SPANISH EMEP ASSESSMENT REPORT

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1. INTRODUCTION

In Spain, the EMEP network started working in the mid-80s, San Pablo de los Montes being the first station. Since then, 15 other stations have been gradually put into operation to reach a suitable national coverage and data series more representative of a background atmospheric pollution network. Nevertheless, the growth of some settlements or the rise in the traffic density in roads near the stations caused the non-fulfilment of the specifications for the EMEP sampling stations description in certain cases. All of those were relocated to more representative sites far from local sources of atmospheric pollution. Some other stations were relocated not due to pollution influence but for administrative readjustments.

This Assessment Report covers the period 1980 to 2000, but the aforementioned relocations have been made since 1995. In general, data before that date suffer from large uncertainties and should be interpreted with care. The names and locations of the 16 Spanish EMEP stations are presented in *Figure 1* and their coordinates are listed in *Table 1*. The ones currently operating are in red.

1: SAN PABLO DE LOS MONTES (Toledo)
2: LA CARTUJA (Granada)
3: ROQUETAS (Tarragona)
4: LOGROÑO (La Rioja)
5: NOIA (A Coruña)
6: MAHÓN (Illes Balears)
7: VÍZNAR (Granada)
8: NIEMBRO-LLANES (Asturias)
9: CAMPISÁBALOS (Guadalajara)
10: CABO DE CREUS (Girona)
11: BARCARROTA (Badajoz)
12: ZARRA (Valencia)
13: PEÑAUSENDE (Zamora)
14: ELS TORMS (Lleida)
15: RISCO LLANO (Toledo)
16: O SAVIÑAO (Lugo)

Figure 1: Spanish EMEP stations

NAME	CODE	Longitude	Latitude	Altitude	Operation period
San Pablo de los Montes	ES1	4º21'07"W	39º22'55"N	917 m.	1984-2000
La Cartuja	ES2	3º36'W	37º12'10"N	800 m.	1987-2000
Roquetas	ES3	0º29'29"E	40º49'14"N	44 m.	1987-2000
Logroño	ES4	2º30'11"W	42º27'28"N	445 m.	1988-2001
Noia	ES5	8º55'25"W	42º43'41"N	683 m.	1992-2000
Mahón	ES6	4º15'E	39º54'00"N	78 m.	1992-1995

NAME	CODE	Longitude	Latitude	Altitude	Operation period
Víznar	ES7	3º28'28"W	37º14'18"N	1.230 m.	1996-*
Niembro-Llanes	ES8	4º51'01"W	43º26'32"N	134 m.	1999-*
Campisábalos	ES9	3º08'34"W	41º16'52"N	1.360 m.	1998-*
Cabo de Creus	ES10	3º19'01"E	42º19'10"N	23 m.	1999-*
Barcarrota	ES11	6º55'22"W	38º28'33"N	393 m.	1999-*
Zarra	ES12	1º06'07"W	39º05'10"N	885 m.	1999-*
Peñausende	ES13	5º52'1"W	41º17'20"N	985 m.	2000-*
Els Torms	ES14	0º43'16"E	41º23'42"N	470 m.	2000-*
Risco Llano	ES15	4º21'09"W	39º31'22"N	1241 m.	2000-*
O Saviñao	ES16	7º42'17"W	42º38'05"N	506 m.	2001-*

Table 1: Spanish EMEP stations coordinates
: Currently in operation

The Spanish measurement programme is summarised in *Table 2*.

Gas	SO_2, NO_2, NO_x, O_3 (**hourly**), VOCs (only at Campisábalos, **twice a week**)
Aerosols	SO_4^{2-}, PM_{10}, $PM_{2.5}$ (**daily**) (TSP until 1st February 2003) HM (only at Campisábalos and Niembro, **once a week**)
Gas + Aerosols	$HNO_3 + NO_3^-$, $NH_3 + NH_4^+$ (**daily**)
Precipitation	pH/H^+, SO_4^{2-}, NO_3^-, NH_4^+, Ca^{2+}, K^+, Cl^-, Na^+, Mg^{2+}, conductivity (**daily**)

Table 2: Present EMEP measurement programme in Spain.

Since the beginning, SO_2 and NO_2 had been determined by manual methods, with a sampling period of 24 hours, being later analysed in the laboratory. To improve the quality of the measurements, several continuous monitors were installed in the stations (for the hourly determination of SO_2, NO_2, NO_x and O_3). After the field comparison carried out by NILU in Zarra, it was proved that SO_2 and NO_2 manual data were poorly representative if they were compared with continuous ones, so the use of SO_2 and NO_2 data monitored with automatic monitors was adopted. This equipment has a much better sensibility than manual methods and allows the assessment of the representativeness of the station location site. On the other hand, it also allows the continue monitoring of these pollutants, according to current Directives.

The Spanish EMEP Assessment Report will present information about SO_2-S, SO_4-S, wet deposition of sulphate, NO_2-N, wet deposition of nitrate, ozone, TSP and pH.

2. EMISSIONS. CONCENTRATIONS. TRENDS

2.1. SO_2 – S

In *Figure 2*, an important reduction in the SO_2-S concentrations through the last decade can be appreciated in all the Spanish stations. The reason for this can be associated with the reduction in SO_2 emissions, both in Spain and in the rest of Europe, due to the application of the Helsinki Protocol of 1985. In Spain all energy activities have contributed to this reduction. More than half of the decrease can be attributed to the introduction of fuel gas desulphurisation (FGD) and the use of lower-sulphur coal and oil. The remainder is mainly due to changes in electricity production including fuel switching (mainly from coal and lignite to natural gas) and improved efficiency, a small part due to the growth in production from renewable sources, included hydro-power. Renewable energies, particularly wind energy, have followed the process of incorporation into the Spanish market, trying to maintain the objectives of the *Renewable Energy Promotion Plan* (12% of primary energy in 2010 from renewable sources).

