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An Evaluation of some Health Risks of the Pollution from Fossil Fuel Combustion

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Paper prepared for the UN / ECE Workshop

**Economic Evaluation of Damage
Caused by Acidifying Pollutants**

London, May 9 - 11, 1994

This document deals with a study in progress. At the present time, its content has not been reviewed by the official sponsorship bodies. Figures of costs are given only for illustrating the approach.

1 Introduction

The Commission of the European Communities, in a cooperative program with the US Department of Energy, is developing an accounting framework for identifying and quantifying the external costs associated with fuel cycles. A general presentation of this program will be set out later in the conference by Mike Holland.

The initial phase consisted of the development of a general methodology by american and british teams¹. This methodology is based on :

- an analysis of the different stages of a fuel cycle and an estimation of pollutants emissions,
- a modelisation of transport and chemical transformation of pollutants from sources to receptors (impact pathways),
- a modelisation of physical effects supported by receptors depending on their exposure (dose-response functions),
- a monetary valuation of physical effects.

In the present phase, INERIS is proceeding to the implementation of this methodology in France for the fossil fuel cycles (coal, oil, natural gas) in the context of the production of electricity.

As part of this programme, we try to evaluate some of the effects on public health of SO₂, NO_x and particulate matter emissions by generation plants.

As in the reference methodology, we particularly consider the effects on human health of particulate matter in air. But our implementation differs from the first EC/US studies on two points :

- we don't only study short range but also long range effects,
- we don't only take account of particulates directly emitted by generation plants but also of particulates resulting of the transformation in sulphates and nitrates of SO₂ and NO_x emissions.

As a consequence, the estimated effects are appreciably more important than in the reference studies.

2 Background

The existence of risks for the human health resulting from the pollution of the ambient air has been well known for a long time. We could call to mind latin authors or recall the prohibition of the use of coal in London by a royal edict in

1273. In the industrial age, the hazards of particulate and SO₂ pollution of urban atmospheres became evident in the conditions of heavy pollution experienced, one more time and particularly, in London in the 1950s, causing the death of thousands of persons.

Since these events a lot of efforts have been made to reduce the emissions of air pollutants and a lot of studies have been carried out to found the environmental policies on scientific facts.

International Organizations have had a leading part in the synthesis. We can particularly mention the work realized by World Health Organization ², United Nations Economic Commission for Europe ³, United Nations Environment Program ⁴, O.E.C.D. ⁵, and Commission of the European Communities ⁶.

Attempts for integrating the health effects of air pollution in a global evaluation of damage have been made in a number of countries, like, for instance, The United States and Germany ⁷.

In France, as in other countries, a continuous effort of research has been made in the fields of toxicology (cf. bibliography in paper ⁸) and epidemiology ⁹.

For example, as early as the seventies, Loewenstein ¹⁰ showed that relation of pollution levels (SO₂ - particulates) with total daily mortality in Paris was very significant for old population.

At the same period, a large study by the cooperative group PAARC on 28 sites distributed in France showed a strong and regular correlation between air pollution in cities (SO₂ concentration), respiratory pathology and decreasing pulmonary function (FEV) ¹¹.

The Authorities in charge of Environment and Public Health in France devote serious attention to the risks of air pollution for human health ¹². A number of articles for the general public show that the opinion is also aware of the problems ¹³.

Since his establishment in 1990, INERIS is involved both in toxicological and epidemiological research ¹⁴ and in damage evaluation ¹⁵. This later field is a relatively new one in France, as few studies had been made up to now for evaluating the economic cost of health effects of air pollution ¹⁶.

3 General approach

The basic approach of the EC/US project is the methodology of Risk Assessment ¹⁷ complemented by economic evaluation.

This approach can be schematized by fig. 2.

