should be coupled with the assessment of aerosol maps in order to confirm the transport of mineral dust from source areas. Air masses may originate from desert areas without atmospheric injection of mineral dust. To avoid the misclassification of days with no mineral dust transport, Staff Working Paper 6771/11 recommends the application of a procedure based on a method developed in Spain and Portugal (Querol et al., 2006). The procedure may be summarised in the following tasks:

1. Identifying Saharan dust outbreak episodes
2. Quantifying Saharan dust outbreak episodes
3. Regional background measuring stations and spatial representativeness
4. Implementation of the method
5. Validation of the method
6. Annual Report
7. Critical discussion

In addition, the application of this procedure will allow for the quantification of African dust contributions to daily and annual PM_{10} levels, which is not possible by back-trajectory analysis. Methodology in Staff Working Paper 6771/11 is thus recommended for future justification of exceedances due to transport of mineral dust from outside the Member State.

(DE) Germany 2008

Methodologies for justification were submitted by two different reporting bodies (Springer VDI and Landesamt für Natur, Umwelt und Verbraucherschutz Nordrhein-Westfalen, Essen):

a) Description of the methodology provided by the Member State

"A pronounced episode of long range transport of Saharan dust occurred from May 27 to June 1 2008. Elevated PM_{10} concentrations were observed as the particle cloud travelled from Southern France via parts of Italy, Switzerland and Austria to North-West Germany. The main transport of Saharan dust occurred aloft, leading to an uneven distribution of elevated PM_{10} concentrations at the ground. While most of Austria was affected near ground level, elevated PM_{10} burdens in Switzerland were only measured at two alpine stations. Chemical analyses and comparison of $PM_{2.5}/PM_{10}$ ratios revealed a change in the PM_{10} composition during the transport. While the high PM_{10} levels in the alpine region were mainly due to Saharan dust, visible in the high amount of PM_{10} and almost no change in $PM_{2.5}$, the PM_{10} maxima in North-West Germany were mainly caused by secondary aerosol and a smaller but still significant share of dust particles. This is confirmed by maxima in the $PM_{2.5}$ fraction occurring at the same time as the PM_{10} maxima and a high amount of ammonium nitrate and ammonium sulphate identified by chemical speciation. The exceedances of the European PM_{10} daily limit value during the episode can thus predominantly be assigned to natural sources in the Alpine region and in Bavaria, but only to a smaller part in North-Western Germany."

Text in English.

The method is qualitative, but not quantitative (allowing for the identification of events, but not for quantification of contributions in terms of PM mass).

Assessment of the methodology:

The methodology provided for justification is rather descriptive and does not provide specific technical data as to how the Saharan dust plume was detected or how the distribution of Saharan dust along the vertical air column was determined. The missing technical data are included in the justification provided by Austria (see above), but the link to this justification (from AT) should be provided in the one given by Germany. Both Member States refer to the same document, even though this was not clearly stated by

Germany and it is only understandable once both texts have been read. As in the case of Austria, the justification provided is based on similar tools to the procedure recommended by Staff Working Paper 6771/11 (which was not available in 2008). Therefore the results are comparable, even though the procedure described in Staff Working Paper 6771/11 is recommended for future justification of natural events.

b) Description of the methodology provided by the Member State
"Staub aus der Sahara verfärbt Regenwasser rötlich-braun".
Text in German.
The method is qualitative, but not quantitative (allowing for the identification of events, but not for quantification of contributions in terms of PM mass).

Assessment of the methodology:
The methodology applied for justification was based on the analysis of meteorological charts, back-trajectories and photographs of red rain deposition on vehicles. The assessment of the African dust episode seems valid, even though the methodology would only be applicable for very specific and evident cases of African dust transport, and it would not be optimal for broad-scale application in national or regional air quality networks (due to the fact that it is not detailed enough). The procedure described in Staff Working Paper 6771/11 is recommended for future justification of natural events.

(ES) Spain 2008 and 2009
Description of the methodology provided by the Member State
"Estudio y evaluación de la contaminación atmosférica por material particulado en España. Informe episodios naturales 2008".
Text in Spanish.
The method is qualitative and quantitative (allowing for the identification of events and for quantification of contributions in terms of PM mass).
The same method was used for 2009.

Assessment of the methodology:
Justification was carried out by applying the procedure recommended by Staff Working Paper 6771/11. The procedure was described in detail by Querol et al. (2006).

(FR) France 2008 and 2009
Description of the methodology provided by the Member State
"analyse_PM2008_RA".
Text in French.
The method is qualitative, but not quantitative (allowing for the identification of events, but not for quantification of contributions in terms of PM mass).
"corrélation avec la houle" (2009). Text in French.