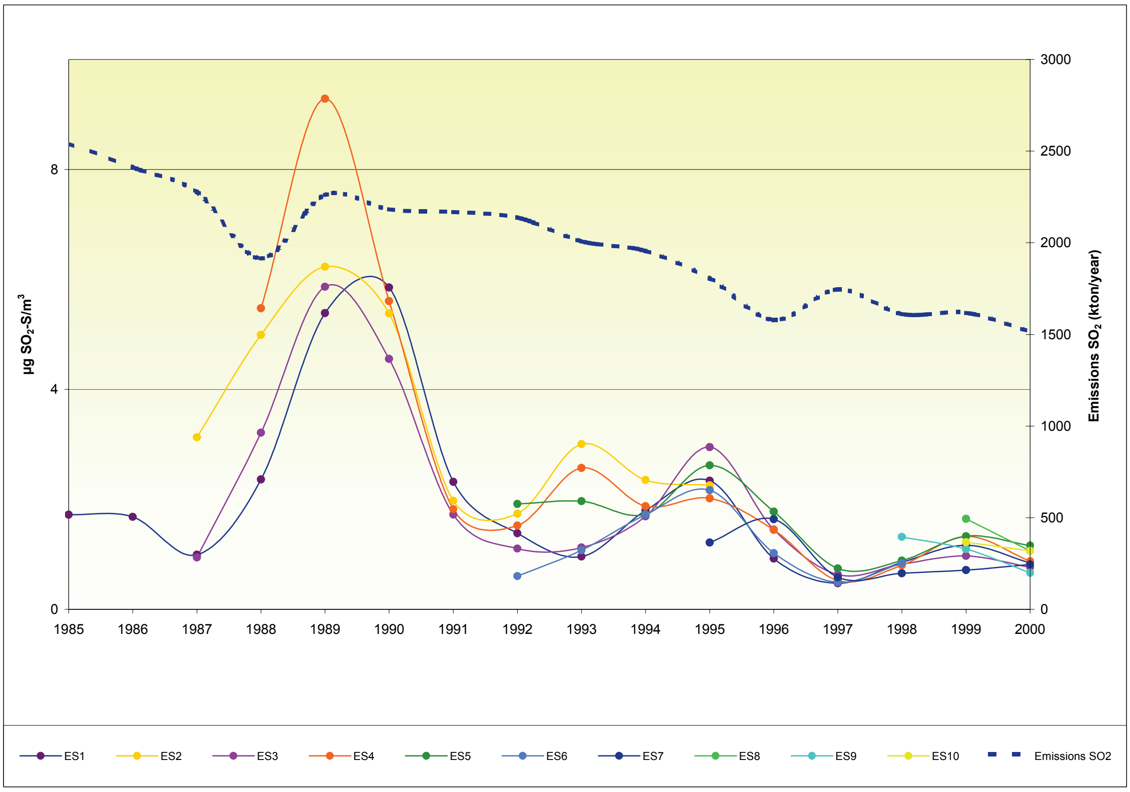

Figure 2: SO_2 annual values (1985-2000). Concentration (µg S/m^3) vs. emission data (kton/year).

The highest values of SO_2-S/m^3 were usually measured at the Noia station because, although annual data were not very high, there were occasionally extreme daily values. The station was located inside a military site, and this made it impossible to have information about the pollutant sources. This was the reason for changing the location of the station. The values were also high at Roquetas, probably due to local pollutant contributions derived from agricultural activities. On the other hand, Niembro had a regular behaviour throughout the year. This station is located in the North of Spain, which is a highly industrial area. It is also a cold area that shows an increase in SO_2 concentration due to the use of the coal heating in wintertime. Víznar shows a lower annual average, without particular contributions of SO_2.

Maximum concentrations in the late 80s may have their origin in meteorological conditions, possibly due to some dry periods with the consequently high-pressure situations that hindered the dispersion of atmospheric pollutants.

During the last years, the concentrations of SO_2-S/m^3 can be considered to be quite low and at the same level as other EMEP countries, although the reduction trend is not statistically significant.

As mentioned before, the field comparison made by NILU at Zarra demonstrated the higher reliability of monitor data against the daily manual data. It is interesting to note, in *Figure 3*, SO_2 monitor data (1998-2001) from 4 locations that show the low concentrations currently registered in Spanish stations.

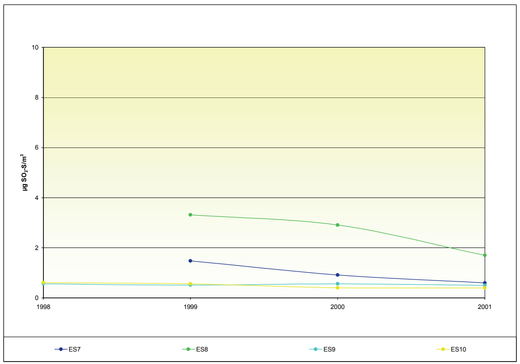

Figure 3: SO_2 monitor data (1998-2001) in µg S/m^3.

2.2. SO_4 – S

The annual mean concentrations of particulate sulphate have also dropped (*Figure 4*), although the reduction is smaller than the one for gaseous SO_2. This is probably due to the fact that particulate sulphate may have a primary origin (marine aerosol) or a secondary origin (from the oxidation of SO_2), so the reduction in anthropogenic emissions of SO_2 does not correspond to the reduction in sulphates. With the exception of the Cabo de Creus station, in the regions where the other stations are placed secondary sulphate is around 90% of total sulphate. However, the sources of ammonium sulphate may have been in very distant areas, and long range transport may account for an important proportion of the measured sulphate. This may explain the slightly different inter-annual trends observed for sulphate and sulphur dioxide.