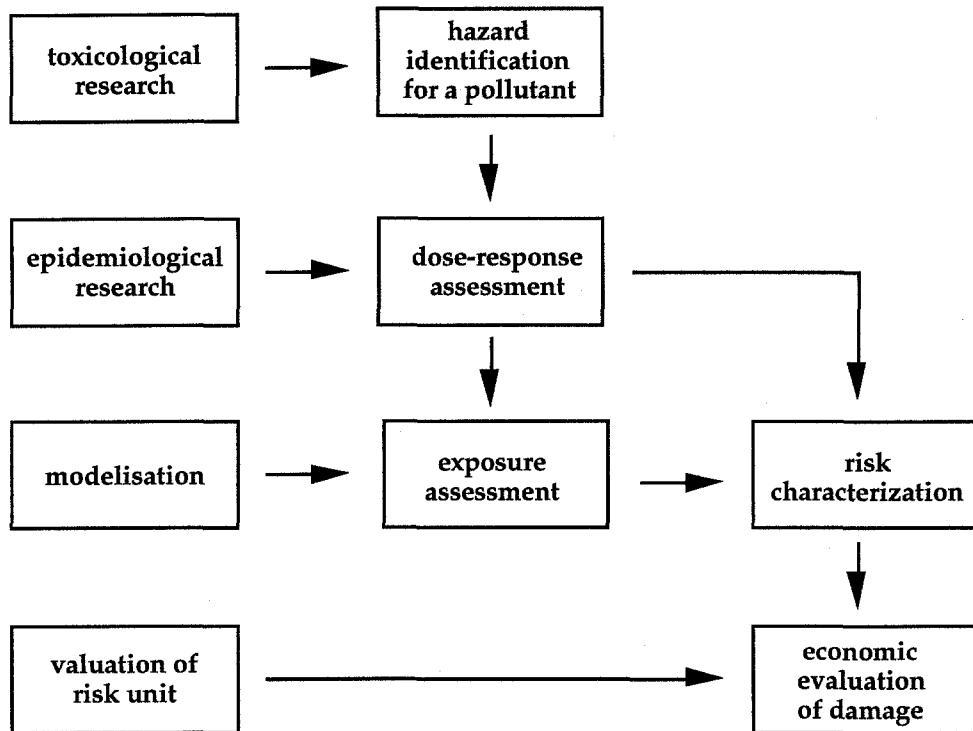


Fig. 2 : The approach of risk assessment
and economic evaluation of damage of a pollutant

In this way, any quantitative evaluation of risks and damage is at first based on a qualitative analysis of the plausibility that a given chemical compound may have a specific effect on man.

At each stage, uncertainty is an underlying feature of any risk assessment :

- toxicological uncertainty : effects on man are inferred from experimentation on animals ; effects at low doses are inferred from experimentation at high doses ;
- epidemiological uncertainty : causal relationship are inferred from statistical correlations ; synergy effects are difficult to discern ;
- modelisation uncertainty : it always exist a risk of neglecting some potential pathway setting in contact a chemical compound and a population.

In this matter of damage evaluation, it often appears that a determinist approach is not really founded and that only a "what if" hypothesis make sens.

4 Hazard identification of SO₂ and NO_x on public health at low concentrations

The hazards for human health resulting of SO₂ and NO_x emissions proceed at first from the direct effects of these compounds but also, indirectly, from the

products of their transformation in the atmosphere (sulphates and nitrate particles, ozone).

It has been said precedently that these hazards are well known for a long time. It must be precised that this assertion is certainly justified for the effects resulting of high pollutants concentrations. But it has been more difficult to demonstrate the effects of more routine situations with lower pollution levels.

However the risks of air pollutants present in the environment even at the low present concentrations appears not necessarily insignificant a priori : one needs only to recall after J. Bignon that man inhales each day 12 000 litres or 15 kg of air (... to compare to 1.5 kg of food and 2 kg of water) ¹⁸ and to consider the phenomenons of penetration, deposition, retention of, for instance, particles inhaled in the respiratory tract ¹⁹.

