

# Particulate matter from natural sources and related reporting under the EU Air Quality Directive in 2008 and 2009

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# Executive summary

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Much of the air pollution that damages human health and the environment today is the result of human activities. But natural sources also emit air pollutants, contributing to the exposure of European citizens and ecosystems to bad air quality — and potentially undermining EU Member State efforts to meet the air quality standards set out in EU legislation.

Recognising the challenge presented by natural air pollution, the Air Quality Directive <sup>(1)</sup> provides that before Member States compare ambient air pollutant concentrations with relevant legally binding limit values they may subtract the contribution of natural sources.

This report provides a first evaluation of Member State reporting on natural air pollution under the Air Quality Directive. Its main objectives are:

- to provide an overview of the main natural sources of air pollutants;
- to summarise methods that can be used to quantify natural source contributions to air pollution levels;
- to analyse Member State reporting on natural air pollutant contributions to limit value exceedances for the years 2008 and 2009;
- to identify recommendations for improvements, particularly proposals relevant to current efforts to develop an electronic reporting mechanism on natural source contributions.

## *Natural sources of particulate matter*

European Commission Staff Working Paper 6771/11 (EC, 2011a) provides guidance on which sources can be regarded as 'natural' and on methods to quantify and subtract the contribution of these sources in the framework of the Air Quality Directive. The Working Paper also defines principles to be applied when evaluating Member State reporting of exceedances due to natural contributions. It does not specify the pollutants for which an exception on natural sources may be requested.

Completed reporting questionnaires for the years 2008 and 2009 reveal that particulate matter (PM<sub>10</sub>) was the only natural contribution to limit value exceedances reported by Member States. Since natural PM<sub>10</sub> sources affect health and the environment, it is important that Member States consider all appropriate measures to reduce excessive exposure and ensure accurate reporting of such natural air pollutant contributions.

According to the Staff Working Paper, when PM<sub>10</sub> limit values are exceeded, contributions from the following natural sources may be subtracted:

- **Wind-blown desert dust**, i.e. natural particulate matter transported from dry regions. Arid zones in North Africa are the major source in the European Union.
- **Sea spray aerosols**, which are finely dispersed particles emitted from the sea surface.
- Particulate matter emitted by **volcanos** and **seismic activities**.
- Particulate matter emitted by **wild-land fires**. Such fires are caused by burning non-managed and managed forests and other vegetation, excluding agricultural burning of stubbles etc.

A pollution source can only be considered 'natural' if it involves no direct or indirect human activity. This can hamper attempts to justify air pollution resulting from events that can occur with or without human intervention, such as wild-land fires.

## *Methodologies to identify particulate matter with a natural origin*

Chemical analysis of particulate matter sampled at regional background measurement stations enables to identify the origin of natural PM. For example:

- African dust consists mainly of silicate minerals and carbonates, with the precise composition dependent on the geographical origin.

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<sup>(1)</sup> Directive 2008/50/EC on Ambient Air Quality and Cleaner Air for Europe (EC, 2008).

- Sodium chloride (NaCl) is the main component in sea spray.
- Particulate matter emitted directly during a volcanic eruption (primary PM) often has a characteristic composition. Sulphur dioxide (SO<sub>2</sub>), usually the major gaseous compound emitted, also contributes to the formation of secondary PM, i.e. particulate matter built from precursor substances in the atmosphere.
- During wild-land fires, mainly fine particulate matter (PM<sub>2.5</sub>), which is a fraction of PM<sub>10</sub>, is emitted directly into the air. Further, non-methane volatile organic compounds (NMVOC) are emitted by such fires, and they participate in chemical reactions, which lead to the formation of organic particulate matter (secondary organic aerosol).

Since PM is dispersed and transported in the atmosphere, measurements need to be interpreted in the light of meteorological information. Meteorological, back trajectory, dispersion, receptor and source allocation models are often applied to interpret PM measurement data. Remote sensing methods using data provided by sensors on satellites and/or light detection and ranging (LIDAR) technologies can further support interpretations. A combination of the approaches listed above can be used for qualitative and for quantitative analyses,

supporting Member State reporting of natural contributions to PM limit value exceedances.

*Exceedances of air pollutant limit values due to natural source contributions reported in 2008 and 2009*

Eleven Member States reported exceedances of PM<sub>10</sub> limit values in 2008 and/or 2009 (Austria, Cyprus, Germany, Greece, France, Italy, Latvia, Malta, Portugal, Spain and the United Kingdom). The daily PM<sub>10</sub> limit value was exceeded more frequently and at more stations than the annual mean. As shown in Table ES.1, the highest numbers of exceedances were reported by Mediterranean Member States (Cyprus, France, Greece, Italy and Spain).

The highest number of stations reporting natural contributions was located in Spain. The main natural source contributing to exceedances was 'transport of natural particles from dry regions outside the Member State' (Saharan dust), followed by sea spray and wild-land fires.

*Recommendations to improve reporting of natural contributions*

It was not within the scope of this report to provide a coherent overview of the contribution of natural

**Table ES.1 Number of stations reporting exceedances of the PM<sub>10</sub> daily limit value before and after subtraction of natural contributions**

| Member State   | Number of stations reporting exceedances of the daily PM <sub>10</sub> limit value |                   |                    |                   |
|----------------|--|-------------------|--------------------|-------------------|
|                | 2008   |                   | 2009               |                   |
|                | Before subtraction   | After subtraction | Before subtraction | After subtraction |
| Austria        | 10   | 8                 | 6                  | 6                 |
| Cyprus         | 1  | 0                 | 1                  | 1                 |
| Germany        | 17   | 16                | 33                 | 33                |
| Spain          | 123  | 72                | 42                 | 24                |
| France         | 31   | 30                | 41                 | 41                |
| United Kingdom | 3  | 2                 | 5                  | 1                 |
| Greece         | 15   | 11                | 7                  | 2                 |
| Italy          | 188  | 187               | 177                | 177               |
| Latvia         | 3  | 3                 | 3                  | 3                 |
| Malta          | 1  | 0                 | 1                  | 0                 |
| Portugal       | 10   | 7                 | 10                 | 7                 |

**Note:** For the United Kingdom the figures include some exceedances determined by supplementary assessment (modelling).



emission sources to air pollutant concentrations or even limit value exceedances in Europe.

In general, the analysis indicates that national reporting on natural contributions for the years 2008 and 2009 was reliable. However, it was hard to detect a common Europe-wide strategy on how to deal with the natural emissions and their contributions to limit value exceedances when analysing Member State submissions. The eleven countries that reported natural contributions to PM<sub>10</sub> limit value exceedances used quite different approaches. It is probable that the natural PM events or episodes reported would have affected more Member States than those that did report.

Commission Implementing Decision 2011/850/EU on reporting under Directives 2004/10/EC and 2008/50/EC was adopted at the end of 2011 (EC, 2011d). It provides that the existing air quality questionnaire

submitted by Member States will be replaced with an electronic reporting mechanism currently under development. In this context, this report's analysis suggests that reporting could be enhanced through the following actions:

- streamlining and clarifying the reporting of natural contributions by harmonising the code lists in future guidance documents;
- providing a clear recommendation concerning the tracer that should be used to identify natural sea spray contributions;
- including in national justification documents a brief English-language summary of the methodologies used, and publishing the documents on publicly accessible websites within Member States.

# 1 Introduction

## 1.1 Background

Much air pollution results from human activities. But natural sources also emit air pollutants, contributing to the exposure of European citizens and ecosystems to bad air quality.

The Air Quality Directive (EC, 2008) provides that before Member States compare ambient air pollutant concentrations with relevant legally binding limit values they may subtract the contribution of natural air pollution sources. Pollutants from natural sources do harm human health and the environment, however, and Member States should therefore consider all appropriate measures to reduce excessive exposure.

European Commission Staff Working Paper 6771/11 (EC, 2011a) provides guidance on which sources can be regarded as 'natural' and on methods to quantify and subtract the contribution of these sources. The Staff Working Paper was officially published in 2011 but drafts were available when the Member States reported 2008 and 2009 data to the European Commission — the years reviewed in the present report. The Staff Working Paper does not specify which pollutants may be subject to natural source contributions, referring simply to 'regulated pollutants exceeding the air quality limit values set in the EU's ambient air quality legislation'.

Before Staff Working Paper 6771/11 was published, the European Commission's Decision 2004/461/EC (EC, 2004a) established the questionnaire on air quality assessment (EC, 2011b) as the tool for annual reporting under the Air Quality Directive (EC, 2008).

This questionnaire and the accompanying guidance were updated in June 2009 to include additional information required for reporting under the Air Quality Directive. The Commission also provided a guidance document to assist in completing the questionnaire (EC, 2011c). Further, the Air Quality Directive expanded the range of sources that could be considered as 'natural' when reporting limit value exceedances (e.g. sea spray — see footnote 2 in the Staff Working Paper).

Forms 21 to 23 of the questionnaire refer to information concerning the exceedance of SO<sub>2</sub> and PM<sub>10</sub> limit values due to contributions from natural sources (Table 1.1). They thus refer to requirements of Article 20 of the Directive ('Contributions from natural sources'). In completing these forms, Member States must cite the methods used to assess the natural contribution to exceedances, documenting any models used (EEA, 2011).

In form 11 of the questionnaire, so-called 'reason codes' for individual exceedances have to be used (Table 1.1). Although one of them is 'Natural sources or natural events' (see Table 6.2), form 11 is not in fact relevant for Article 20 of the Air Quality Directive.

The questionnaires submitted by Member States containing 2008 and 2009 data indicate that natural contributions to exceedances were only reported for particulate matter, PM<sub>10</sub> (see Box 1.1). No exceedances of the sulphur dioxide (SO<sub>2</sub>) limit values, which might have been linked to volcanic eruptions and/or geothermal activity inside or outside Member States, were reported. Therefore,

**Table 1.1** Forms in the questionnaire on air quality assessment relevant for reporting the contribution of natural sources to limit value exceedances

| Form number |   |
|-------------|---|
| 11          | Individual exceedances of limit values and limit values plus the margin of tolerance of pollutants listed in the First and Second Daughter Directives (*) |
| 21          | Exceedance of the limit values for SO <sub>2</sub> due to natural sources   |
| 22          | Natural SO <sub>2</sub> sources: optional additional codes to be defined by the Member State  |
| 23          | Exceedance of limit values of PM <sub>10</sub> due to natural events  |

**Note:** (\*) The 'First Daughter Directive' is Council Directive 1999/30/EC relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air (EC, 1999). The 'Second Daughter Directive' is Directive 2000/69/EC of the European Parliament and of the Council relating to limit values for benzene and carbon monoxide in ambient air (EC, 2000).

**this report only addresses reported exceedances of the PM<sub>10</sub> daily and annual limit values (Table 1.2).**

The Staff Working Paper (EC, 2011a) sets out six key principles that the Commission applies when evaluating Member State claims that an exceedance is due to natural contributions:

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

Special attention should be paid to the first principle, which implies that a source can only be considered 'natural' if it involves no direct or indirect human activity. This can hamper attempts to justify air pollution resulting from events that can occur with or without human intervention, such as wild-land fires. For such fires to be classified as natural sources they must have been initiated by natural causes (e.g. lightning). Human-related causes such as littering of forests should not be reported as natural events.

## 1.2 Natural air pollution contributions that may be subtracted

The Staff Working Paper (EC, 2011a) provides a non-exhaustive list of sources whose contributions may be subtracted from national air pollution figures:

- **Wind-blown desert dust particles (African dust)**

This term refers to the transport of natural particles from dry regions, i.e. resuspended and transported (wind-blown) desert dust particles that have a strong impact on atmospheric visibility and aerosol composition and on particulate matter levels. Arid zones in North Africa are the major source.

- **Sea spray aerosols**

Sea spray aerosols, containing sea salt, are finely dispersed particles formed by the action of the wind on the sea and emitted into the air.

- **Volcanic dust particles**

Particles emitted by volcanic eruptions and seismic activities.

- **Wild-land fire particles**

Such fires are caused by burning non-managed and managed forests and other vegetation, excluding agricultural burning of stubbles etc. Again, however, the Staff Working Paper notes that the fire must have a natural cause in order to be considered a 'natural' source.

Methodologies for identifying and quantifying the contribution of these sources are described and discussed in the Staff Working Paper.