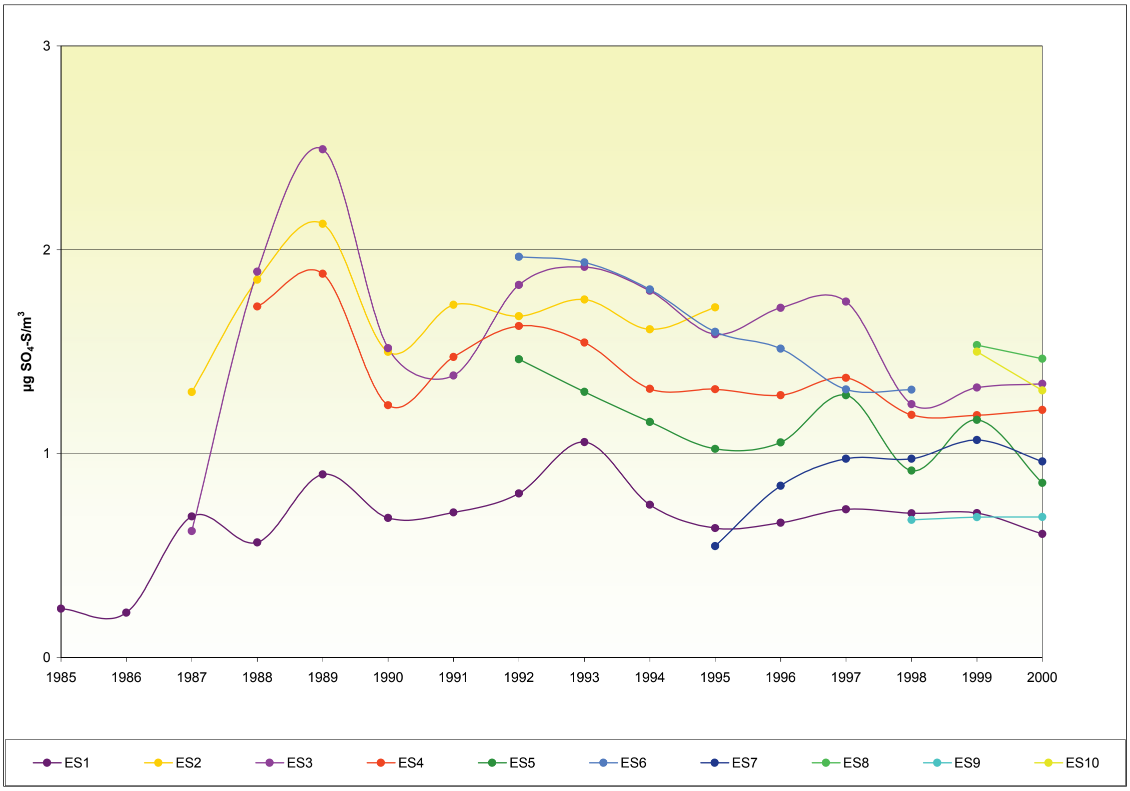

Figure 4: SO_4 annual values (1985-2000) in µg S/m^3.

It must be pointed out that the levels of sulphate measured at San Pablo de los Montes are lower by 1 µg/m^3, when compared to the other stations. This is probably due to the lower industrial emissions in the central peninsular area with respect to some coastal regions.

2.3. Wet deposition of sulphate

As it can be seen in *Figure 5*, the decreases both in SO_2-S and in airborne sulphate concentrations were not translated into a decrease in wet deposition of sulphate, as it has maintained a steady value in an almost constant band between 100 and 500 mg S/m^2 for most stations. Data from some stations very close to the sea are not included due to the influence of marine aerosols (see below item 2.7).

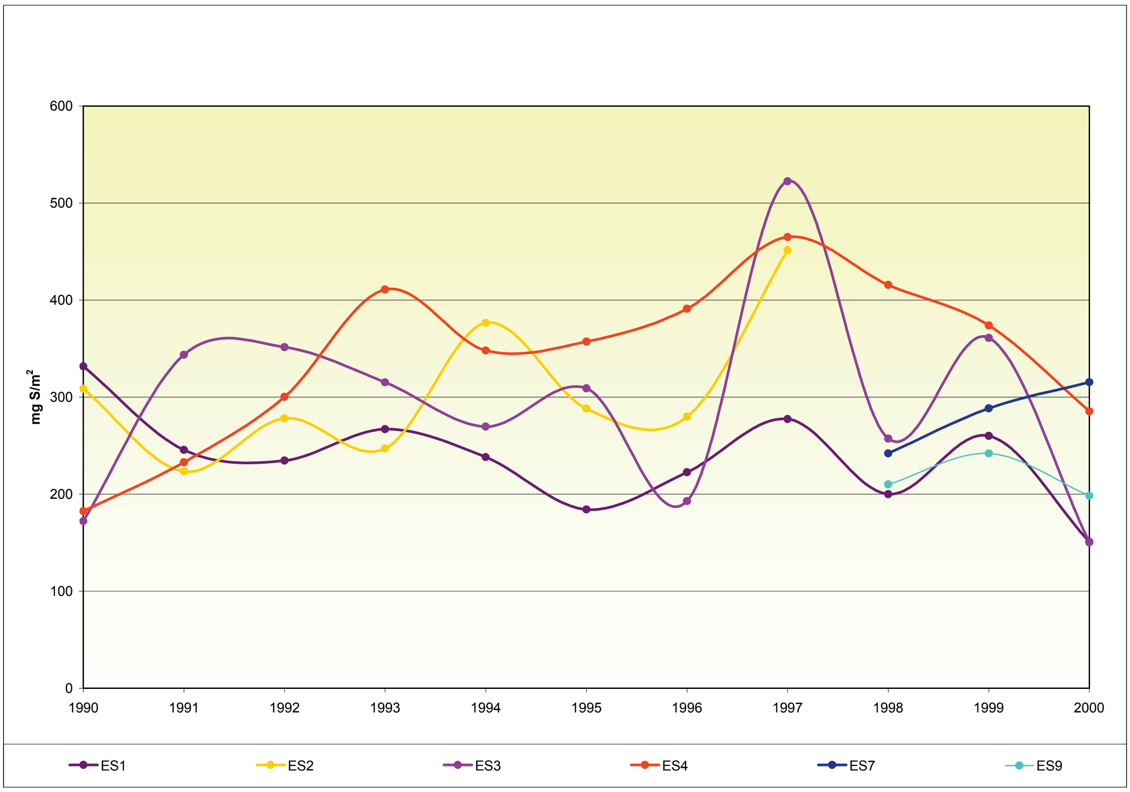

Figure 5 – Annual values of sulphate in wet deposition (1990–2000) in mg S/m^2 per year

2.4. NO_2 – N

Figure 6 shows an increment of NO_2 emissions despite the reduction policies adopted; this may be due to the important growth in the amount of cars in Spain.