A large number of epidemiological research projects which has been achieved in the world during the last decade concludes to the existence of effects on man of SO₂, NO_x, ozone and especially particulates even at low concentrations.

Some studies made in France lead to converging results. We would like to mention :

- a study realized as early as 1974 - 1982 in the North region giving a clear indication of a real pathological influence of pollutants (SO₂, particulates and specifically sulphates...), even on moderate concentration levels, lower than european standards ²⁰ ;
- another study on the site of Toulouse in 1985-1986 showing a strong relationship between the concentration in air of particulate ammonia derivatives (sulphates and nitrates) and acute respiratory diseases of children ²¹.
- a study made in 1989 in Paris showing that the frequency of asthma attacks is significantly higher in the days where particulate sulphate concentrations are the most important ²².

A synthesis of the results of the research on the evaluation of health effects has been made recently by a large advisory board as part of the program ERPURS ²³. The board has recorded that many research works realized in routine situations representative of the present environment conclude to a statistical relationship between air pollutants levels and human health. More accurately, the evidence of a relationship between mortality by respiratory causes or, secondly, cardiac causes and particulate pollution is admitted ²⁴.

It is necessary to make clear that the risks in question are short term risks. The evaluation of risks in the long term, resulting from exposure to pollutants for many years, is a problem very difficult to treat scientifically, in view of the statistic requirements of epidemiological studies and the constraints of

observation of living persons. Some attempts have been made to evaluate risks in the long term but the results may be considered as more speculative²⁵.

In consideration of the results of all the cited studies, the EC/US project and the french implementation focus at this stage the health damage evaluation on the risks for health of particulates and, secondly, of SO₂. The risks of ozone could be taken in count later.

5 Quantification of effects of particulate matter : dose-response functions

The precise quantification of dose-response functions at low pollutant concentration requires epidemiological studies realized on a very large scale. That is necessary for discerning weak effect signals on high background noises and distinguishing with which pollutant these effects are more particularly in relationship, even when the variations of concentrations of different pollutants are highly correlated.

A number of such epidemiological studies has been done over the last years especially in The United States on behalf of the Environmental Protection Agency²⁶. These studies are diachronic, that is they try to investigate correlations between daily variations of pollution at a particular site and changes in morbidity and mortality. Successive generations of research, applied to increasingly large populations - for instance, analysis of the daily death rate of 10 million people over 10 years - with increasingly stringent methodologies, appear to confirm the conclusions of the early analyses concerning the effects of particulate pollution and quantify short-term statistical relationship between particulate concentration and the state of health of a population : mortality, lung function deficits, disease symptoms, increased hospitalization.

For instance, the relationships between pollution and mortality appearing as a result of those analysis are approximately linear. These relationships seem identical for different climates (warm-cool, dry-humid...). It has not been possible, in the range of current pollution levels being investigated, to show any threshold below which a change in the level of pollution would no longer have any effect.

A meta-analysis made by J. Schwartz²⁷ come to the conclusion that the dose-response function is of the following form :

*Relative variation of the daily mortality rate
observed in a population exposed to pollution*

$$= 0,6 \cdot 10^{-3} \text{ (confidence interval : } 0,5 \cdot 10^{-3} - 0,7 \cdot 10^{-3})$$

x Variation of daily mean air concentration of particulates ($\mu\text{g TSP}/\text{m}^3$)

In a predictive form, the dose-response relationship can be expressed as follows:

Individual Health Risk (Number of events attributable to pollution per head per year)

= *Response Factor*

x Variation in annual mean concentration of pollutant in the local atmosphere

On the basis of the relation of Schwartz and, if one supposes that the basic mortality of the population is 1% a year and that the effective agent in particulate matter is PM₁₀ (about 50% of TSP in the american context), the value of the response factor in the preceding case could be :

$$RF_{death\ PM10} = 1.2 \times 10^{-5} \text{ death per head, per year, per } \mu\text{g PM}_{10}/\text{m}^3$$