A second non-exhaustive list in the Staff Working Paper sets out sources that the Commission does not consider to be eligible for subtraction when PM limit values are exceeded:

- **Primary biological aerosols**

Primary biological aerosols include, for example, spores (very small in size and part of PM<sub>10</sub>) or pollen (typically larger in size than PM<sub>10</sub>), which originally derive from biological

**Table 1.2 Limit values for PM<sub>10</sub> as given in the Air Quality Directive (see also Box 1.1)**

| Size fraction    | Averaging period | Value                | Comments   |
|------------------|------------------|----------------------|--|
| PM <sub>10</sub> | One day          | 50 µg/m <sup>3</sup> | Not to be exceeded on more than 35 days per year |
| PM <sub>10</sub> | Calendar year    | 40 µg/m <sup>3</sup> | –  |

**Note:** The deadlines for achieving the limit values and possible extensions of those deadlines are set out in the Air Quality Directive.

**Source:** EC, 2008.

### Box 1.1 Dust, aerosols, particles, particulate matter, $PM_{10}$ and $PM_{2.5}$

**Dust** is a common word used for dry particles of earth or other matter that can be raised and blown away by the wind. In atmospheric science the word **aerosol** is normally used. An aerosol is a suspension of fine solid particles or liquid droplets in the air (most aerosols can actually be found in clouds).

This report also employs the word **particle** when addressing **particulate matter (PM)** in the air. The EU's air quality legislation covers the mass concentrations, measured in micrograms per cubic meter ( $\mu\text{g}/\text{m}^3$ ), of two size fractions suspended in the air:

- **$PM_{10}$**  is particulate matter with an aerodynamic diameter of 10  $\mu\text{m}$  or less;
- **$PM_{2.5}$**  is particulate matter with an aerodynamic diameter of 2.5  $\mu\text{m}$  or less (also termed 'fine PM').

processes and which are emitted into the atmosphere without change in their chemical composition.

- **Secondary organic biogenic aerosols**

These are the organic fraction of PM formed in the atmosphere ('secondarily') via a chain reaction of volatile organic compounds emitted by soils and vegetation. Secondary organic biogenic aerosols are a common fraction of  $PM_{10}$  and can be important during the summer, particularly in areas covered with vegetation. However, exceedances of the  $PM_{10}$  limit values in such areas are rare.

- **Resuspension of dust particles**

Major sources of resuspended particles addressed here are roads and pavements in cities. The suspension process can be caused naturally (e.g. by wind) or by human activities (e.g. by passing vehicles). The contribution of each cannot be quantified adequately at present.

Decision 2004/461/EC provides a list of natural sources and standard codes for use in justifying natural contributions to limit values exceedances. The events listed in the decision do not exactly match the list of sources or events given in the Staff Working Paper (see Chapter 3) because the Decision predates the expanded list of sources included in the Air Quality Directive (EC, 2008).

### 1.3 Report structure

Chapters 2 to 5 of this report provide an overview of major natural emission sources affecting air quality in Europe, summarising the major natural events and available qualitative and quantitative tools to identify those events.

Chapter 6 uses the years 2008 and 2009 to critically analyse Member State reporting of natural contributions to exceedances of the  $PM_{10}$  limit values under the Air Quality Directive. The analysis addresses the methodologies used to justify natural contributions to air pollution and concludes with recommendations on how to improve reporting.

## 2 Wind-blown desert dust particles – African dust

Most of the naturally emitted mineral dust released to the atmosphere is from arid or semi-arid areas. The major dust source areas relevant for the European Union (EU) are located along the west coast of North Africa (León and Legrand, 2003; Prasad and Singh, 2007).

### 2.1 Long-range transport of particulate matter emitted from dry regions

North Africa is considered a typical example of a hot desert area where rain is extremely rare. The average annual rainfall in the Western Sahara is below 25 millimetres (mm) and even lower in the Eastern Sahara (five mm on average). Further, precipitation occurs very irregularly, i.e. no rain may fall in some areas, followed by a single intense thunderstorm (Encyclopaedia of Earth, 2012). In environments with extremely low precipitation levels and very high average temperatures (> 50 °C in summer) episodes of massive dust resuspension are common, leading to high levels of PM in the air (African dust).

The warming of the surface during the day leads to strong vertical thermal turbulence, which can reach altitudes of up to 4 000–5 000 meters in summer (Dubief, 1979). Such turbulence is usually followed by periods of nocturnal stability. This cycle inhibits the deposition of resuspended particles propelled to considerable atmospheric heights (Moulin et al., 1998), leading to particle residence at these altitudes lasting weeks or even months. The result is so-called 'dry smog'. Material from this semi-permanent reservoir of emitted dust can be transported over long distances by atmospheric mechanisms.

In Europe, African dust can significantly increase ambient particulate matter levels, particularly in southern European countries (Bergametti et al., 1989). It is a well-recognised source contributing to exceedances of the PM limit values set in the EU legislation (Querol et al., 1998; Rodriguez et al., 2001; Viana et al., 2002; Escudero et al., 2005 and 2007; Gerasopoulos et al., 2006; Kallos et al., 2006; Kocak et al., 2007; Mitsakou et al., 2008; Pey et al., 2009a).

According to studies, a variety of meteorological scenarios can lead to the transportation of African dust air masses towards Europe (Box 2.1).

A recently published paper on African dust in the Mediterranean region (Querol et al., 2009) summarises the seasonality, occurrence and intensity of African dust episodes over the whole Mediterranean basin. A number of PM<sub>10</sub> data series from monitoring sites across the European Mediterranean region were evaluated. The study revealed clearly increasing PM<sub>10</sub> values from north to south and from west to east across the basin, almost matching the PM<sub>10</sub> African dust load patterns (Map 2.1). The authors noted that PM<sub>10</sub> background levels were 5–10 µg/m<sup>3</sup> higher in the eastern Mediterranean basin compared to the western Mediterranean basin.

Significant seasonal PM<sub>10</sub> trends were observed when comparing eastern and western sites, largely driven by the occurrence of African dust events. More frequent dust events in spring and early summer affecting the eastern sites caused higher PM levels. Likewise, the summer maximum observed in the western Mediterranean roughly coincided with the most intense periods of African dust outbreaks. This is apparent despite the fact that recirculation of aged air masses significantly increases the background PM<sub>10</sub> levels in this region.

Further, Querol et al. (2009) identified important inter-annual variations in the contribution of dust to overall PM<sub>10</sub> levels, particularly 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 eastern Mediterranean basin corresponded with anomalously low contributions over the western Mediterranean basin, and vice versa.

De Leeuw (2012) cites the national questionnaire report submitted by Cyprus as an example of African dust intrusions causing high PM<sub>10</sub> concentrations. In 2010 mean concentrations exceeding 200 µg/m<sup>3</sup> were recorded on several days at the regional background station in Agia Marina; on one day the value exceeded 1 000 µg/m<sup>3</sup>.

### 2.2 Chemical composition of African dust

The regular dust intrusions moving from the African continent derive from different sources

**Box 2.1 Meteorological scenarios leading to long-range transport of African dust**

For the **western Mediterranean**, Rodríguez et al. (2001) and Escudero et al. (2005) defined three scenarios typically favouring the transportation of mineral particles from the Sahara and Sahel deserts. The transport may be due to:

1. A low atmospheric pressure system over the Atlantic (near Portugal) and/or over the Morocco area, normally in early autumn and spring.
2. A high pressure system (at surface level) over the Iberian peninsula and western Mediterranean and/or over northern Africa. Such situations are generally observed in January–March, causing dust plumes to develop over the Atlantic with a well-defined convex morphology. The plumes can be transported towards the Iberian peninsula from the west.
3. A North African anticyclone — a wind flow in the upper levels of the atmosphere associated with a high atmospheric pressure system. This is the most common scenario transporting African dust towards western Europe, mainly occurring in summer.

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 responsible for the transport of large amounts of mineral dust particles:

4. In spring and early summer, Saharan thermal lows can develop in the south of the Atlas Mountains 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). Cyclones travel eastward along this thermal gradient and finally cross the Mediterranean between Libya and Egypt. They constitute the main atmospheric scenario responsible for the transport of desert dust over the eastern Mediterranean Basin.
5. Severe episodes can also be associated with the combination of a deep trough over the west Mediterranean and north-west Africa and relatively high pressures towards the eastern part of the Mediterranean. This scenario is related to the evolution scenario 1 in the western Mediterranean.

and therefore contain different mixtures of mineral particulate matter. In combination with other tools, analysis of the composition of particulate matter is therefore an important method to detect episodes of dust transported from Africa (Figure 2.1).

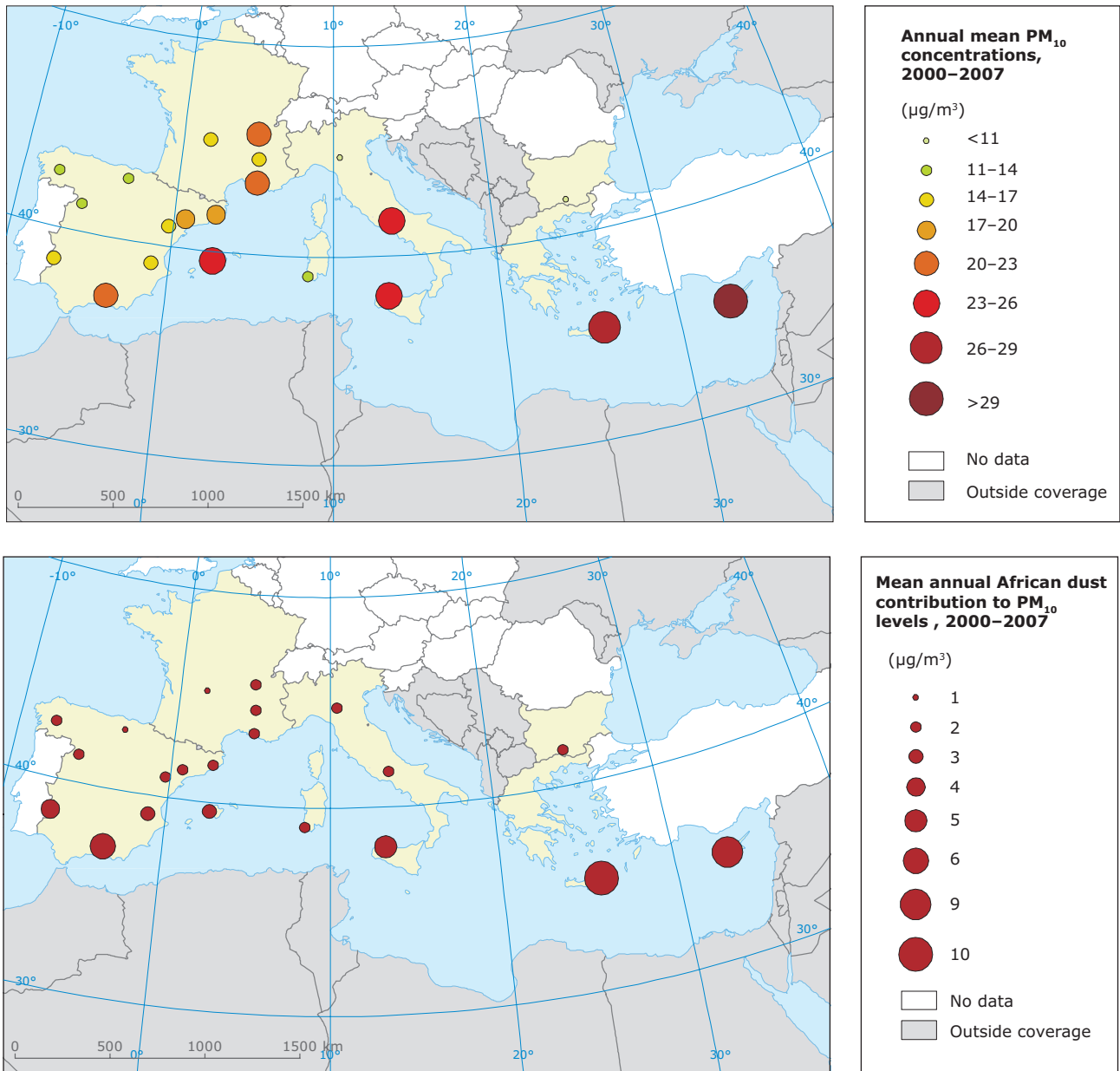
In addition to the sources listed in Box 2.2, regional winds (Harmattan and monsoon winds) can also have a big influence on the chemical composition of resuspended soil dust. The Harmattan winds transport diatomaceous dusts (?) from dry basins westwards, where they mix with particles from the surrounding basement massifs. As a result, dust clouds are enriched with 'hard rock' minerals such as hornblende and several trace elements. In contrast, the summer monsoon dust (carrying certain clay minerals) blows into the Sahara-Sahel dust corridor from sub-Saharan Africa. The chemistry of those particles reflects terrains that are deeply chemically weathered (containing more immobile elements such as zirconium, hafnium and rare earth elements).