As for concentration levels, we can see a great fluctuation through the years. Top values were measured in the middle 90s, probably due to the drought that took place from 1990 to 1995. The following sharp decrease has no clear explanation, but it occurs in 1996, a year with a total precipitation much higher than average.

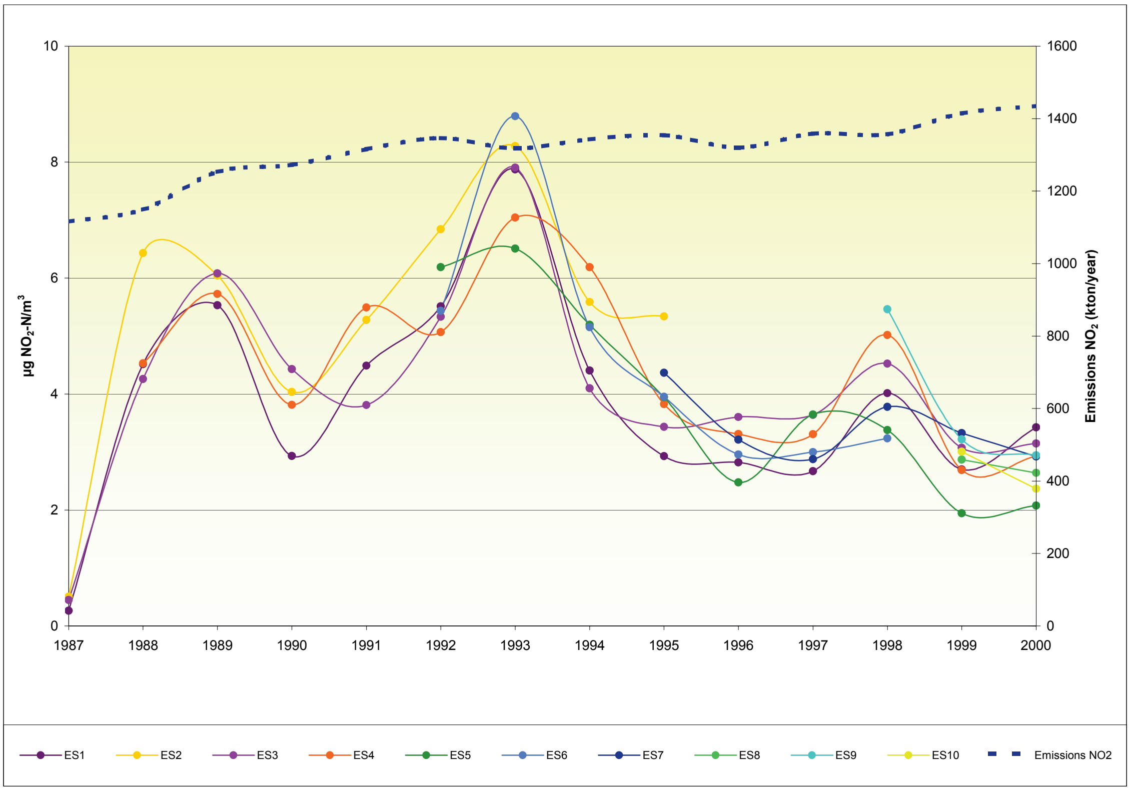

Figure 6: NO_2 annual values (1987-2000). Concentration (µg N/m^3) vs. emission data (kton/year).

Finally, since 1998-1999, there is a stabilisation of the concentrations (showed in *Figure 7*, where monitor data for NO_2 from 4 Spanish stations are plotted).

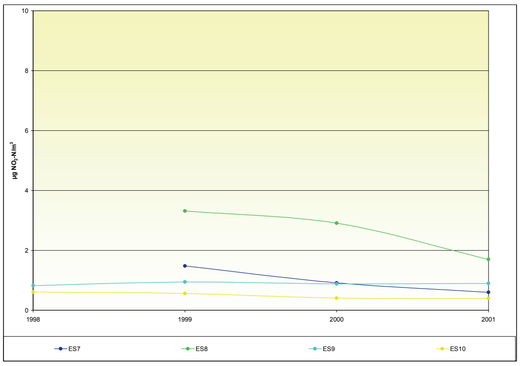

Figure 7: NO_2 monitor data (1998-2001) in µg N/m^3.

The highest daily levels of NO_2-N measured in some EMEP stations can be explained by their proximity to high traffic density roads (this is the case of Logroño) or to cities (the case of La Cartuja and Roquetas). This is the reason why they were relocated.

2.5. Wet deposition of nitrate

In *Figure 8* and, as it was the case of sulphate, no clear trend for wet deposition of nitrate can be found (here again the stations influenced by the marine aerosols are not considered).