Assessment of the methodology:
A combination of tools was applied for the identification of an African dust event (G2): meteorological charts, PM_{10} and $PM_{2.5}$ grain size distribution, and chemical analyses. The identification of the event seems valid, as it coincides with the dates identified as affected by African dust transport by neighbouring Member States. The justification provided is based on similar tools compared to the procedure recommended by Staff Working Paper 6771/11 (which was not available in 2008).However, this is only true for qualitative identification of the dust events, not for quantification of dust contributions. Therefore the results are comparable with results from the procedure recommended by Staff Working Paper 6771/11, but the latter is recommended for future justification of natural events.

No justification is provided for the sea salt contributions ("embruns marins") reported by 2 stations in 2008 (see Table 4.6a). As for 2009, the justification provided ("corrélation avec la houle") is insufficient to carry out any technical assessment. It is recommended that a detailed description of the methodology will be provided for future justifications of natural events. For the justification of sea salt contributions, Working Paper 6771/11 recommends the use of a method "based on a chemical analysis of 24-hour PM_{10} samples to determine exceedances due to sea salt contribution at the individual PM_{10} sampling point. Estimation of sea salt natural contribution on an annual basis can be adequately achieved through application of campaign-based measurements (as described below) and modelling. However, such annual estimates cannot be interpreted as the contribution at a particular day. Member States are also advised not to use such annual estimates in the source apportionment that steers abatement measures in an air quality plan, as it may lead to future exceedances due to overestimation of natural contributions during high pollution episodes.". However, this Working Paper was not available in 2008.

(GB) Great Britain 2008 and 2009

Description of the methodology provided by the Member State

"The method uses mapped annual mean particulate chloride concentrations on a 5 km x 5 km grid interpolated from measurements at 28 rural monitoring sites in the CEH rural network (http://www.cara.ceh.ac.uk/hno3network/index.html). Sea salt has been estimated from chloride by applying a scaling factor of 1.648 to take account of the sodium ion. The function was then used to estimate the change in the number of exceedance days for the sea salt concentration at the vicinity of the limit value.

The method is applied on an annual and a daily basis. Results are compared, and a reasonable consistency between the annual and daily methods is found".
Text in English.
The method is qualitative and quantitative (allowing for the identification of events and for quantification of contributions in terms of PM mass).

Assessment of the methodology:

- The method used by UK is based on interpolation of measurement data based on chloride levels. However, according to Staff Working Paper 6771/11, "examples of methodologies that estimate the sea salt contribution on the basis of tracer elements such as sodium and chloride are available, but for the time being there is insufficient spatial coverage to produce reliable maps of sea salt concentration on a daily basis". Staff Working Paper 6771/11 recommends the use of a method "based on a chemical analysis of 24-hour PM_{10} samples to determine exceedances due to sea salt contribution at the individual PM_{10} sampling point. Estimation of sea salt natural contribution on an annual basis can be adequately achieved through application of campaign-based measurements (as described below) and modelling. However, such annual estimates cannot be interpreted as the contribution at a particular day. Member States are also advised not to use such annual estimates in the source apportionment that steers abatement measures in an air quality plan, as it may lead to future exceedances due to overestimation of natural contributions during high pollution episodes". However, this Working Paper was not available in 2008.
- Staff Working Paper 6771/11 presents one other possibility: "a simpler and widely used method to calculate the sea salt contribution considers only sodium or chloride as a tracer for the sea salt contribution. The contribution is calculated assuming that sea salt is made up only by NaCl and that all Na and Cl are associated in sodium chloride." However, it also states that the use of chloride only

is potentially subject to both positive and negative artefacts, to finally discourage the use of chloride as a tracer of sea spray: “it is not recommended to use chloride as a reference ion to calculate the sea salt contribution because chloride observations have a large margin of uncertainty.” However, it is stated in the UK justification report that “daily sodium data are not available, so chloride is the only option for the daily method.” And that “we believe that the use of chloride as a marker is less likely to lead to an overestimate than the use of sodium.”

- Sea salt is not the only source of particulate chloride in the atmosphere. Hydrogen Chloride (HCl) is emitted from coal burning but reductions in coal use and flue gas abatement are likely to have reduced atmospheric HCl and ammonium chloride concentrations considerably. There will also be loss of chloride from marine aerosol due to reactions with nitric acid.
- The composition of sea salt is dominated by chloride and sodium. Other components contributing more than 1% by mass are sulphate, magnesium, calcium and potassium. In the case of UK, it was decided to simplify the analysis by assuming that the sea salt consists of sodium chloride only. Thus the measured chloride concentration was scaled by a factor of 1.648. An alternative approach would be to scale by 1.809 to take account of the full composition of sea salt.

(GB) Gibraltar 2008 and 2009

Description of the methodology provided by the Member State

“The Spanish authorities have, for the preparation of ongoing mandatory reporting to the Commission for Spain, identified days in 2009 on which regional background sites across the Iberian Peninsula were significantly affected by African dust (Pey, J., 2009), referred to here as ‘African dust days’, using a qualitative methodology (Querol, et al. 2007)”

Text in English.