Although Africa is the major source of dust, intrusions can also derive from other locations. Using measurement equipment on the CALIPSO (Cloud Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite (see also Box 2.4) it could be shown that in March 2007 a dust cloud emerged during a storm blowing over dry agricultural areas in the Ukraine, known as 'Chernozems' ('black soil' in Russian). Because of drought, the soil was extremely dry and thus sensitive to wind erosion. The event left a clear footprint at PM<sub>10</sub> measurements at stations throughout central and western Europe. Chemical analysis revealed that the PM contained high potassium levels. Potassium is a natural chemical element found in high concentrations in humus of the top soil layers of Chernozems (Bessagnet et al, 2008).

Information on the geochemistry of mineral particulate matter cannot be used as a single tracer of African dust transport. Rather, it has to be combined with other available tools such as

(?) Diatoms are microscopic unicellular algae (most of them are aquatic) with siliceous cell walls.

**Map 2.1 Mean annual  $PM_{10}$  concentrations and net African dust contributions to  $PM_{10}$  levels across the Mediterranean Basin (2000 to 2007 average)**



**Note:** The unit given in the legend (for the dots) is  $\mu\text{g}/\text{m}^3$ . The analysis presented in this figure does not cover Portugal, where contributions to  $PM_{10}$  levels due to African dust events can also be very high.

**Source:** Based on Querol et al., 2009.

meteorological information. In comparison, the '(geo)chemical signature' of pollution episodes due to anthropogenic sources is generally characterised by more carbon and metal components. Compared to natural dust material, those pollutants are more

concentrated in the fine particulate matter fraction,  $PM_{2.5}$ , rather than in  $PM_{10}$  (Allen et al., 2001; Utsunomiya et al., 2004; Birmili et al., 2006).

**Box 2.2 Overview of chemical composition of mineral dust as a function of source regions**

- **Saharan basement massifs**

The particulate matter originating from this source is rich in relatively soluble major elements (sodium, potassium, calcium and magnesium) and contains abundant rare earth elements. Much of this particulate material can be considered as sedimentologically and therefore geochemically immature. This means that it has been derived relatively recently from the weathering and erosion of exposures of certain igneous and metamorphic rocks in the desert (Evans et al., 2004).

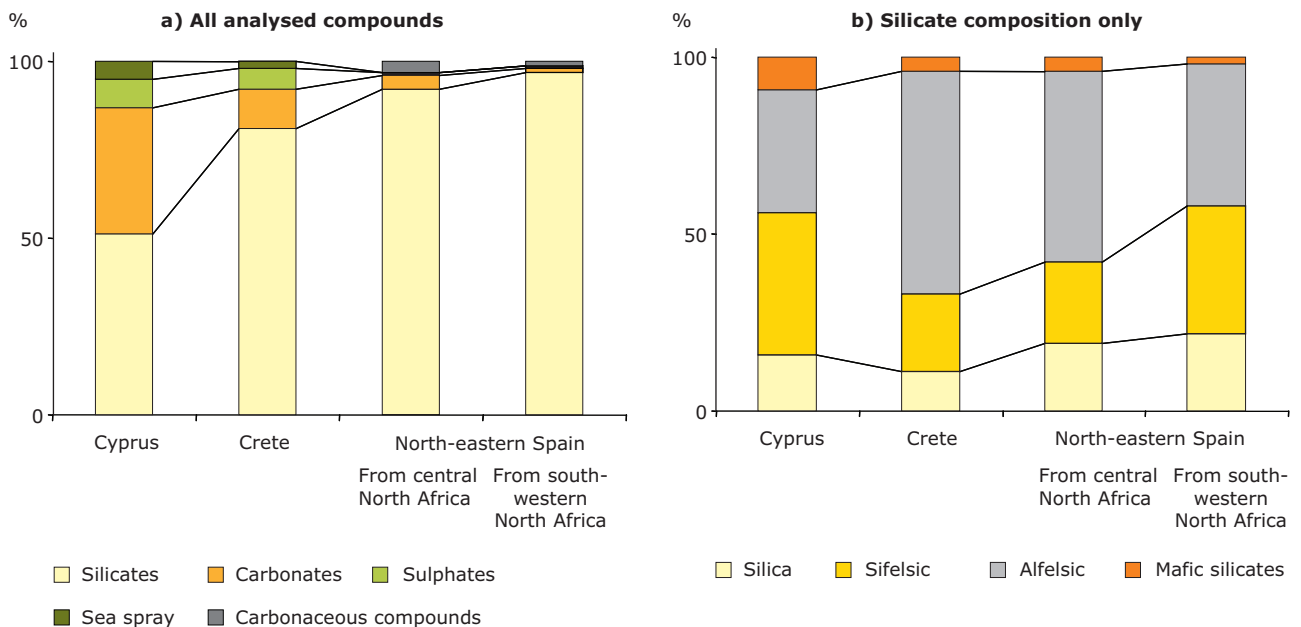
- **Saharan sedimentary basins**

This group of particulates is depleted in primary silicate minerals rich in magnesium and iron. They therefore have a simple composition comprised mostly of clay minerals, quartz and diatoms (silicate minerals enriched with lighter elements such as silicon, oxygen, aluminium, sodium and potassium). Such particles typically have a so-called polycyclic history. This involves a repeated transport by water (rivers, streams) and/or by air and sedimentation that can be traced back millions of years (Evans et al., 2004).

- **Atlantic margin-type particulates**

Many of these aerosols have a geochemistry strongly influenced by the weathering and erosion of marine limestones and marls with origin in the Mesozoic-Cenozoic era (i.e. from about 250 million years ago to the present). Such materials are rich in carbonate (and therefore in calcium, magnesium and strontium). They commonly contain iron oxides, as well as magnesium and iron rich clays.

**Figure 2.1 Chemical composition of African dust analysed in different parts of southern Europe**



**Note:** The analysis is based on scanning electron microscopy (SEM-EDX) PM composition (in terms of the number of particles, not the mass) of PM<sub>10</sub> filter samples collected during north African pollution episodes at locations on Cyprus, Crete and in north-eastern Spain (the latter including air masses coming from central and south-western Africa).

Geochemically, rock-forming silicates can be divided into two main classes: felsic (silica- and aluminium-rich) and mafic (magnesium- and iron-rich) types. Thus, silicates belong in most cases to either felsic varieties (such as quartz, feldspar, white micas and aluminous clay minerals), or mafic species (such as pyroxenes, amphiboles, dark micas, chlorite and their decomposition products, for example secondary chlorite, chlorite-clay mixtures, saponitic smectite, and vermiculite).

**Source:** Based on Querol et al., 2009.



## 2.3 Tools to identify desert dust particulate matter

A number of methodologies are currently used to identify the occurrence of dust outbreaks. Generally, a combination of methods is applied, since using a single approach could lead to misinterpretations or episodes could be overlooked. There are two types of methodologies: those analysing episodes **qualitatively** and those assessing the amount of natural particulate matter affecting an area **quantitatively**.

### 2.3.1 Qualitative analyses of desert dust particulate matter

#### *Interpretation of particulate matter data series together with meteorological information*

One of the common methodologies used to identify natural dust outbreaks is the interpretation of the particulate matter (PM) data series from a given measurement site. In most cases the assessment is supported by a number of meteorological tools (including air-mass back-trajectories and meteorological maps) and local parameters measured at the site such as wind direction and relative humidity (which typically decrease during African dust events) and temperature (which generally increases during these episodes).

Measurements at regional background stations (located far from local sources) are recommended to identify natural dust episodes (Querol et al., 1998; Rodríguez et al., 2001 and 2002; Pérez et al., 2008; Pey et al., 2010). Studies based on measurements at urban or industrial sites are also common but require complementary measurements in order to distinguish natural source contributions from anthropogenic ones. For example, in a typical urban area where road traffic emissions are the main source of particulate matter pollution, peaks of PM are typically concurrent with those of

nitrogen oxides (NO<sub>x</sub>) and carbon monoxide (CO). In industrial environments, it is often possible to link sulphur dioxide (SO<sub>2</sub>) increments to PM peaks. Thus, at such locations the impact of natural dust outbreaks should cause PM peaks unrelated to NO<sub>x</sub>, CO or SO<sub>2</sub> emissions (Viana et al., 2002 and 2003; Moreno et al., 2005 and 2006a).

#### *Remote sensing*

Sun-photometers and other passive remote sensors on satellites are able to provide information on the physical and optical properties of particulate matter in the air. There is a clear increment in aerosol optical depth at all wavelengths when dust plumes are affecting a given site or area (Box 2.3). However, these column-integrated methodologies do not provide information on the vertical structure of a PM plume.

Other satellite data are also used to assess the occurrence of natural dust events (e.g. Moulin et al., 1998). The Sea-viewing Wide Field-of-view Sensor (SeaWiFS, 2012) provides data on quantitative global ocean bio-optical properties and dust plumes visible over oceans (Chou et al., 2002). Similarly, the Moderate Resolution Imaging Spectroradiometer (MODIS, 2012) is a key instrument aboard the Terra and Aqua satellites, viewing the entire Earth's surface every one to two days (Remer et al., 2005). These instruments enable understanding of global dynamics and processes (such as dust plume movements) occurring on land, over the oceans and in the lower atmosphere generally.

Light detection and ranging (LIDAR) technologies (Box 2.4) are increasingly used to analyse the atmosphere, particularly in terms of its vertical resolution. This technique detects natural dust plumes effectively, 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

#### **Box 2.3 Aerosol optical depth and mineral-bearing particulate matter from deserts**

The combination of the increment in aerosol optical depth with the so-called Angström coefficient is generally a useful tool to confirm the existence of dust events. The Angström coefficient is the name of the exponent in a formula 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 relatively large particles (> 2.5 microns) to the atmospheric column. This is a typical indication that mineral-bearing particulate matter transported from desert regions is involved (Lyamani et al., 2005).

**Box 2.4 Light detection and ranging (LIDAR) technology and networks**

LIDAR is an optical remote sensing technology that can measure the distance to or other properties of a target by illuminating the target with light (often using pulses from a laser). Generally, the aerosol backscatter coefficient and extinction coefficient are the most important parameters that can be extracted from LIDAR signals.

The importance of these types of measurements is reflected in the existence of two main global networks on remote sensing: the Aerosol Robotic Network (AERONET, 2012) and the Asian LIDAR Observation Network (NIES, 2012). These networks include both aerosol optical depth measurements and vertical section profiles of aerosols. The European (regional) version of the AERONET network is the European Aerosol Research LIDAR Network (EARLINET, 2012). Its main purpose is to compile an aerosol dataset describing the vertical, horizontal and temporal distribution, including variability at the continental scale. The dataset is also used to validate and improve numerical air quality and climate models that predict the future chemical composition of the atmosphere.

to analyse the bottom 300 meters of the troposphere, which are most relevant when addressing air quality issues.

***Analysis of atmospheric deposition samples***

Atmospheric deposition is the process by which air pollutants are transferred from the atmosphere to the Earth's surface. Evidence of dust outbreaks is clear when red-coloured rain or snow is observed ('wet' deposition). Several studies worldwide characterise these types of episodes (e.g. Bergametti et al., 1989; Schwikowski et al., 1995; Avila et al., 1998; Kubilay et al., 2000). In addition, so-called 'dry' deposition samples are usually collected in regions with little rain and subsequently studied by microscopy in order to identify dust outbreak impacts. In studies performed in Jordan (Abed et al., 2009) and Libya (O'Hara et al., 2006), the researchers found that natural dust particles consist mainly of clay minerals (alumino-silicates), quartz ('sand') and carbonates (calcite and dolomite), with traces of phosphates. The chemical composition reflects the lithology of the north African Sahara.