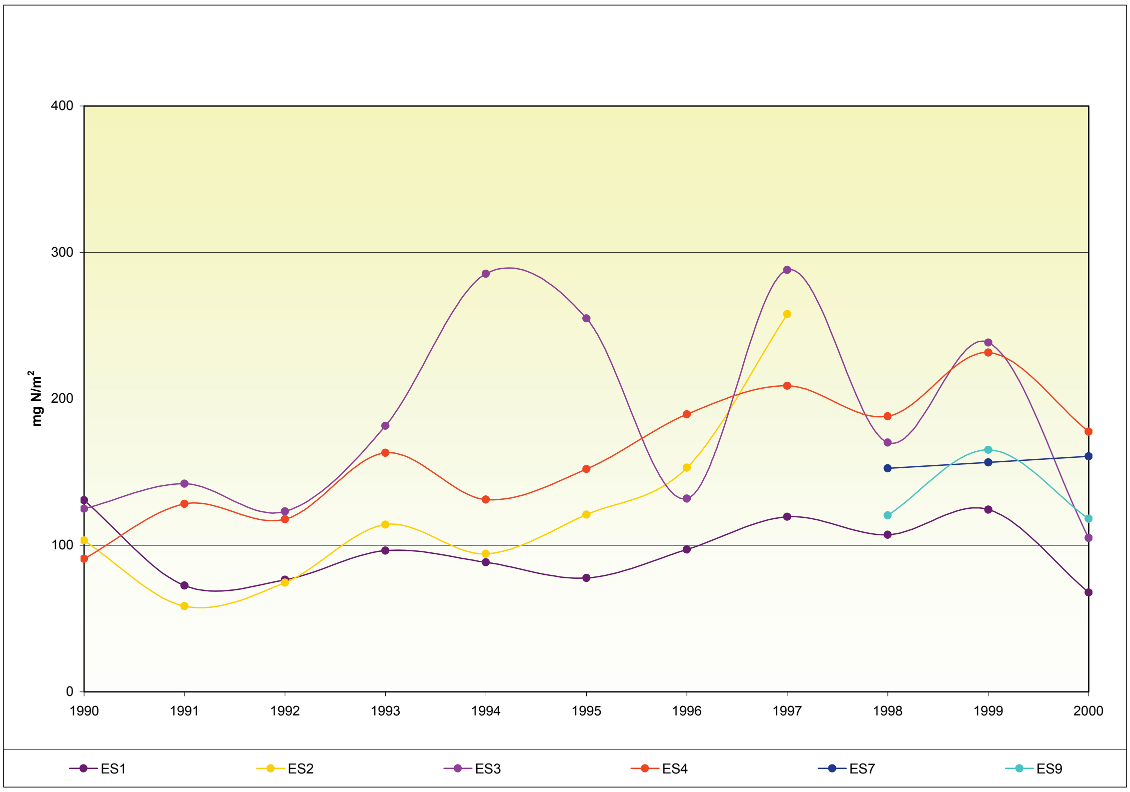

Figure 8 – Annual values of nitrate in wet deposition (1990–2000) in mg N/m^2 *per year*

2.6. O_3

Ground-level ozone is one of the most urgent problems regarding atmospheric pollution in Spanish EMEP stations. This is due, on the one hand, to the large number of sunshine hours and, on the other, to the great concentration of primary precursors (NO_X, VOC).

The following figures (*Figure 9* and *Figure 10*) illustrate the AOT40 values, the AOT40 critical levels for the protection of forests (20.000 $\mu g/m^3 \cdot h$) and for the protection of vegetation (6.000 $\mu g/m^3 \cdot h$). These figures show that all the Spanish stations (except Niembro) exceed these values. Niembro is one of the least sunny stations in Spain and has an oceanic climate with frequent rains. On the other hand, the maximum concentrations are measured in Víznar and Cabo de Creus.

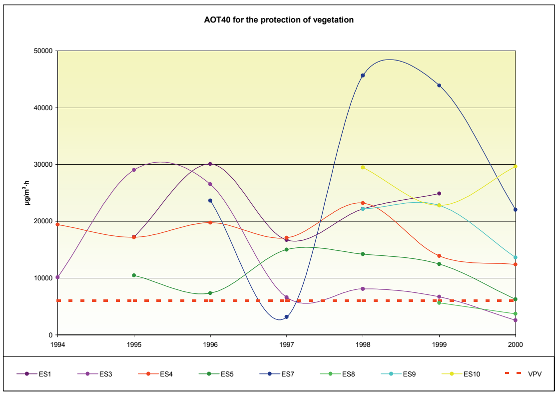

Figure 9: AOT40 for the vegetation protection in $\mu g/m^3 \cdot h$ *(May- July).*

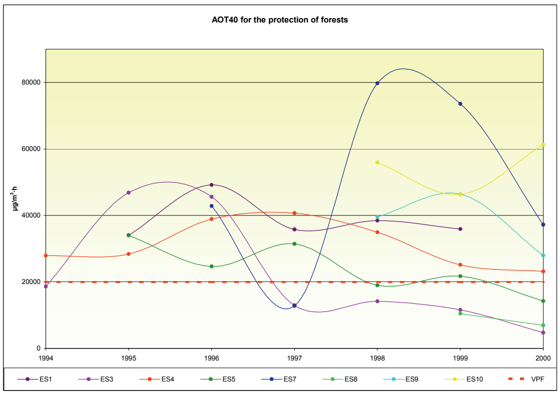

Figure 10: AOT40 for the forest protection in $\mu g/m^3 \cdot h$ *(April- September).*

2.7. TSP

(TSP were measured from 1986 until February 1st 2003; PM_{10} and $PM_{2.5}$ are measured in every Spanish station from March 1st 2001)

Background TSP levels in Spain (*Figure 11*) are relatively high when compared to those recorded in central and northern EU. This is attributed to the following major causes:

a) Mineral dust input from the transport of African air masses;
b) Mineral dust re-suspension from poorly developed soils in relatively semi-arid zones;
c) Low rainfall with the consequent reduction of the PM scavenging potential;
d) Development of regional air mass aging events caused by continuous re-circulation of air masses, typically occurring from spring to summer over the Iberian Peninsula.

Furthermore, in coastal stations very close to the shoreline an important marine aerosol contribution may be found.

The highest daily TSP levels are recorded at La Cartuja, Víznar and Cabo de Creus. In all these cases this is due to natural inputs: in the first two stations, the African dust outbreaks and, in Cabo de Creus, the marine aerosols. This is proved by the rate TSP/PM_{10} recorded (according to 2001 and 2002 data) for Víznar and Cabo de Creus stations, much higher than elsewhere. However, in Víznar, in the last years, there is an annual increasing trend that may be caused by the emissions from a nearby highway recently constructed.