The method is qualitative and quantitative (allowing for the identification of events and for quantification of contributions in terms of PM mass).

Assessment of the methodology:

Justification was carried out by applying the procedure recommended by Staff Working Paper 6771/11. The procedure was described in detail by Querol et al. (2006). This justification applied only to mineral dust, and not to sea salt aerosols.

(GR) Greece 2008 and 2009

Description of the methodology provided by the Member State

“μοντέλο μεταφοράς σκόνης SKIRON, δορυφορικές εικόνες TOMS και SEAWIFS”.

Text in Greek.

The method is qualitative and quantitative (allowing for the identification of events and for quantification of contributions in terms of PM mass).

Assessment of the methodology:

Even though the justification was provided by this Member State in Greek, it is understandable that aerosol maps SKIRON (forecast.uoa.gr) and TOMS (ozoneaq.gsfc.nasa.gov), as well as satellite imagery SEAWIFS (oceancolor.gsfc.nasa.gov/SeaWiFS/) were used. The natural exceedences reported by GR in 2008 and 2009 were due to transport of mineral dust from outside the Member State (G2), wildland-fires inside (D1) and outside (D2) the Member State:

a) *Mineral dust from outside the Member State*: it is assumed (from the description of the methodology provided by the Member State) that transport of mineral dust was identified by means of the analysis of aerosol maps and satellite images. This is a valid approach, but it should be complemented with back-

trajectory analysis to confirm the transport of the dust-laden air masses. For future justifications, the application of the procedure described by Staff Working Paper 6771/11 is recommended, which will in addition allow for the quantification of African dust contributions to daily and annual PM_{10} levels. This is not possible by the analysis of SKIRON and TOMS maps and SEAWIFS images.

b) *Wildland-fires inside and outside the Member State*: Staff Working Paper 6771/11 states that "wild-land fires are usually of anthropogenic origin, and may be prevented or controlled with appropriate actions to a significant extent. Member States should therefore effectively address this contribution in view of the possibility of subtraction only if the emissions are transported from regions outside the Member State and when provisions of Directive 2008/50/EC related to the transboundary pollution have been applied. If the fire has extended from another Member State any deductions of the common contribution need to be accompanied with the description of measures taken at the short term to eliminate the fire and reduce the exposure of the population". Currently, no exhaustive method for the identification and the quantification of the impact of wild-land fire episodes have been developed and communicated to the Commission in the framework of the annual report on air quality assessment. In absence of developed and implemented methodologies, a summary of a possible method for the quantification of wild-land fire contributions that fulfils the key principles is described in Staff Working Paper 6771/11. Satellite images are listed as tools to identify the aerosol plumes originating from the wild-land fire. It is essential that the Member States justify that the wildland-fire was originated by natural causes, when originating inside the Member State. This was not the case in GR in 2009. For the 2008 justification, evidence should have been provided regarding the fact that provisions of Directive 2008/50/EC related to the transboundary pollution were applied. This was not the case, either.

(IT) Italy 2008 and 2009

Description of the methodology provided by the Member State

No justification was provided.

Assessment of the methodology:

It is recommended that a methodology is provided for the justification of natural events. Given that the natural sources reported were mineral dust from outside the Member State (G2) and natural events (S8), it is recommended that the methodology described in Staff Working Paper 6771/11 is used for future justification of natural events.

(LV) Latvia 2009

Description of the methodology provided by the Member State

"5%", this was the only description provided by the MS.

Assessment of the methodology:

No justification was provided. It is recommended that a methodology is provided for the justification of natural events. Given that the natural sources reported were grouped under the more general code S8, it is not possible to infer what type of natural sources the justification refers to.

(MT) Malta 2008 and 2009

Description of the methodology provided by the Member State

"Justification report on natural PM_{10}".
"The basis for the identification of Sahara dust events is the methodology also described in the guidance document provided by the European Commission (2009). The PM_{10} measured in Malta and in particular at the background station in Gharb (MT0007A) were analysed in detail by Axisa (2009) using backward trajectories, satellite images, PM2.5/PM10 ratios and chemical speciation of low volume sampler filters in order to identify their likely origin".
Text in English. T
The method is qualitative and quantitative (allowing for the identification of events and for quantification of contributions in terms of PM mass).

"The calculation of sea salt concentrations is based on the method provided by the guidance (European Commission, 2009) to calculate the sea salt concentration from measured chloride and sodium concentrations. In 2008, a total of 38 PM_{10} low volume sampler filters were collected from the traffic site in Msida and analysed for ions. As described in the guidance the concentrations of chloride and sodium were used to calculate the contribution of sea salt using the following formula (sea salt = $1.176*(Na^{+}+Cl^{-})$)".
Text in English.
The method is qualitative and quantitative (allowing for the identification of events and for quantification of contributions in terms of PM mass).