***Aerobiological studies***

Long-range transport of air masses from desert regions includes not only mineral dust but also other components such as bio-aerosols. A study on the Canary Islands analysed pollen populations according to the origin of different air masses. During African dust outbreaks some species only typical of desert regions were detected (Izquierdo

et al., 2011). Similar findings were observed by Alastuey et al. (2005) during an intense African dust episode, again affecting the Canary Islands. In that study the authors found in the collected PM samples numerous silica skeletons of fresh water diatoms typical of lakes or ponds of northern Africa.

***Radiometric analysis***

Radiometry comprises a set of techniques for measuring radioactive radiation. A number of radiometric elements can be used as tracers of soil dust transport towards a given region. Recent studies on the Canary Islands focused on the analysis of caesium ( $^{137}\text{Cs}$ ), potassium ( $^{40}\text{K}$ ) and Beryllium ( $^7\text{Be}$ ) isotopes (Hernández et al., 2005 and 2008) <sup>(3)</sup>. The authors found a clear indication that the analysed dust originated in Africa. The increment in the activity of these radionuclides, measured in microbecquerel per cubic metre ( $\mu\text{Bq}/\text{m}^3$ ), was up to 14 times above the levels normally found in soil material sampled on the islands.

**2.3.2 Quantitative analysis of desert dust particulate matter*****Statistical analysis***

Escudero et al. (2007) and Pey (2008) have developed a statistical methodology to quantify the daily African dust load in  $\text{PM}_{10}$  samples. The technique, which was developed via a collaboration between Spanish and Portuguese institutions, is described in MARM (2010). This approach is usually applied for

<sup>(3)</sup> Isotopes are variants of a particular chemical element, i.e. atoms of the same element can have different numbers of neutrons.

EU Member State reporting and is recommended by the European Commission (EC, 2011a).

African dust events can be detected by a set of tools including synoptic charts, aerosol maps and back-trajectory analysis. Once the relevant days are identified, this method assumes that during an dust event at a given regional background site  $PM_{10}$  concentrations can be divided into the regional contribution and the dust input. Once the  $PM_{10}$  regional load is determined, the net 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 levels can be obtained by applying a monthly moving 40th percentile to the  $PM_{10}$  time series from a given regional background station, excluding those days coincident with African dust inputs. The 40th percentile method is a good indicator of background concentrations during advective conditions (here the transfer of dust by the horizontal movement of an air mass). 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.

### *Numerical modelling*

In principle, modelling of dust plume evolution and concentrations originating from natural sources can be used to quantify the contribution to particulate matter concentrations in a certain area. In relation to the Air Quality Directive 'in

principle' means that the models have to fulfil the model quality objectives given in the Directive, here for daily mean  $PM_{10}$  concentrations<sup>(4)</sup>. In practice, however, quantifying daily mean  $PM_{10}$  values to the required level of uncertainty (50 %) is difficult. To reduce uncertainties, modelling results have to be applied in combination with monitoring data, which can comprise both satellite and ground-based observations. Mircea et al. (2008) and Mitsakou et al. (2008) represent two relevant examples where models were used to help identify and quantify the contribution of wind-blown dust from the Sahara.

An international project of dust monitoring and forecasting has been established pre-operationally within the framework of GMES (Global Monitoring for Environment and Security; see also Box 2.5). The GMES programme is an EU initiative dedicated to implementing integrated observation and modelling systems to monitor the state of the environment. A component developed in the MACC (Monitoring Atmospheric Composition and Climate) project (MACC, 2012) aims at delivering global and regional (European) forecasts and maps of air pollutant concentrations. Dust in the atmosphere is addressed in the global service with models that assimilate satellite information to provide an evaluation of dust patterns at the global scale on a daily basis. Such a system can be used as an alert or awareness system to inform public and authorities about the occurrence of such dust events, thus also those which are likely to cause exceedances of the daily  $PM_{10}$  limit value.

### **Box 2.5 Sahara dust forecasts**

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 and 2006b);
- the Monterey National Research Laboratories and the Tel-Aviv University Weather Research Center;
- GMES Atmospheric Services through the MACC project (MACC, 2012).

Outputs such as particulate matter concentrations close to the Earth's surface, total atmospheric dust load, dry and wet deposition, and vertical profiles of dust can be obtained from some of these modelling systems.

<sup>(4)</sup> For more details, also concerning the interpretation of 'uncertainty', see the Forum for Air Quality Modelling in Europe (Fairmode) guide (EEA, 2011). Fairmode is a joint response action of the European Environment Agency (EEA) and the European Commission Joint Research Centre (JRC). Its aim is to bring together air quality modellers and users to promote and support the harmonised use of models by EU member countries, with emphasis on their application to the European Air Quality Directive (Fairmode, 2012).

Some but not all regional scale air quality models, such as the French model CHIMERE also contain modules that parameterise wind-blown dust emissions (Vautard et al., 2005). Models commonly used for back trajectory modelling include FLEXTRA and FLEXPART (Stohl et al., 2002; Stohl, 2009) and HYSPLIT (ARL, 2009). Those models have been used for a variety of applications to identify the origin of natural emissions.

### 2.4 Chemical analysis of particulate matter samples

The chemical composition of a collected dust sample can be determined using various methodologies to analyse the mineral matter included in the sample. The most commonly used methods are:

- real-time measurements using automatic instruments such as optical particle counters;
- total digestion of samples collected on filter substrates by means of a reference method and subsequent analysis by different techniques;
- X-ray diffraction and particle-induced x-ray emission.

In many cases, mineral dust originates not only from desert regions but from a mixture of local, regional

and distant sources (Amato et al., 2009). In such cases, the interpretation of chemical elemental ratios can help to relate material to natural dust outbreaks and its sources. Examples include the silicium to aluminium (Si/A) or calcium to iron (Ca/Fe) ratios.

In order to minimise the interference of non-long-range transport sources when defining the typical regional background composition of particulate matter, the use of measurements at respective background monitoring sites is highly recommended. At those locations, most of the dust particles derive from long-range transport (Marenco et al., 2006; Salvador et al., 2007; and Pey et al., 2009a).

#### *Receptor modelling*

A number of studies have identified and quantified the natural dust contributions to particulate matter concentrations by using receptor modelling techniques (see EEA, 2011). In order to carry out such studies a reliable set of data on chemical PM composition is needed. The main aim of receptor modelling activities is to distinguish between different mineral dust sources. One such source is the contribution of long-range transport processes. Examples of receptor modelling applications are summarised by Wang et al. (2007), Nicolás et al. (2008), Koçac et al. (2009) and Viana et al. (2010).

### 3 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 main source of water vapour in the atmosphere. Oceanic surfaces cover 71 % of the Earth's surface and the air-ocean interface is an important source of particulate matter released to the air. For many chemical species in the atmosphere the role of the ocean remains the greatest uncertainty (Fowler et al., 2009). Marine natural particulate matter comprises primary and secondary aerosol components.

Primary marine PM or sea-spray aerosol is a major source of global natural aerosol mass and has an influence on the global climate. Sea-spray aerosol is a combination of inorganic sea salt and organic matter. The main component of sea salt is sodium chloride (NaCl) with traces of magnesium (Mg) and sulphate (SO<sub>4</sub><sup>2-</sup>). The aerosols are emitted due to wind stress at the ocean surface, i.e. via the bubble-bursting processes typically resulting from whitecap generation, leading to the production of film and jet drops. Sea-spray particles range in size from less than one micrometre up to a few micrometres. Sea

spray thus contributes to PM<sub>10</sub> concentrations in the air.

In coastal regions sea salt may contribute up to 80 % to the annual mean particulate mass (PM<sub>10</sub>) (Putaud et al., 2004). By analysing 89 sites in Europe, Manders et al. (2010) found the average concentrations above land ranged between 0.3 and 13 µg/m<sup>3</sup>, with maximum concentrations found along the Irish coastline. Sea salt was a dominant component of particles larger than 0.8 µm at all sites, with a smaller resuspension component associated with frequent precipitation events (Ceburnis et al. 2006).

Since sea salt (mainly NaCl) is quantitatively the major contributor to the marine aerosol mass, it is the component to be considered when evaluating the natural contribution of marine aerosol to PM levels. Sea aerosols in the atmosphere tend to occur as episodic events during strong winds. They are most relevant for the PM<sub>10</sub> daily mean limit values: discounting the sea salt contribution can have an impact on the calculation of the exceedances of the daily limit value for PM<sub>10</sub>.

#### Box 3.1 Biogenic primary sources of marine components emitted to the air

Although the dominant mass fraction of sea-spray aerosol is sea salt, long-term field measurements at Mace Head on the west coast of Ireland<sup>(5)</sup> suggest a significant biogenic primary source of marine organic components (O'Dowd et al., 2004). The water insoluble organic fraction in fine marine aerosol collected during periods of phytoplankton bloom in the North Atlantic was observed to be the most important contribution. The water-insoluble organic fraction, dominating the organic composition in the fine PM size range (< 1 microns), is 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 still remains to be identified (O'Dowd and de Leeuw, 2007). The most relevant marine secondary organic aerosol is methanesulphonic acid, which is derived from marine biologically produced dimethyl sulphide. Some dicarboxylic acids have been associated with secondary formation mechanisms (Kawamura and Sakaguchi, 1999) and a new secondary organic aerosol component produced through the reaction of gaseous amines with sulphuric acid has recently been found in marine aerosol (Facchini et al., 2008; Muller et al., 2009).

<sup>(5)</sup> Located on the west coast of Ireland, the Atmospheric Research Station at Mace Head, Carna, County Galway is unique in Europe. It offers westerly exposure to the North Atlantic Ocean and the opportunity to study atmospheric composition under northern hemispheric background conditions as well as European continental emissions when the winds favour transport from that region (Mace Head, 2012).

**Box 3.2 Sea salt aerosol and climate change interactions**

There is broad interest in sea spray because of its role in several chemical reactions in the atmosphere (e.g. Sørensen et al., 2005; O'Dowd et al., 1999 and 2000) and in particular because of its role in climate change interactions (IPCC, 2007). Sea salt aerosols have two effects on the earth's radiation budget:

- a direct effect in which aerosol particles scatter and absorb the solar and thermal radiation;
- an indirect effect in which they change the particle size and lifetime of cloud droplets acting as cloud condensation nuclei, leading to a cloud albedo change (i.e. the property of clouds to reflect sunlight).

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 areas.

**3.1 Tools to identify sea salt aerosols****3.1.1 Chemical analysis of sea salt aerosols**

One way to calculate the sea salt contribution to atmospheric particulate matter concentrations is based on the average composition of sea water. Sea salt aerosols consist mainly of chloride (c. 55 % by weight) and sodium (c. 31 % by weight). In theory, chloride could be used as a reference ion to calculate the sea salt contribution to particulate matter levels in the air. However, samples can be subjected to chloride depletion due to interaction with nitric acid ( $\text{HNO}_3$ ) or sulphuric acid ( $\text{H}_2\text{SO}_4$ ). They can also be affected by hydrogen chloride (HCl) emitted due to human activities. In contrast, water-soluble sodium could be regarded as a conservative sea salt tracer. It has only a limited number of non-sea salt sources if local dust resuspension is not a significant source, for example due to winter salting of streets (Millero et al., 2004; Manders et al., 2010). However, using sodium may overestimate the natural sea spray mass due to the presence of sodium nitrate ( $\text{NaNO}_3$ ). The question is whether  $\text{NaNO}_3$  is regarded as natural or anthropogenic. In practice, the sodium is likely to come from natural origins (sea spray) and nitrate from anthropogenic origins (nitrogen oxide,  $\text{NO}_x$  oxidation). In its Staff Working Paper (EC, 2011a), the European Commission recommends the chemical determination of both sodium and chloride at each monitoring station at which sea salt contributions are reported.

**3.1.2 Modelling of sea salt aerosol levels**

In addition to chemical analysis of particulate matter, a number of modelling tools are available

to simulate the contribution of sea salt in the lower troposphere (Tsyro et al., 2011). Several studies use modelling applications as the primary source of information for assessing the contribution of sea salt to exceedances of the  $\text{PM}_{10}$  daily and annual limit values (EEA, 2011). Examples from the Netherlands include van Jaarsveld and Klimov (2011), which applied the OPS-ST model, and Manders et al. (2009), which applied the LOTOS-EUROS model to calculate sea salt contributions using a relatively low model resolution of approximately  $6 \times 6 \text{ km}^2$ . Generally, the modelling studies remain quite uncertain on the temporal scale of a single day, with estimated uncertainties in salt concentrations of the factor two to three. However, the long-term average concentrations (over five years) are better represented by the models (van Jaarsveld and Klimov, 2011).