On the basis of other epidemiological analysis, it appears also possible to relate deaths to SO₂ concentration (28) :

$$RF_{death\ SO2} = 0.2 \times 10^{-5} \text{ death per head, per year, per } \mu\text{g SO}_2/\text{m}^3$$

... and to relate the incidence of different signs of disease, such as coughs, bronchitis, asthma, or other events, to a population's exposure to particles, SO₂ and NO_X (29). For instance :

$$RF_{symptom-day\ PM10} = 2 \text{ s.-d. per head. per year, per } \mu\text{g PM}_{10}/\text{m}^3$$

$$RF_{asthma\ attack\ PM10} = 0.01 \text{ a. a. per head., per year, per } \mu\text{g PM}_{10}/\text{m}^3$$

$$RF_{restricted\ activity-day\ PM10} = 0.6 \text{ r.a.d. per head., per year, per } \mu\text{g PM}_{10}/\text{m}^3$$

6 Expression of the health risk

We shall consider a source of pollutants emitting particles, SO₂ and NO_X, which generates a pollution field around itself. The particulate pollution in particular results from the diffusion of particles emitted directly by the source, but also from the secondary formation of sulphates and nitrates from the SO₂ and NO_X.

The health risk to exposed population resulting from those emissions may be put into the mathematical form :

Risk to Exposed Population (Number of events attributable to pollution per year)

$$= \Sigma_{pollution\ field} (\text{Individual Risk } x \text{ Exposed Population})$$

$$= \Sigma_{pollution\ field} (\text{Response Factor } x \text{ Variation in mean concentration of pollutant in the local atmosphere } x \text{ Exposed Population})$$

Risk to Exposed Population

= *Response Factor*

x Population-weighted average variation of pollutant concentration

x Total population over which population-weighted pollutant concentration is determined

If we term collective exposure the product

Collective Exposure

= *Population-weighted average variation of pollutant concentration*

x Total population over which population-weighted pollutant concentration is determined

we have :

$$\text{Risk to Exposed Population} = \text{Response Factor} \times \text{Collective Exposure}$$

For emissions of pollutants like fine particles, SO_2 and NO_x , the pollution field to consider is very large. For example, in average, it is necessary to go away from the source to distances beyond 1000 or 2000 km before 80 % of the emitted atoms of sulphur or nitrogen are deposited on the ground.

Consequently, it is necessary to use long distance pollution transportation models, such as EMEP (Co-operative Programme for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe) for calculating variations of pollutants concentrations and collective exposure attributable to a source.

7

Exposure calculation for a simple case

At the present time, the EMEP model don't produce any grid calculation for the air concentrations of sulphates and nitrates resulting from emissions of SO_2 and NO_x in a pinpoint source like a coal-electricity generation plant.

We therefore attempted to construct a model with a very simple mathematical form to represent the diffusion and chemical modification in the atmosphere of gaseous pollutants. A short description of the model may be found in Annex 1.

In order to illustrate the calculation of collective exposure we shall examine an entirely notional but transparent case rather than a realistic and hence complicated example.

We shall consider a source of atmospheric pollution located at the centre of a circular area; the population within this area is distributed uniformly. We may assume for example that, as France, the area covers $550,000 \text{ km}^2$ - that is a circle of 420 km radius - and the population density is 100 inhabitants per km^2 .

With the simple model described in Annex 1, we have calculated the average pollutant concentrations in the area and the collective exposures resulting from the emissions flow of one tonne of fine particulate per year, one tonne of SO₂ per year, one tonne of NO_x per year. As an illustration, the results are :

Average pollutant concentrations in the area resulting from the source :

fine particulate	:	3.8×10^{-6}	µg / m ³ per tonne part. emitted /year
SO ₂	:	2.6×10^{-6}	µg / m ³ per tonne SO ₂ emitted /year
Sulphate	:	0.6×10^{-6}	µg / m ³ per tonne SO ₂ emitted /year
NO ₂	:	3×10^{-6}	µg / m ³ per tonne NO ₂ emitted /year
Nitrate	:	1×10^{-6}	µg / m ³ per tonne NO ₂ emitted /year