Assessment of the methodology:
The methodology applied for the justification of natural exceedances due to African dust contributions is based on an early version of the procedure provided by the European Commission (2009). This would be a valid approach given that the final version was only published in 2011. However, the report from MT states that "as described in the Guidance of the European Commission (2009), the regional and temporary background concentration was determined by averaging the background measurements approximately 15 days before and after each particular episode. The resulting surplus was then subtracted from the respective values of the traffic station in Msida". This was applied for the 2008 and 2009 data. The guidance document states that the calculation should refer to the 40^{th} percentile of the background PM_{10} levels, not the average of the background levels. The calculation of the average instead of the 40^{th} percentile would probably result in an underestimation of the African dust contributions. It is recommended that this bias is corrected for future justifications of natural events.

The methodology applied for the justification of natural exceedances due to sea salt contributions is based on the method provided by the Staff Working Paper 6771/11 (European Commission, 2009), and it therefore is judged to be valid. Sea salt concentrations were calculated correctly by measuring ion concentrations at point sites where sea salt contributions were reported. Where no ion measurements were available, the correlation between wind speed and sea salt concentration was analysed". This extrapolation is not recommended by Staff Working Paper 6771/11. However, this document was not available at the time of justification given here.

(PT) Portugal 2008 and 2009
Description of the methodology provided by the Member State
"Annexed report".
Text in Portuguese.
The method is qualitative and quantitative (allowing for the identification of events and for quantification of contributions in terms of PM mass).

Assessment of the methodology:
Justification was carried out by applying the procedure recommended by Staff Working Paper 6771/11. The procedure was described in detail by Querol et al. (2006).

For year 2008, at the time of justification, the use of the 30th percentile was recommended, as opposed to the 40th percentile recommended in the final version. This issue was already corrected in the justification of the 2009 natural exceedances due to African dust contributions.

A methodology for the justification of exceedances due to wildland-fires was also provided in 2008 and 2009, even though no exceedances were ascribed to this natural cause according to forms 23.

Annex II: List of forms in the AQ questionnaire

Form 0	General information, update history
Form 1	Contact body and address
Form 2	Delimitation of zones and agglomerations
Form 3	Stations and measuring methods used for assessment under first, second and fourth DD
Form 4	Stations used for assessment of ozone, including nitrogen dioxide and nitrogen oxides in relation to ozone
Form 5	Stations and measuring methods used for the assessment of recommended volatile organic compounds (3rd DD) and other relevant PAH and metals in ambient air and deposition (4th DD)
Form 6	Stations and measurement methods used for the assessment of other ozone precursor substances
Form 7	Methods used to sample and measure PM_{10} and $PM_{2.5}$, ozone precursor substances, arsenic, cadmium, nickel, mercury, PAH: optional additional codes to be defined by the Member State
Form 8	List of zones and agglomerations where levels exceed or do not exceed limit values or limit values plus margin of tolerance for pollutants listed in first and second DD
Form 9	List of zones and agglomerations where levels exceed or do not exceed target values or long term objectives for ozone and arsenic, cadmium, nickel, B(a)P and $PM_{2.5}$
Form 10	List of zones and agglomerations where levels exceed or do not exceed upper assessment thresholds or lower assessment thresholds, including information on the application of supplementary assessment methods
Form 11	Individual exceedances of limit values and limit values plus the margin of tolerance of pollutants listed in first and second DD
Form 12	Reasons for individual exceedances: optional additional codes to be defined by the Member State
Form 13	Individual exceedances of ozone thresholds
Form 14	Exceedance of target values of ozone, arsenic, cadmium, nickel, benzo(a)pyrene and $PM_{2.5}$
Form 15	Annual statistics of ozone, arsenic, cadmium, nickel, and benzo(a)pyrene
Form 16	Annual average concentrations of ozone precursor substances of mercury and other relevant PAH and deposition rates of mercury and other relevant PAH
Form 17	Monitoring data on 10 minutes mean SO_2 levels
Form 18	Monitoring data on 24hr mean $PM_{2.5}$ levels
Form 19	Tabular results of and methods used for supplementary assessment
Form 20	List of references to supplementary assessment methods referred to in Form 19
Form 21	Exceedance of limit values for SO_2 due to natural sources
Form 22	Natural SO_2 sources: optional additional codes to be defined by Member State
Form 23	Exceedance of limit values of PM_{10} due to natural events
Form 24	Exceedance of limit values of PM_{10} due to winter sanding
Form 25	Consultations with other MS on transboundary pollution
Form 26	Exceedances of limit values laid down in Directives 85/203/EEC
Form 27	Reasons for exceedances of limit values laid down in Directives 85/203/EEC: optional additional codes to be defined by the Member State