At the European scale the regional air quality model LOTOS-EUROS with a resolution of c.  $10 \times 10 \text{ km}^2$  has been used to simulate sea salt distribution (Schaap et al., 2008). 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 airborne sea salt is represented well by the models, the assessment of absolute concentrations remains a challenge. The main reason is that the balance between sea salt sources and sinks is not yet fully understood. The respective emission parameterisations show considerable variability (O'Dowd and de Leeuw, 2007), with an uncertainty of a factor of two to three. The uncertainty in emissions is due to the different sea salt aerosol source functions used in the models.

## 4 Volcanic dust particles

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, carbon dioxide (CO<sub>2</sub>), sulphur dioxide (SO<sub>2</sub>), hydrogen chloride, fluoride and bromide (HCl, HF and HBr), and a long list of other components emitted in lower abundances (von Glasow et al., 2009). Mercury is amongst the most toxic elements emitted by volcanic eruptions (Ferrara et al., 2000).

The sudden eruption of a volcano has the potential to produce transient peaks in PM<sub>10</sub> levels in EU Member States. Within Europe volcanic activity can mainly be found in certain areas in the Mediterranean area (notably Italy and Greece) and on Iceland. Fine volcanic ash emitted directly from these point sources usually affects nearby rural or urban areas, but will also be spread widely at high atmospheric altitudes (see also Box 4.1). Emissions of gaseous SO<sub>2</sub> from volcanoes contribute to the formation of secondary particulate matter in the air. A recent example in Europe was the eruption of the Eyjafjallajökull glacier on Iceland in April and May 2010 (MACC, 2010). Although the plume reached altitudes of six to seven kilometres, the impact on air quality could be observed at some ground-based measurement stations, e.g. the mountain station Schauinsland in Germany (de Leeuw, 2012). All in all, however, those volcanic emissions had no significant negative effects on air quality in Europe.

### 4.1 Tools to identify volcanic dust particulate matter

Natural contributions from volcano eruptions to ambient atmospheric PM levels can affect the air quality situation for an extended period of time, sometimes weeks. Tracking the development of gaseous tracers such as SO<sub>2</sub> at air quality measurement stations throughout potentially affected Member States is a useful method to follow the impacts of such events in time and space. Another option, which has been investigated and used within the GMES/MACC service (MACC, 2012), is the use of satellite data.

Currently, there is no specific tool available to quantify the effect of volcanic eruptions in local and regional air quality networks. Possible methods are summarised in the Staff Working Paper (EC, 2011a). The tools listed there are based on the comparison of air pollutant concentration levels at the measurement site or in the area under investigation with:

- rural and remote stations in other, non-affected areas;
- levels assessed during periods not affected by the volcanic eruption;
- the analysis of possible plume trajectories, e.g. using satellite data and/or model calculations.

#### Box 4.1 Volcanic activity

Volcanic activity on Earth is focused within active zones of tectonic plate margins. The impact of volcanic ash emissions after an eruption can have a global impact, however, because some of the ash components may be injected into the stratosphere. The stratosphere is the region of the upper atmosphere extending upward from the tropopause (the boundary between troposphere and stratosphere) to about 50 km above the Earth's surface.

One of the clearest examples of volcanic activity's atmospheric effects was the eruption of Mt Pinatubo in the Philippines on 12 June 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<sub>2</sub>. It injected large amounts of aerosols into the stratosphere, more than any eruption since Krakatoa (a volcanic island in Indonesia) 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, and stratospheric ozone depletion temporarily increased substantially (Rantucci, 1994).

The following actions are recommended by the Staff Working Paper (EC, 2011a):

1. Conducting a study of satellite images and back-trajectories (see also Chapter 2) in order to assess the impact of an event in time and space.
2. Using models describing the development of plumes from suspected source areas to demonstrate the relationship between potentially high levels of  $PM_{10}$  and  $SO_2$  and a volcano event, for example by reverting to the GMES pre-operational Atmospheric Service (MACC, 2012).
3. Compiling time series of measurement data from regional background sites, which are relevant for a particular volcano event (in time) and the assessed area (in space). Modelled time series have to be thoroughly validated using measurement data.
4. Identifying peaks in the concentrations of  $PM_{10}$  and  $SO_2$  in long time series, and comparing and assessing whether exceptionally high concentrations can be related to certain volcano events.
5. Comparing the time distribution of relevant volcano events with that of the coincident high  $PM_{10}$  peaks and reviewing 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.
6. Using the average of the  $PM_{10}$  and  $SO_2$  concentrations registered in the 15 days before and 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 the contribution of the volcanic eruptions. If the duration of an event is significant compared to a 30 days 'buffer', a more elaborate scheme might 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 or area can be used if properly justified.
7. The spatial extent of the contribution needs to be explicitly justified through modelling and back trajectories. The spatial representativeness of the measuring station, if determined on the basis of averaged time series, is most probably not adequate for application during a specific event.



## 5 Wild-land fires

Wild-land fire emissions have a significant effect on atmospheric particulate matter concentrations. They usually occur during the summer and in forested areas (e.g. in Portugal in 2005 and in Greece in 2007 and 2009). According to the EU Fire Database (JRC, 2012), an average of 95 000 fires occurred annually in Europe during the period 2000–2005, which resulted in almost 600 000 hectares<sup>(6)</sup> of burnt forestland per year. The emissions from these fires are of special relevance in the Mediterranean countries where summers are drier and hotter compared to other parts of Europe, and where fire outbreaks are commonly fanned by strong winds. Most of these fires occurred in France, Greece, Italy, Portugal and Spain where a combined average of 500 000 hectares of forestland burn every year (Barbosa et al., 2009). Map 5.1 shows the total area burnt in Europe during 2008.

Wild-land fires have a pronounced effect on both local and regional air quality. Emissions include primary air pollutants, i.e. particulate matter (PM), carbon monoxide (CO) and nitrogen oxides (NO<sub>x</sub>). Further, the pollutants ozone (O<sub>3</sub>) and secondary organic aerosol form in the atmosphere when gaseous components, such as non-methane volatile organic compounds (NMVOC) and NO<sub>x</sub>, are released by fires and undergo (photo-)chemical processing (Urbanski et al., 2009).

The European Commission's Joint Research Centre (JRC) estimated that more than 90 % of all fires in the Mediterranean region and 87 % of fires in the boreal region of Russia are actually caused by human activities (JRC, 2007). Analysis of the geographical distribution of forest fires within Member States can help make such an assessment. If, for example, fires occur in particularly humid regions (Figure 5.1), this might point to anthropogenic causes.

It is difficult to identify the causes of wild-land fires. In order to be considered 'natural events', EU legislation requires that they should have 'natural causes', for example lightning. Human activities causing forest fires may include direct ignition (accidental or intentional) but also littering of forest areas with materials with the potential to start a fire or the absence of proper forest

management strategies (thus resulting in forest areas which are uncared for and prone to fires). Correctly identifying and further justifying natural or anthropogenic causes is a key issue for reporting of natural sources to the Commission under the Air Quality Directive.

Finally, it becomes even more difficult to justify natural contributions to exceedances of limit values due to wild-land fires if the pollution is imported from another country.

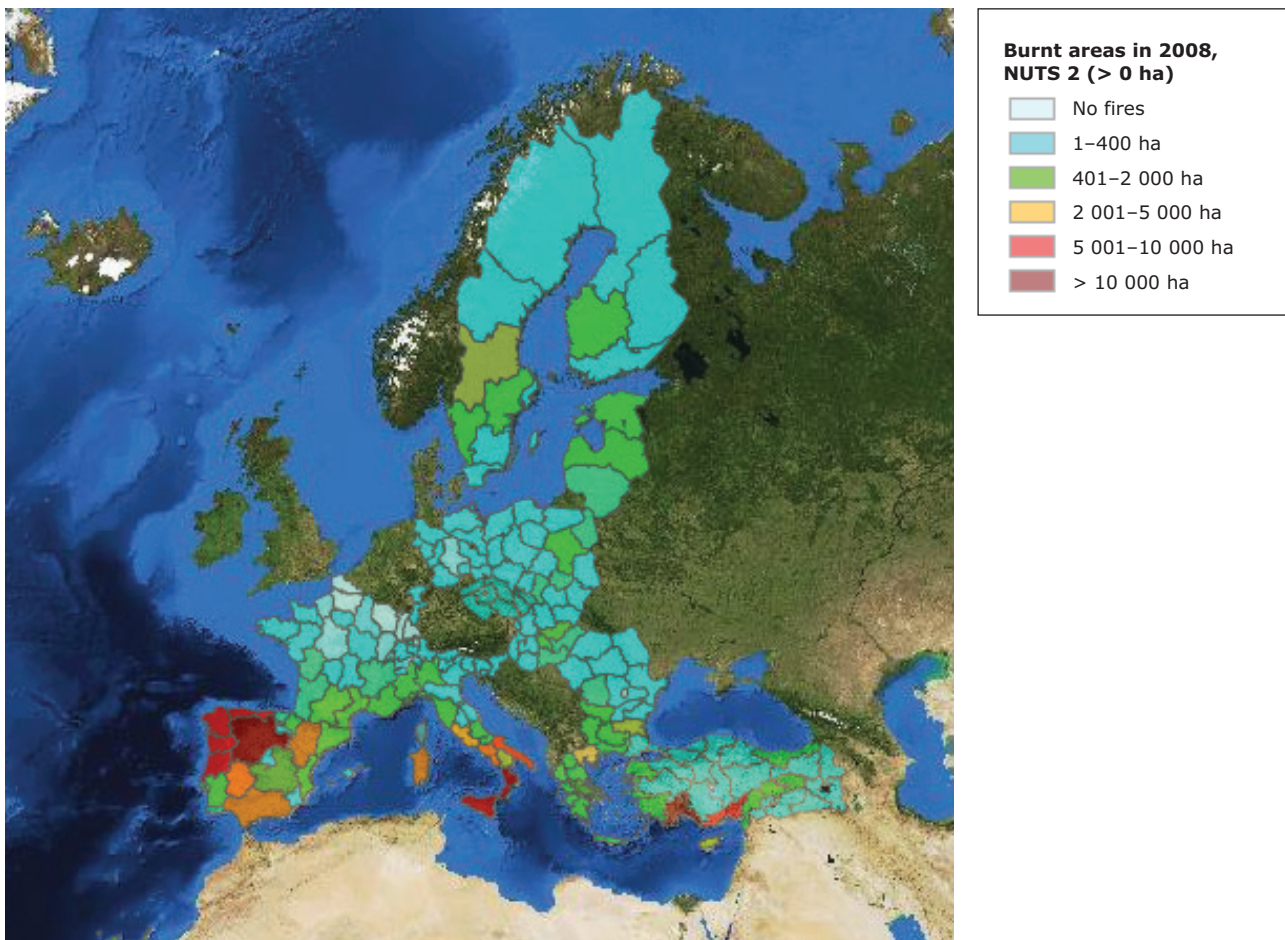
### 5.1 Tools to identify wild-land fire particulate matter

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 (Chapter 4). Member States should only cite this potential contribution if they have evidence regarding the natural origin of the fire. Countries can also report natural contributions to exceedances when fire-related emissions have been transported from regions outside the Member State and if provisions of the Air Quality Directive related to the transboundary pollution have been applied (according to Article 25 'Transboundary air pollution'). The Staff Working Paper (EC, 2011a) states that '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 a 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'. The Staff Working Paper is not clear on whether wild-land fires taking place outside the Member State also need to be natural in origin in order to be considered as a 'natural' contribution.

In addition, if the fire has extended from one Member State to another Member State's territory, any deductions of the common contribution need to be accompanied by the description of short-term measures taken to eliminate the fire and reduce the exposure of the population (EC, 2011a).

<sup>(6)</sup> One hectare is 100 metres x 100 metres = 10 000 square metres (m<sup>2</sup>).

**Map 5.1 Total area burnt in Europe during 2008**



**Note:** No data were available for the areas not shaded.

**Source:** Published with permission of JRC, 2012.

In accordance with the technical reference guide compiled by Fairmode (EEA, 2011), the Staff Working Paper (EC, 2011a) recommends an integrated approach to assess the contribution of wild-land fires. The approach includes the use of:

- geo-referencing the fire occurrence;
- air quality modelling and back trajectories;
- satellite and measurements at ground based monitoring sites;
- chemical analysis of particulate matter.