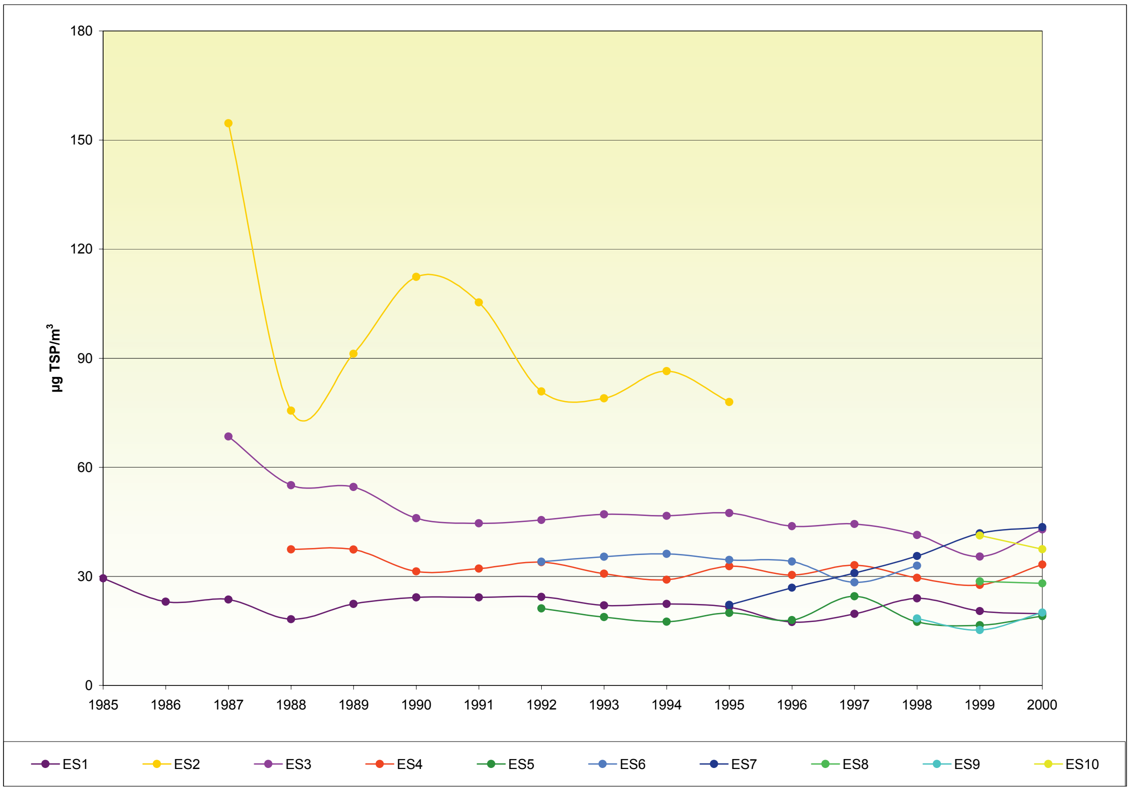

Figure 11: TSP annual concentrations (1985-2000) (µg/m³).

2.8. pH

The pH of rain is considered neutral at 5.6. Below this value precipitation is considered acid rain. In general, Spanish EMEP stations do not have an acid rain problem, as shown in *Figure 12*, which represents precipitation pH monthly mean values. The behaviour of Noia and Niembro differs from the other stations. Both stations are located in the North of Spain and have many monthly mean values below 5.6. The remaining stations have values between 5 and 7 and those values below 5 are either episodic values that probably have their origin in the North of Europe (which happens twice a year) or values measured with a low precipitation volume (which makes the sample very concentrated and not very representative). On the contrary, Spain suffers occasionally from basic rains ("red rains"), events associated with southern winds.

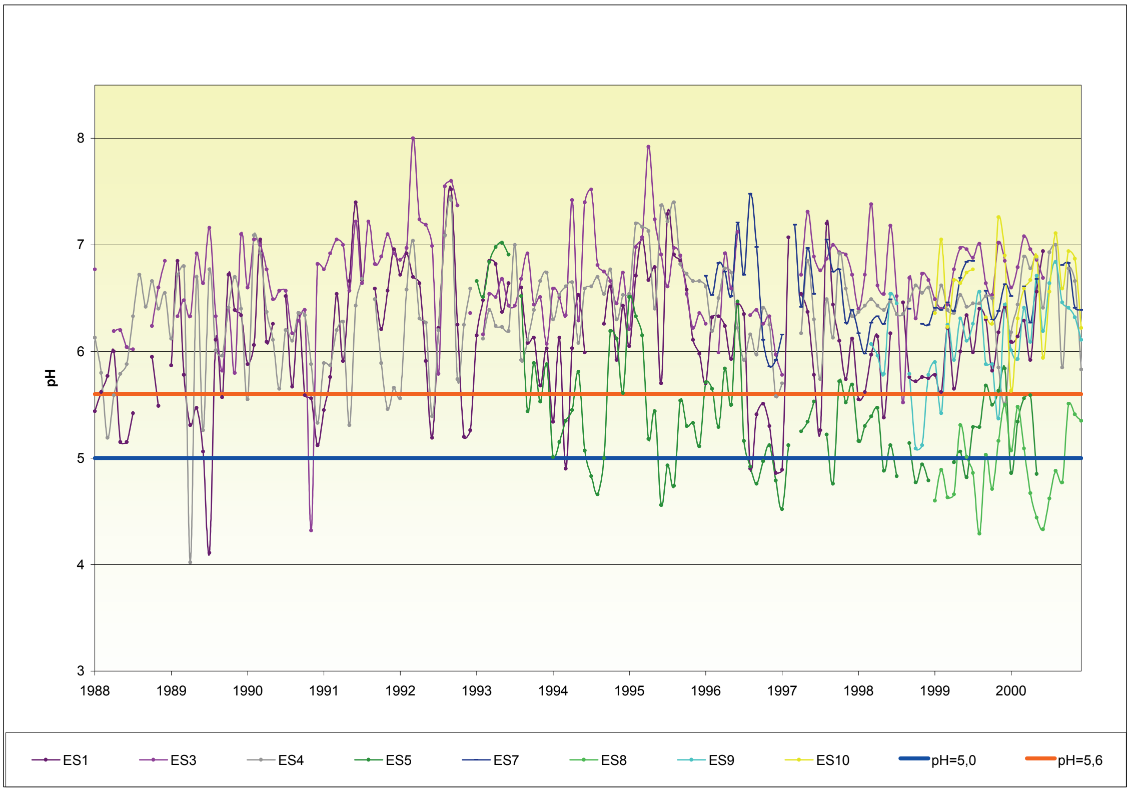

Figure 12: Precipitation pH monthly mean values.