Collective exposures in the area resulting from the source :

fine particulate	:	200	person. µg / m ³ per tonne part. emitted /year
SO ₂	:	140	person. µg / m ³ per tonne SO ₂ emitted /year
Sulphate	:	30	person. µg / m ³ per tonne SO ₂ emitted /year
NO ₂	:	160	person. µg / m ³ per tonne NO ₂ emitted /year
Nitrate	:	55	person. µg / m ³ per tonne NO ₂ emitted /year

Note

It will be seen that although the case proposed is notional it is not unconnected to existing situations.

One of the EC/US project's reference studies calculates, using the Harwell model, that the Lauffen power plant generates the following mean pollution concentrations in Germany for each tonne of pollutant emitted:

Mean concentrations (10^{-6} µg/m³) for each tonne emitted

	German case	Our example
particles	3.3	3.8
SO ₂	3.2	2.6
NO ₂	4.2	3

Similarly a study by the Brookhaven National Laboratory ³⁰, estimates that an annual emission of one tonne of SO₂ in the United States results in an average collective exposure to sulphates of 31 person.µg/m³: this is precisely the figure obtained in our example...

8 Health risks calculation

The expression of risk above-mentionned is :

$$\text{Risk to Exposed Population} = \text{Response Factor} \times \text{Collective Exposure}$$

For example, it is possible to calculate value of death risk in the notional case considered on the basis of the dose-response relationship of Schwartz and Hatzakis.

The death risk generated by the emission of particulates is :

Risk (death) particles

$$= 1.2 \times 10^{-5} \text{ death per head, per year, per } \mu\text{g PM}_{10} / \text{m}^3 \\ \times 200 \text{ person. } \mu\text{g / m}^3 \text{ per tonne part. emitted / year}$$

Or:

$$\text{Risk (death) particles} \approx 2.5 \times 10^{-3} \text{ attributed deaths per tonne of part. emitted}$$

Similarly we can calculate:

$$\text{Risk (death) SO}_2 = 0.2 \times 10^{-5} \text{ death per head, per year, per } \mu\text{g SO}_2 / \text{m}^3 \\ \times 140 \text{ person. } \mu\text{g / m}^3 \text{ per tonne SO}_2 \text{ emitted / year}$$

$$\text{Risk (death) SO}_2 = 0.3 \times 10^{-3} \text{ attributed deaths per tonne of SO}_2 \text{ emitted}$$

Since an emission of sulphur dioxide generates sulphates, i.e., particles, we also have:

$$\text{Risk (deaths) sulphate} = 1.2 \times 10^{-5} \text{ death per head, per year, per } \mu\text{g part/m}^3 \\ \times 30 \text{ person. } \mu\text{g / m}^3 \text{ per tonne SO}_2 \text{ emitted / year}$$

$$\text{Risk (deaths) sulphate} = 0.4 \times 10^{-3} \text{ attributed deaths per tonne SO}_2 \text{ emitted}$$

By adding the primary and secondary risks of SO₂ we have:

Risk (death) SO₂ + Sulphate

$$\approx 0.7 \times 10^{-3} \text{ attributed deaths per tonne of SO}_2 \text{ emitted}$$

Similarly, one may calculate :

Risk (death) nitrate

$$\approx 0.7 \times 10^{-3} \text{ attributed deaths per tonne of NO}_X \text{ emitted}$$

Knowing the risks data determined by epidemiological studies, it would be possible to evaluate in the same way the number of cases of coughs, bronchitis and asthma or even the number of working days lost through sickness, per tonne of particulate, SO₂ or NO_X emitted.

9 Monetary value

In the reference studies of the EC/US project, the hypothesis proposed for the value of one "statistical risk of death" is

$2,6 \cdot 10^6$ ECU, that is near 20 million FF.