For geo-referencing the FIRMS Web Firer Mapper can be used to extract the coordinates (FIRMS, 2012).

When air quality models are applied it is important to validate the modelling runs with observed data, either ground-based (terrestrial) or satellite-based.

Best available estimates for wild-land fire emissions are needed: knowledge of the burned area, burning period, fuel (biomass) characteristics, fire behaviour, fuel consumption and pollutant-specific emission factors are required. The information needed can be derived from retrieval algorithms based on earth observation. A methodology has been developed within the GMES/MACC Atmospheric Service projects (Ottmar et al., 2009; MACC, 2012). Global emission rates of various chemical species which are released by wild-land fires can be downloaded (on a daily basis) from the MACC website. They are derived from Fire Radiative Power observations made by the SEVIRI and MODIS instruments, which are provided by the EUMSAT Land Surface Analysis Satellite Applications Facility (LSA SAF; Europe) and the National Aeronautics and Space Administration (NASA; United States of America), respectively, and from high resolution land use data (Kaiser et al., 2012).

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Estimates of emissions at the European scale are available, based on the European Union's fire database (Barbosa et al., 2009). This database contains input provided by individual Member States for each year (see for example Map 5.1). It includes fire event and burned area maps obtained from satellite images, and a map of fuel (biomass) types. In addition to emissions information, some other aspects, such as plume rise of wild-land fires, are currently uncertain. Several recent studies and projects addressing those

fires, particularly in countries such as Finland, France, Greece and Portugal, indicate that contributions to air pollution can be significant (e.g. Miranda, 2004, Hodzic et al., 2007; Miranda et al., 2008).

Even if (improved) emission estimates are available, however, the question remains whether the wild-land fire has a 'natural' origin. Clear guidance on how to determine whether this is the case is so far not available.

## 6 Member State reporting of exceedances of PM<sub>10</sub> limit values due to contributions of natural sources

'Contribution from natural sources shall mean concentration of pollutants resulting from emissions not caused directly or indirectly by human activities' (EC, 2011).

This chapter presents an analysis of the reporting of natural events under Directive 2008/50/EC based on the questionnaires submitted by Member States to the European Commission for the years 2008 and 2009. No Member State reported exceedances of the sulphur dioxide (SO<sub>2</sub>) limit values, which might have been linked to volcanic eruptions or to geothermal activity. This chapter therefore only evaluates the contribution of three main natural sources to exceedances of the daily and annual PM<sub>10</sub> limit values:

- wind-blown desert dust particles (African dust);
- sea spray aerosol particles;
- wild-land fires particles.

Tables 6.1 and 6.2 set out the standard codes for single natural events and their descriptions, as defined in Decision 2004/461/EC. These are natural sources

as specified in Directive 1999/30/EC (EC, 1999). Directive 2008/50/EC added further sources that can be subtracted, including a constant presence of natural particles such as those originating from sea spray.

According to the Staff Working Paper (EC, 2011a), Member States should respect the following 'key principles' to ensure a robust demonstration and quantification of natural contributions:

- the contributions must not be caused by direct or indirect human activities;
- the quantification of the natural contribution must be sufficiently precise;
- the quantification of the natural contribution must be consistent with the averaging period of the limit value;
- the quantification of the natural sources must be spatially attributed;
- the contributions must be demonstrated based on a systematic assessment process;

**Table 6.1 Natural events contributing to limit value exceedances for PM<sub>10</sub> – standard codes**

| 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-fire inside the Member State  |
| D2                 | Wild-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 |

**Note:** These are the codes to be used in forms 21 to 23 in the questionnaire on air quality assessment (EC, 2011b), in relation to Article 20 of the Air Quality Directive ('Contribution from natural sources').

**Source:** Table 5 in Decision 2004/461/EC.

**Table 6.2 Reasons for individual exceedances – standard codes**

| Reason code | Description  |
|-------------|--|
| S1          | Heavily trafficked urban centre  |
| S2          | Proximity to a major road  |
| S3          | Local industry including power production                                    |
| S4          | Quarrying of mining activities   |
| S5          | Domestic heating   |
| S6          | Accidental emission from industrial source                                   |
| S7          | Accidental emission from non-industrial source                               |
| S8          | Natural sources or natural events  |
| 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  |

**Note:** These are the 'reason codes' to be used in form 11 of the Questionnaire. However, form 11 is actually not relevant for Article 20 of the Air Quality Directive ('Contribution from natural sources').

**Source:** Table 2 in Decision 2004/461/EC.

- the quantification of the natural sources must be demonstrated for each pollutant separately.

The questionnaire on air quality assessment, designed to aid reporting under the Air Quality Directive, is a Microsoft Excel workbook with 27 main sheets/forms and several sub-forms (EC, 2011b). The following two forms were used by Member States for reporting on the contribution of natural sources to exceedances of the PM<sub>10</sub> limit values:

- Form 11** (including reason codes) regarding individual exceedances of limit values and limit values plus the margin of tolerance of pollutants listed in the First and Second Daughter Directives. The First Daughter Directive addresses exceedances of the PM<sub>10</sub> limit values.
- Form 23** (including justification codes) regarding exceedances of limit values of PM<sub>10</sub> due to contributions of natural sources.

## 6.1 Member States reporting exceedances due to pollution from natural sources

### 6.1.1 Exceedances of the PM<sub>10</sub> daily limit value

Ten Member States (Austria, Cyprus, France, Germany, Greece, Italy, Malta, Portugal, Spain and the United Kingdom) reported exceedances of the PM<sub>10</sub> daily limit value due to natural contributions

in 2008, and eight in 2009 (Cyprus, France, Greece, Italy, Latvia, Portugal, Spain and the United Kingdom). As shown in Table 6.2, the highest numbers of PM<sub>10</sub> exceedances per station due to such contributions were reported by Mediterranean Member States (Cyprus, France, Greece and Spain, with no data available for Italy for 2009). The lowest numbers per station were reported by Austria, Germany and Latvia.

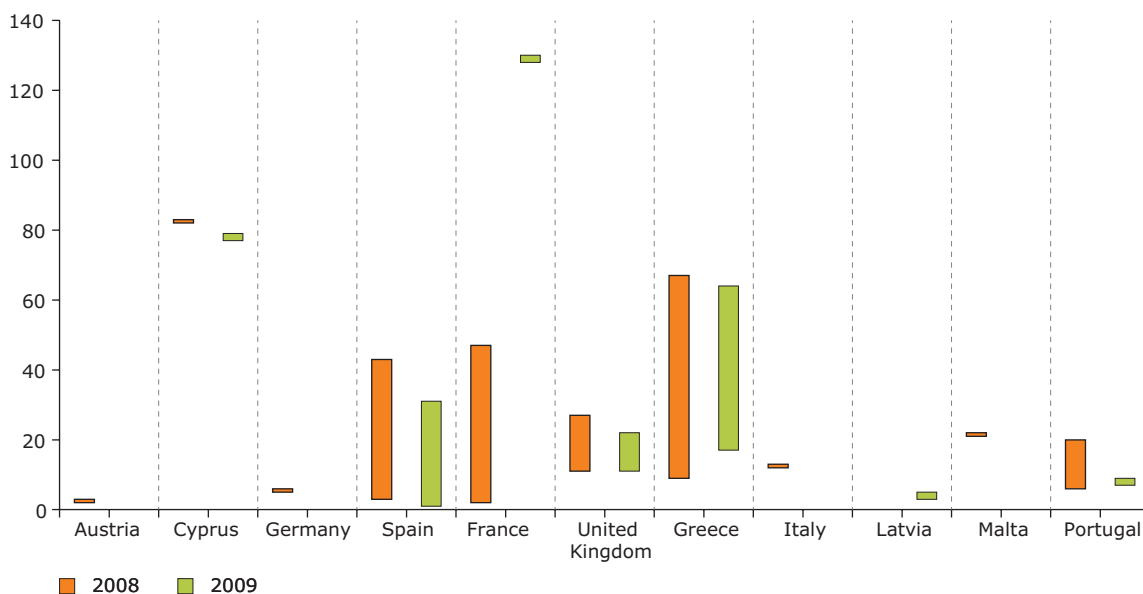
The number of exceedances due to natural contributions reported per station and year is subject to large variability. This is mainly due to year-to-year changes in meteorology controlling natural emission events and the dispersion of natural and anthropogenic air pollutants over Europe.

The main natural contribution to exceedances of the PM<sub>10</sub> daily limit value reported by the different Member States was 'transport of natural particles from dry regions outside the Member State' (code G2 in Table 6.1). This cause was named by 70 % of the Member States reporting natural events in 2008 (Cyprus, France, Germany, Greece, Italy, Portugal and Spain) and 50 % of the Member States in 2009 (Cyprus, Greece, Portugal and Spain).

Exceedances due to the more general term 'natural sources or natural events' (code S8 in Table 6.2) were reported by 30 % of the relevant Member States in 2008 (Austria, Malta and the United Kingdom) and 50 % in 2009 (Italy, Latvia, Spain and the United Kingdom). The analysis of the methodologies

**Figure 6.1 Range of PM<sub>10</sub> exceedances of the daily limit value due to natural contributions reported per Member State in form 11 in 2008 and 2009**

Maximum and minimum number of natural contributions to exceedance/station

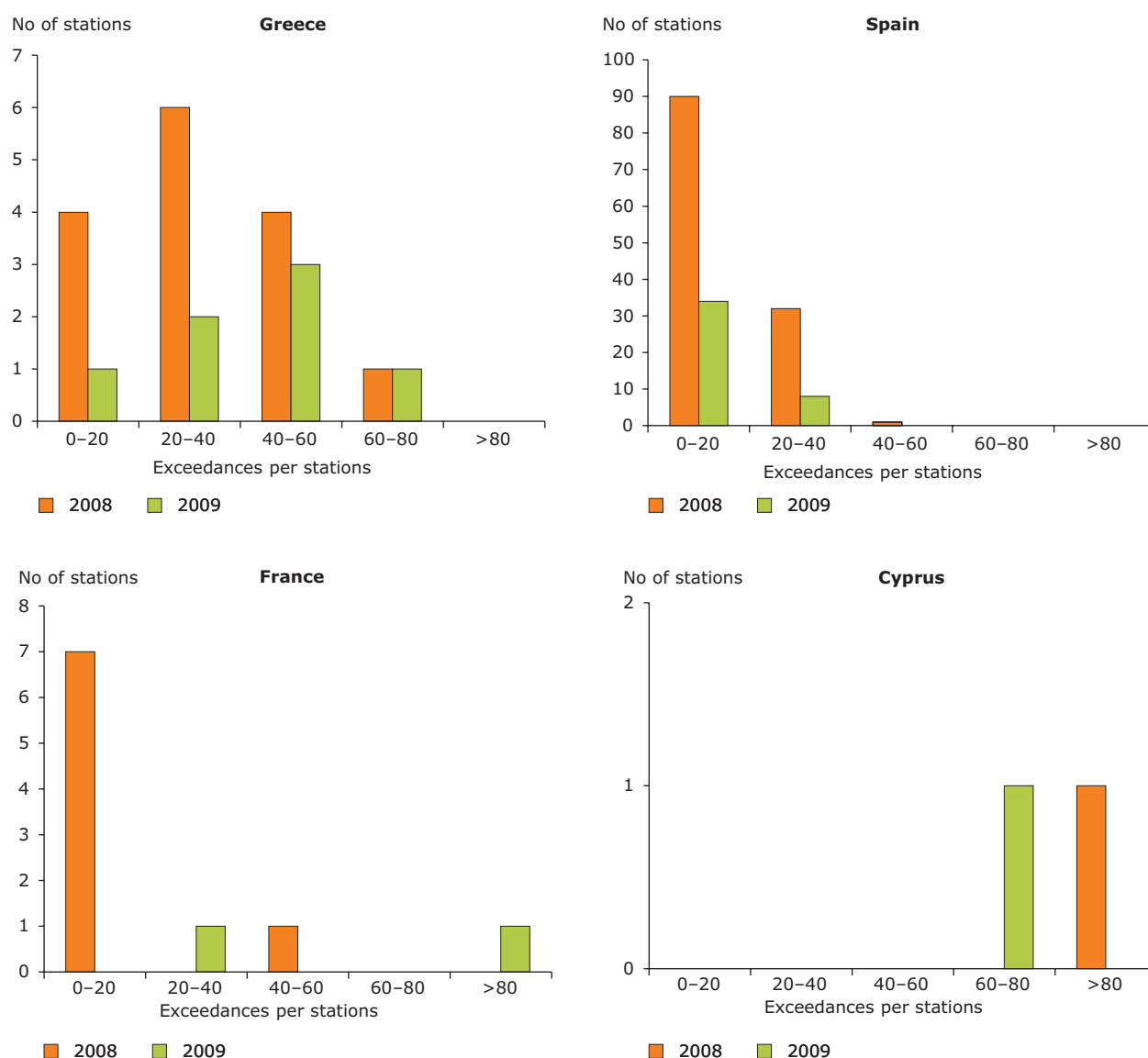

**Source:** Based on Viana et al., 2011.

**Table 6.3 Number of stations where the total PM<sub>10</sub> concentration exceeded the daily and annual limit values before subtracting natural source contributions but not afterwards (2008 and 2009)**