2.9. Wind sector analysis

This section aims to provide some examples of wind sectors analysis to prove wind direction has, generally, no influence on pollutants concentrations. In general, only data for one station, ES1, San Pablo de los Montes, are represented, as all the other stations showed similar patterns. So, in *Figure 13*, SO_2 1985-1996 average annual mean concentrations by sectors are represented for ES1 station. The form of the figure shows that there is no influence of wind direction and the contribution of SO_2 comes from all wind directions. The higher contribution from the South is counteracted by the little frequency of the wind from that direction (*Figure 14*).

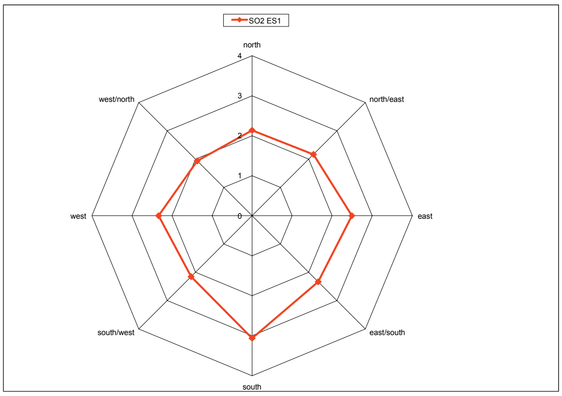

Figure 13: SO_2 concentrations by sectors (ES1), (1985-1996).

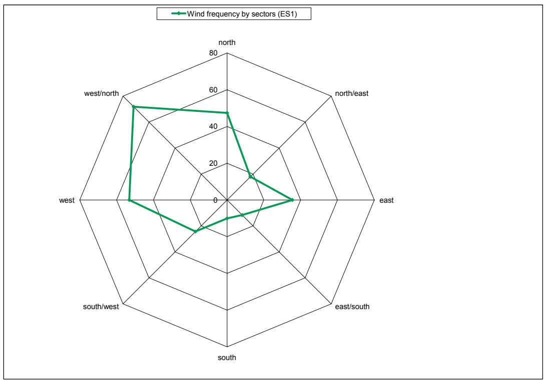

Figure 14: Wind frequency by sectors (ES1), (1985-1996).

Figure 15 and *Figure 16* show similar outputs for SO_4 concentrations and NO_2.

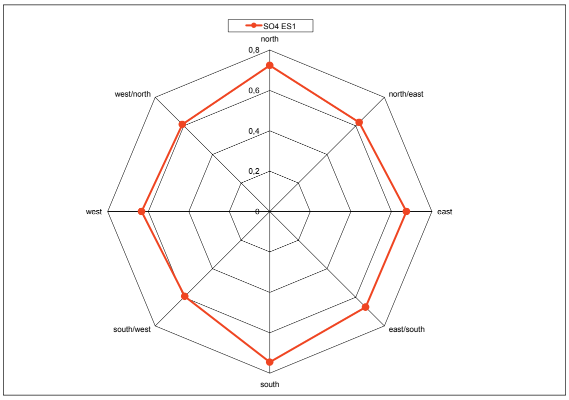

Figure 15: SO_4 concentrations by sectors (ES1), (1985-1996)

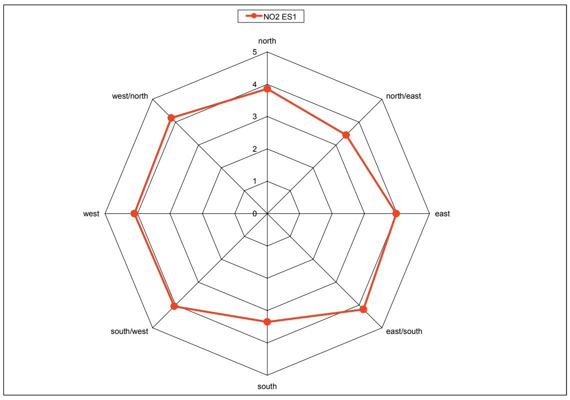

Figure 16: NO_2 concentrations by sectors (ES1), (1985-1996).

As for suspended particles, *Figure 17* and *Figure 18* show the TSP concentrations by sectors in the stations ES2 and ES1, respectively. The first one demonstrates the influence of Saharan dust over La Cartuja. The figure for San Pablo de los Montes (ES1) shows a much lower and more regular concentration.

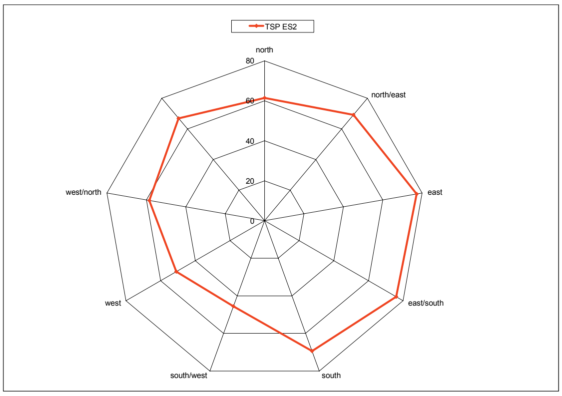

Figure 17: TSP concentrations by sectors (ES2), (1985-1996).

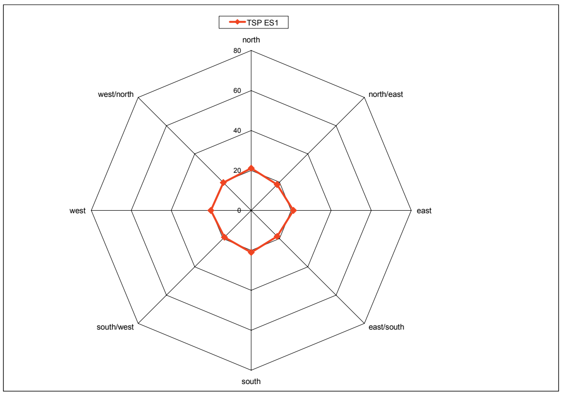

Figure 18: TSP concentrations by sectors (ES1), (1985-1996).