| Member State   | 2008                                 |                                      |                                      |                                      | 2009                                 |                                      |                                      |      |
|----------------|--------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|------|
|                | Daily limit value                    |                                      | Annual limit value                   |                                      | Daily limit value                    |                                      | Annual limit value                   |      |
|                | No of stations/<br>% of total number | No of stations/<br>% of total number | No of stations/<br>% of total number | No of stations/<br>% of total number | No of stations/<br>% of total number | No of stations/<br>% of total number | No of stations/<br>% of total number |      |
| Austria        | 2                                    | 1 %                                  | —                                    | —                                    | —                                    | —                                    | —                                    | —    |
| Cyprus         | 1                                    | 50 %                                 | 1                                    | 50 %                                 | 1                                    | 50 %                                 | 1                                    | 50 % |
| Germany        | 2                                    | < 1 %                                | —                                    | —                                    | —                                    | —                                    | —                                    | —    |
| Spain          | 123                                  | 30 %                                 | 41                                   | 10 %                                 | 42                                   | 10 %                                 | 12                                   | 3 %  |
| France         | 8                                    | 2 %                                  | 3                                    | 1 %                                  | 2                                    | 0.3 %                                | 1                                    | <1 % |
| United Kingdom | 3                                    | 4 %                                  | 1                                    | 1 %                                  | 5                                    | 7 %                                  | —                                    | —    |
| Greece         | 15                                   | 88 %                                 | 12                                   | 71 %                                 | 7                                    | 41 %                                 | 6                                    | 35 % |
| Italy          | 4                                    | 1 %                                  | 1                                    | <1 %                                 | 59                                   | 15 %                                 | —                                    | —    |
| Malta          | 1                                    | 33 %                                 | —                                    | —                                    | —                                    | —                                    | —                                    | —    |
| Portugal       | 3                                    | 6 %                                  | 1                                    | 2 %                                  | 3                                    | 6 %                                  | —                                    | —    |
| Latvia         | —                                    | —                                    | —                                    | —                                    | 3                                    | 33 %                                 | —                                    | —    |

**Note:** The percentage values in the table represent the percentage of the total number of stations reporting PM<sub>10</sub> data in each Member State. For the daily limit values, the stations referred to are those where subtracting the natural source contribution reduced the number of exceedance days from more than 35 to 35 or less.

For Italy no data were available regarding natural contributions to exceedances of the annual limit value in 2009 (form 11h).

**Figure 6.2** Number of exceedances of the PM<sub>10</sub> daily limit value per station and Member State, classified in five ranges

**Source:** Based on Viana et al. 2011.

submitted for reporting suggests that this code referred to African dust transport in all cases, and in addition also to sea salt aerosols in the United Kingdom. Finally, other causes were reported by single Member States: 'wild-land fires inside and outside the Member State' (D1 and D2, by Greece), an unspecified cause (H1) by Greece for 2008 and 2009, and 'embruns marins' (sea salt) by France for both years.

Given the nature of sea-salt contributions, it is somewhat surprising that this type of event was only reported by two Member States (France and the United Kingdom). This could be explained by the fact that this PM source is not included in Table 5 of the

Staff Working Paper (presented as Table 6.1 above). As a result, other Member States may not have listed it, despite being affected by sea spray events.

In almost all Member States where it was applied, the subtraction of natural source contributions reduced the number of stations surpassing the 35 exceedances per year threshold (Table 6.4). The only exception was Latvia (in 2009). For the United Kingdom this is not strictly speaking the number of stations because modelling results were also reported in form 23 for some zones (see note under Table 6.4), in accordance with guidance provided in Decision 2004/461/EC (EC, 2004a).

**Table 6.4** Number of stations reporting exceedances of the PM<sub>10</sub> daily limit values before and after subtraction of natural source contributions

| Member State   | Number of stations reporting exceedances of the daily PM <sub>10</sub> limit value |                   |                    |                   |
|----------------|--|-------------------|--------------------|-------------------|
|                | 2008   |                   | 2009               |                   |
|                | Before subtraction   | After subtraction | Before subtraction | After subtraction |
| Austria        | 10   | 8                 | 6                  | 6                 |
| Cyprus         | 1  | 0                 | 1                  | 1                 |
| Germany        | 17   | 16                | 33                 | 33                |
| Spain          | 123  | 72                | 42                 | 24                |
| France         | 31   | 30                | 41                 | 41                |
| United Kingdom | 3  | 2                 | 5                  | 1                 |
| Greece         | 15   | 11                | 7                  | 2                 |
| Italy          | 188  | 187               | 177                | 177               |
| Latvia         | 3  | 3                 | 3                  | 3                 |
| Malta          | 1  | 0                 | 1                  | 0                 |
| Portugal       | 10   | 7                 | 10                 | 7                 |

**Note:** For the United Kingdom the figures include some exceedances determined by supplementary assessment (modelling). When assessing compliance with PM limit values, the UK always reports the highest determined exceedance, including when that result was calculated by modelling and not based on measurement results obtained at monitoring stations.

Italy did not ask for a subtraction in 2009.

In the other countries, subtracting natural source contributions generally reduced the number of stations exceeding the threshold in 2008 and 2009 by between one and five. In Spain, the natural source correction reduced the number of stations surpassing the threshold by 51 in 2008 and 18 in 2009.

In the case of Italy, a subtraction due to natural sources was not requested in 2009; form 23 of the questionnaire had not been submitted to the Commission.

### 6.1.2 Exceedances of the PM<sub>10</sub> annual limit value

As shown in Table 6.5, the number of Member States reporting exceedances of the PM<sub>10</sub> annual limit value due to natural contributions was lower than for the daily limit value: seven Member States in 2008 (Cyprus, France, Greece, Italy, Portugal, Spain and the United Kingdom) and four in 2009 (Cyprus, France, Greece and Spain). The main reason was probably the low impact of (often episodic) contributions from natural sources on annual mean levels.

**Table 6.5** Number of stations reporting exceedances of the PM<sub>10</sub> annual limit value before and after subtraction of natural contributions

| Member State   | Number of stations reporting exceedances of the PM <sub>10</sub> annual limit value |                   |                    |                   |
|----------------|---|-------------------|--------------------|-------------------|
|                | 2008  |                   | 2009               |                   |
|                | Before subtraction  | After subtraction | Before subtraction | After subtraction |
| Cyprus         | 1   | 0                 | 1                  | 0                 |
| Spain          | 42  | 21                | 12                 | 6                 |
| France         | 5   | 4                 | 7                  | 6                 |
| United Kingdom | 1   | 0                 | -                  | -                 |
| Greece         | 12  | 9                 | 6                  | 0                 |
| Portugal       | 1   | 0                 | -                  | -                 |



**Table 6.6 Reasoning of Member States reporting natural contributions to PM<sub>10</sub> annual concentrations in 2008 and 2009**

| Natural contribution in 2008 |             |                        |                                  |                                       |                                       |
|------------------------------|-------------|------------------------|----------------------------------|---------------------------------------|---------------------------------------|
| Member State                 | No stations | Natural event code(s)  | Annual mean (µg/m <sup>3</sup> ) | Max. annual mean (µg/m <sup>3</sup> ) | Min. annual mean (µg/m <sup>3</sup> ) |
| Cyprus                       | 1           | G2                     | 13                               | 13                                    | 13                                    |
| Spain                        | 41          | G2                     | 4                                | 10                                    | 2                                     |
| France                       | 3           | G2, embruns marins, S8 | 1                                | 1                                     | 0.5                                   |
| United Kingdom               | 1           | S8                     | 5                                | 5                                     | 5                                     |
| Greece                       | 12          | G2;D2                  | 3                                | 7                                     | 1                                     |
| Italy                        | 1           | G2                     | 1                                | 1                                     | 1                                     |
| Portugal                     | 1           | G2, S8                 | 2                                | 2                                     | 2                                     |
| Natural contribution in 2009 |             |                        |                                  |                                       |                                       |
| Cyprus                       | 1           | G2                     | 13                               | 13                                    | 13                                    |
| Spain                        | 12          | G2                     | 1                                | 4                                     | 0.5                                   |
| France                       | 1           | Embruns marins         | 17                               | 17                                    | 17                                    |
| Greece                       | 6           | H1, G2, D1             | 8                                | 9                                     | 6                                     |

**Note:** For the explanation of the natural event codes, please see Tables 6.1 and 6.2. Embruns marins = sea spray.

Mean annual contributions of natural sources to PM<sub>10</sub> levels ranged in 2008 were:

- 1–3 µg/m<sup>3</sup> in Italy, France, Portugal and Greece;
- 4–5 µg/m<sup>3</sup> in Spain and the United Kingdom;
- 13 µg/m<sup>3</sup> on Cyprus.

In 2009, the mean contributions were lower in Spain (1 µg/m<sup>3</sup> versus 4 µg/m<sup>3</sup> in 2008), higher in Greece (8 µg/m<sup>3</sup> versus 3 µg/m<sup>3</sup> in 2008), and the same on Cyprus (13 µg/m<sup>3</sup> in both years). The case of France is not comparable, given that African dust was the natural cause reported for 2008 and sea spray for 2009. With the exception of the United Kingdom, Greece and France (in 2009), the reported natural contributions were linked to the transport of African dust to the Member States.

In all Member State reporting, the subtraction of the mean annual natural contribution resulted in a reduction of the number of stations exceeding the PM<sub>10</sub> annual limit value of 40 µg/m<sup>3</sup> (Table 6.5). In Cyprus, France, Portugal and the United Kingdom, subtraction due to natural contributions reduced the number of stations exceeding the annual limit by one. In Greece, the reduction was three in 2008 and six in 2009. In Spain, the reduction was 21 in 2008 and six in 2009.

## 6.2 Assessment of methodologies applied by the Member States in 2008 and 2009

Member States applied different methodologies to justify natural contributions to particulate matter levels in the air.

1. **African dust:** Portugal, Spain and the United Kingdom (Gibraltar) applied the methodology recommended by the Staff Working Paper (EC, 2011a). Malta used an earlier version of this method, which is mostly comparable to the officially recommended one. Austria, Cyprus, Germany and the United Kingdom applied qualitative methodologies with different degrees of comparability with respect to the method recommended by the Staff Working Paper. France used modelling data issued from PREV'AIR (2012), the national air quality forecasting and mapping platform, which provides, inter alia, global dust maps. No methodologies were submitted by Italy or Latvia for justification for African dust contributions or the unspecified cause S8 (EC, 2004a).
2. **Sea salt aerosols:** None of the Member States applied the methodology recommended by the Staff Working Paper. The United Kingdom

applied a modelling approach. France did not provide specific details on the methodology used for justification.

3. **Wild-land fires:** Contributions from this natural source were reported by Greece. Satellite images were the only tools listed for justification.

### 6.3 Assessment of stations and specific days reported as natural exceedances

Based on information given in questionnaire form 23, in 2009 natural events (mostly African dust episodes) contributed to exceedances of the PM<sub>10</sub> daily limit value for about:

- 30 % of the days reported in Cyprus;
- 10 % of the days reported in Spain;
- 35 % of the days reported in France;
- 7 % of the days reported in the United Kingdom;
- 21 % of the days reported in Greece and Italy;
- 5 % of the days reported in Portugal.

When comparing these results to the data on African dust events in the Mediterranean area published by Querol et al. (2009), the number of exceedances attributed to natural sources appears to be within the expected average in the cases of Cyprus, Greece, Portugal, Spain and the United Kingdom. In addition, the seasonality of the natural contribution to exceedances matches the occurrence of African dust outbreaks (Figure 6.3). Those comparisons can be seen as a confirmation that the justification given by Member States for this type of contribution is reasonable. The number of natural contributions to exceedances of the daily limit value as attributed by Italy was high with respect to the total number of natural events that occurred in 2009. However, Italy did not ask for a subtraction in 2009.

### 6.4 Analysis of the 2008 and 2009 questionnaires: general observations and recommendations

It was not within the scope of this report to provide a coherent overview of the contribution of natural emission sources to air pollutant concentrations or even limit value exceedances in Europe. In general,

the present analysis has shown that the eleven countries that reported natural contributions to PM<sub>10</sub> limit value exceedances used quite different approaches. Further, during natural PM events or episodes, additional Member States than those that did report might have been affected.

It was difficult to identify a common European-wide strategy on how to deal with natural emissions and their contributions to limit value exceedances when analysing the 2008 and 2009 questionnaires and Member State submissions. Certain issues emerged, however, as summarised below. Where feasible in the context of this report, recommendations are presented on how to address these issues.

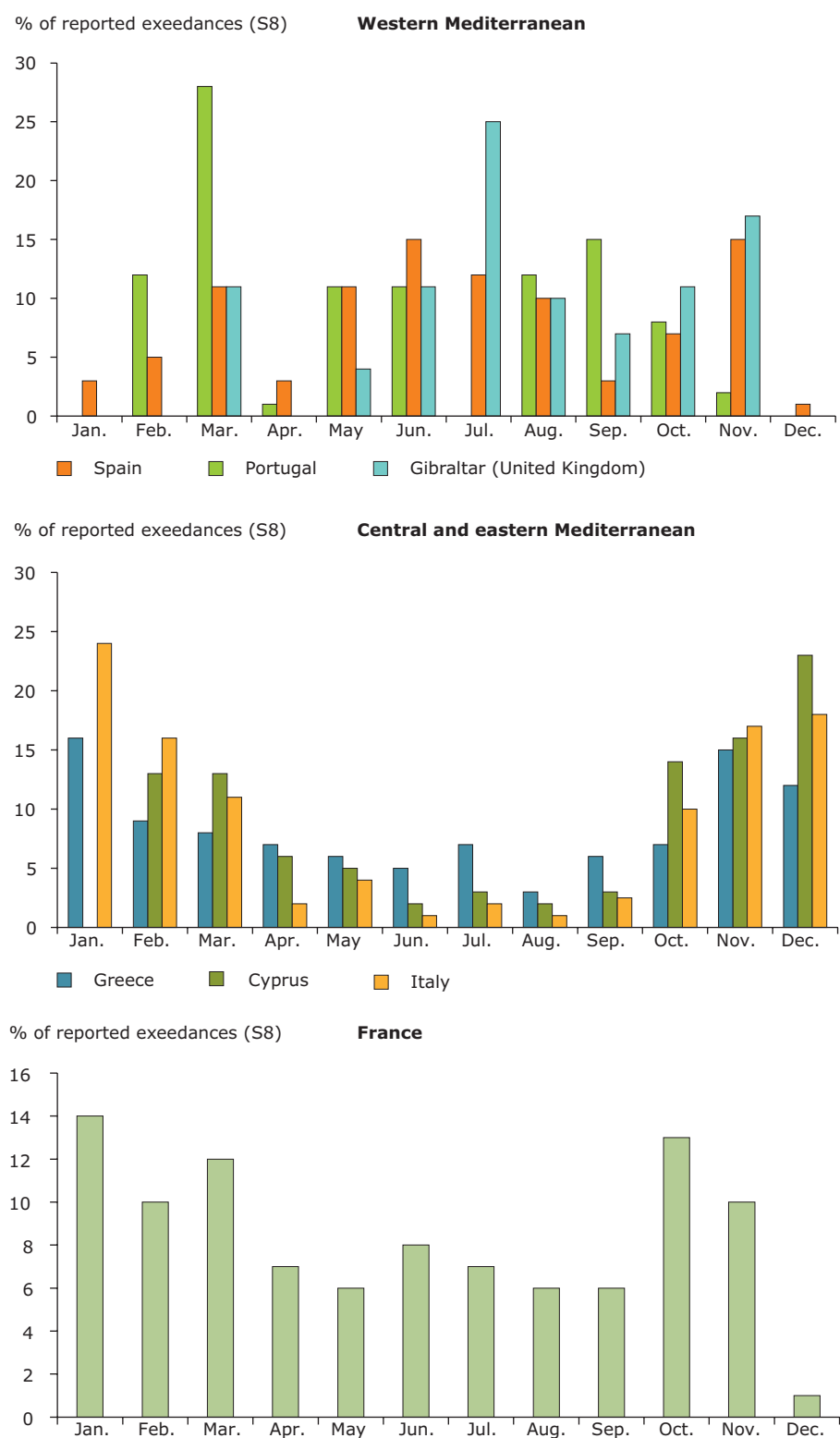
The Commission Implementing Decision (2011/850/EU) on reporting under Directives 2004/10/EC and 2008/50/EC was adopted at the end of 2011 (EC, 2011d). Pursuant to that decision, the questionnaire will be replaced with an electronic reporting mechanism (currently under development). The European Commission is drafting a complementary guidance document. At the moment the draft guidance does not include a code list naming the reasons for the exceedance. This remains to be defined in accordance with the common data type 'exceedance situation' (Annex II of the Decision). As such, the recommendations listed below that address the questionnaire should be considered within the context of this process.

#### 6.4.1 Sea salt aerosols

Contributions from sea-salt aerosols are not included in Tables 5 and 2 of Decision 2004/461/EC (presented as Tables 6.1 and 6.2 in this report) because that decision implements Directive 1999/30/EC. Reporting on sea spray contributions was only included in Directive 2008/50/EC and is thus included as a cause eligible for subtraction in the Staff Working Paper (EC, 2011a). The absence of a standard code to report sea spray as a natural cause means that certain Member States (e.g. France) reported contributions to PM<sub>10</sub> exceedances using a reference ('embruns marins') without a code.

**Recommendation 1: Reporting could be enhanced by including reporting on sea spray events in future revisions of the Staff Working Paper and by creating a standard code for such an entry. This would eliminate the discrepancy that exists currently between the wordings given in Decision 2004/461/EC and the Staff Working Paper.**

**Figure 6.3 Monthly frequency of exceedances of the PM<sub>10</sub> daily limit value due to natural sources (S8)**



**Note:** Top: western Mediterranean Member States (including Gibraltar, United Kingdom).

Middle: central and eastern Mediterranean Member States.

Bottom: France is shown separately because many of the contributions were attributed to sea spray events.

**Source:** Viana et al. 2011.

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 Decision 2004/461/EC).

**Recommendation 2: Reporting could be enhanced by harmonising the 'reasons for exceedance' with the list of 'natural event' codes in form 23 of the questionnaire and Decision 2004/461/EC.**

One option would be to take S8 (natural sources and natural events) out of Table 2 of the Decision (see Table 6.2 of this report) and request the Member States to include the detailed codes for the natural sources from Table 5 (see Table 6.1 of this report). This would make the coding relevant for Article 20 ('Subtraction of natural sources') of Directive 2008/50/EC. The table has to be expanded to include additional sources, including sea spray, when reporting reason codes on form 11. As a minimum table 5 needs to be expanded to include sea spray so that the correct codes can be included in form 23.

**6.4.2 Analysis of the types of natural events reported**

In general, Member States did not always use the codes provided by Decision 2004/461/EC. The main reason is that the coding in the questionnaire is incomplete. France reported exceedances due to 'embruns marins'; Germany used the term 'Saharastaub' for African dust; Malta introduced the codes 'S8a' and 'S8b'; and Greece provided the codes 'H1' and 'H43'. None of these terms are specified in Decision 2004/461/EC.

Certain Member States specified the causes of the natural events and used the codes G2, H1, D1 and D2 (see Table 6.1). Others countries used only the more general term given by code S8 'natural sources and natural events' (Table 6.2). Some Member States reported both natural and non-natural causes. This was the case for Italy in 2009, which provided, in addition to S8 ('natural sources'), codes for anthropogenic sources for all sites: S1, S5 and S3 (see Table 6.2).

Some 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. This was also true for all the days with natural source contributions reported for each station. The one station on Cyprus and all Spanish stations reported that they were affected by G2 (transboundary transport of PM), in 2009 except for one station reporting S8. In other Member States:

- all French stations reported being affected by 'embruns marins';
- all stations in the United Kingdom reported being affected by S8;
- all Greek stations reported being affected by H1, G2 and D1;
- all Latvian stations reported being affected by S8;
- all Polish <sup>(6)</sup> stations reported being affected by G2;
- all Italian stations reported being affected by S1, S5, S3 and S8.

It is in fact unlikely that all stations within a Member State are affected by exactly the same natural sources and during all reported days.

**6.4.3 Assessment of methodologies applied in 2008–2009**

The applied methodologies were explained in the national language by some Member States and in English by others. The language heterogeneity is an initial obstacle when assessing the methods used.

**Recommendation 3: The language issue might be solved by submitting the title of the justification document and a brief summary of the methodologies used in English, in addition to the national language.**

Not all Member States provided a description of the methodology used, or they provided codes not included in Decision 2004/461/EC. Examples are Italy and Cyprus in 2008 and Latvia in 2009.

The majority of Member States referred to one document for justification, with the exception of the

<sup>(6)</sup> Poland reported natural contributions to exceedances (in form 11) in 2009 only but did not request a subtraction (in form 23).

United Kingdom in 2008 and 2009 (referring to one additional document for Gibraltar) and Germany in 2008.

All justification documents were publicly available but while some could be easily retrieved (for example in the cases of the United Kingdom and Germany), others were difficult to trace.

**Recommendation 4: The justification documents could be published on the websites of the respective Member State air quality networks.**

Certain Member States used methodologies not included in the Staff Working Paper to justify contributions due to natural sources or events. It is important to note, however, that the justifications assessed in this report were submitted in 2008 and 2009, whereas the Staff Working Paper was officially published in 2011.

For some Member States reporting exceedances due to the general code S8 (natural events), assessing the methodology used for justification made it possible to identify the type of natural source (for example, sea salt aerosols in the United Kingdom and African dust on Malta). However, this was not the case for other Member States (e.g. Latvia).

**Recommendation 5: If exceedances are reported under 'natural sources or natural events' (code 8), reporting could be enhanced if Member States also specified which natural sources are meant (African dust, sea salt etc.).**

The methodologies for quantifying sea salt contributions presented in the Staff Working Paper are somewhat contradictory. On one hand, the possibility of calculating the sea salt contribution using only sodium or chloride as a tracer is included. On the other hand, the same document discourages the use of chloride as the only tracer, given that it is potentially subject to both positive and negative artefacts.

**Recommendation 6: Reporting could be enhanced if a clear recommendation concerning the use of sodium and/or chloride as sea salt tracers would be provided.**

Possible clarifications hinge also on whether sodium nitrate ( $\text{NaNO}_3$ ) is regarded as 'natural' or anthropogenic. In practice, the sodium is likely to come from natural origins (sea spray) and the nitrate from anthropogenic origins ( $\text{NO}_x$  oxidation).

#### 6.4.4 Comparison between data reported in the questionnaire forms 23 and 11

The codes given in Tables 2 and 5 of Decision 2004/461/EC were used simultaneously for reporting in forms 23a and 23b.

**Recommendation 7: Concerning the future, reporting would be enhanced if the Commission clarified which types of codes (from Tables 2 or 5) should be used for reporting in forms 11h, 23a and 23b of the questionnaire.**

Certain Member States submitted data in form 11 but did not submit the corresponding information in form 23. This procedure is only correct if the 35 exceedances per year threshold, defined for the  $\text{PM}_{10}$  daily limit value, is not surpassed (e.g. in the case of Iceland in 2009)

The number of stations reporting exceedances of the  $\text{PM}_{10}$  daily limit value due to natural events (S8), according to form 11, was compared with the number of stations surpassing the 35 exceedances per year threshold due to natural causes (form 23). Inconsistencies were detected between the two sets of responses. The number of stations in form 11 should be equal to or larger than the number given in form 23, given that in form 23 only stations surpassing 35 annual exceedances are included.

The number may also be larger in form 11 because the form is based on reason codes, which may be assigned qualitatively. Contrastingly, Article 20 of the Air Quality Directive requires that Member States make a quantitative assessment to include stations in form 23 (EC, 2008).

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