3. COMPARISON WITH THE MODEL

The following figures (*Figure 19* to *Figure 22*) represent the annual mean values measured compared with those predicted by the EMEP model for SO_2, NO_2, particulate SO_4 and NH_3+NH_4 for San Pablo de los Montes (ES1). We can notice in all the figures, except in the one for sulphate, that discrepancies between both values are huge (especially for SO_2 and NO_2). This makes us assert that in the case of Spain, additional improvements of the model are needed.

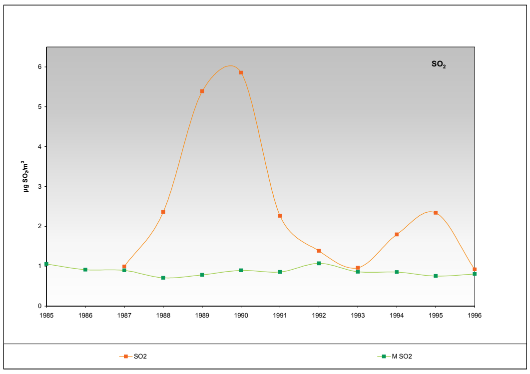

Figure 19: SO_2 (measured vs. modelled (in green)).

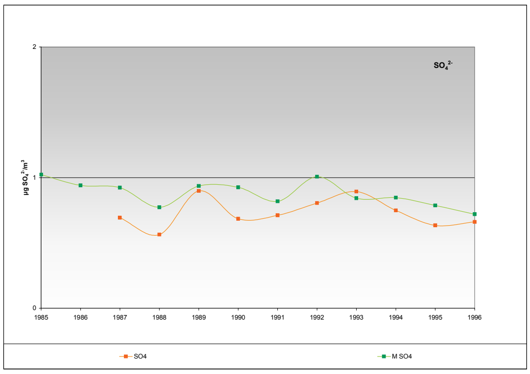

Figure 21: SO_4 (measured vs. modelled (in green)).

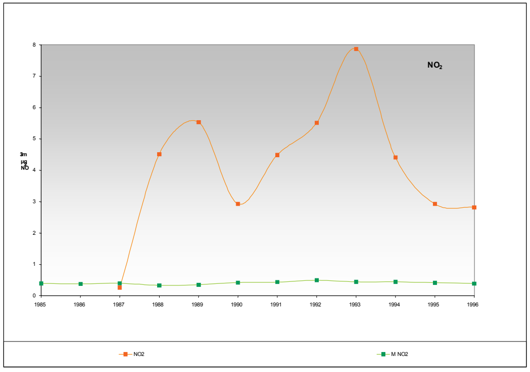

Figure 20: NO_2 (measured vs. modelled (in green)).

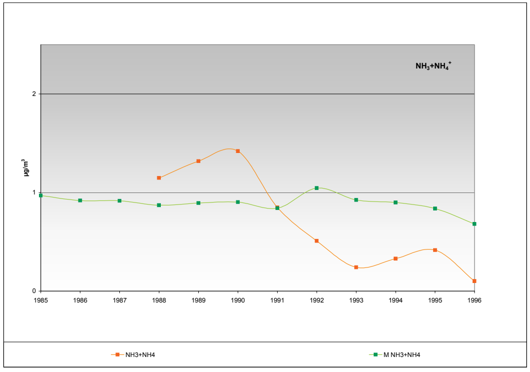

Figure 22: NH_3+NH_4 (measured vs. modelled (in green)).

4. ENVIRONMENTAL STATUS AND PLANS

First of all, the reduction in SO_2 emissions has caused a reduction both in SO_2-S and particulate sulphate concentrations, but wet deposition of sulphate has kept almost constant. NO_2 concentrations have also dropped in spite of a slight rise in its emissions; but, again, wet deposition of nitrate keeps without significant trends.

Regarding the other pollutants, Spain has a particular behaviour in the following aspects: ozone, due to the large number of sunshine hours; TSP, due to natural inputs; and precipitation pH, which is generally basic due to soils and winds from Sahara. Only in the last two cases above (TSP and pH) wind direction influences the pollutants concentration.

SO_2 and NO_2 continuous monitoring was adopted in 2000, as a field comparison proved that it was more reliable than manual data. As a result, and in order to improve the overall network quality, Spain is now carrying out a field comparison campaign in all the stations and a calibration program of the equipments. Measurements of new pollutants (heavy metals, NH_3, VOCs …) are also being implemented.

5. FURTHER MEASURES TO BE TAKEN

Considering the good results achieved by the implementation of the Protocols under the Geneva Convention, the transposition of European Directives (*First Daughter Directive, National Emission Ceiling Directive (NO_x, SO_2, VOCs and NH_3) and the Large Combustion Plants Directive* with their National programme reduction) and further measures would continue the progress in the reduction of pollutant concentrations. In Spain, short and medium-term environmental objectives are encouraging. Social and political conscience towards a more respectful environment and an improvement in air quality have boosted some measures that, without any doubt, anticipate a future in which control and prevention are the keys to decision-making in the different productive systems. A new National Energetic Efficiency Strategy is being developed and soon the mechanisms for its implementation will be established. Fulfilling the objectives and requirements of the Spanish Integrated Prevention Pollution Control Law and the Renewable Energy Promotion Plan, together with the whole framework of Air Quality Directives, is currently the main challenge for Spain regarding the achievement of ideal background pollution levels. In transport, cleaner technologies and lower-sulphur fuels have accomplished significant reductions in emissions of local and regional air pollutants, but additional efforts are needed to reach the targets. However, reaching Spanish national emission reduction targets for 2010 will require substantial further reductions through additional policies including energy taxation policies and measures like promotion of combined heat and power, encouraging the use of renewable energies and increasing the eco-efficiency of the industrial processes.

6. ACKNOWLEDGMENTS

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