French economists ³¹ working on the project consider that this figure is high, particularly if one consider the price of human life taken in consideration in matter of road safety investments, that is 3.3 million FF.

Other economists insist on the fact that the death risk may touch more probably persons very old or seriously ill before, that is people having a lower expectation of life than the average man ³².

However, on the above-mentionned basis, and in the notional case considered, the translation of the physical evaluation of risk in a monetary form give, for example :

Cost (death) particles	\approx	50,000 FF / tonne particulate emitted
Cost (death) SO ₂ + sulphate	\approx	14,000 FF / tonne SO ₂ emitted
Cost (death) nitrate	\approx	14,000 FF / tonne NO _X emitted

Taking account of the cited morbidity effects of particulate pollution and of the economic cost of those effects, a health external cost could be calculated in the same hypothetical way :

External Health Cost particles	\approx	100,000 FF/tonne part. emitted
External Health Cost SO ₂ + sulphate	\approx	23,000 FF / tonne SO ₂ emitted
External Health Cost Nitrate	\approx	30,000 FF / tonne NO _X emitted

10 A health component in the external cost of a thermal power station

We shall consider a modern coal power plant, equipped with desulphurisation by flue gas scrubbing and selective catalytic reduction of nitrogen oxides, at the centre of the circular geographical area constituting our illustrative example.

Basic data

Net plant output: 630 MW
"Equivalent" operating time: 4010 hours/year
(giving an annual output of 2.53 TWh)

Emission of particles:	0.2 g/kWh	or 480 tonnes/year
Emission of SO ₂ :	0.8 g/kWh	
Emission of NO ₂ :	0.8 g/kWh	or 1925 tonnes/year

Health risk

The annual death rate attributed to the plant in the illustrative case is:

$$\begin{aligned}\text{Risk (deaths) plant year} &= (480 * 2.5 \times 10^{-3}) + (1925 * 0.7 \times 10^{-3}) + (1925 * 0.7 \times 10^{-3}) \\ &\quad \text{attributed deaths} \\ \text{Risk (deaths) plant year} &\approx 4 \text{ attributed deaths}\end{aligned}$$

External health cost

The external health cost is calculated in the same way:

$$\begin{aligned}\text{External Health Cost plant year} &= (480 \times 100,000) + (1925 \times 23,000) + (1925 \times 30,000) \text{ FF}\end{aligned}$$

$$\text{External Health Cost plant year} \approx 150 \text{ MF}$$

Expressing this external cost per kWh generated, we obtain for our typical case:

$$\text{External Health Cost} \approx 6 \text{ cF / kWh}$$

11 Conclusion of the exercise

The above evaluation is primarily an illustration of the approach used in the External Costs of Fuel Cycles project.

However it highlights the importance of taking into account the secondary chemical forms of sulphur and nitrogen pollution, which was not considered in the calculations of the EC/US reference studies, unlike the other studies on the impacts of energy systems.

In fact, sulphates and nitrates constitute a major part of the production of particulates from human origin (cf. ³³). Those secondary particles had been

taken into account in a number of international health impact studies mentionned in our introduction, as also in some french studies³⁴.

With the renewed attention paid in recent years to the impact of particulate pollution on health, awareness of the health risk related to secondary particles has become predominant amongst scientists working in the field³⁵.

We could not conclude without recalling that the calculations made in our exercise concern only some measurable short term effects of a pollution. It is not impossible that those measurable effects are, as it has been said³⁶, only the emerging part of the iceberg.

Annex 1

Long-range dispersion and chemical modification of SO₂, NO_x and particulate matter in the atmosphere:

A simplified transport model

Introduction

The EC/US external environmental costs project does not require great precision in the geographical representation of the pollution induced by a source, for this would be futile in the light of the considerable uncertainties affecting the other stages of the evaluation process. Nor does it require the pollution to be monitored over time, needing only the mean values at one particular place.

We therefore attempted to construct a model with a simple mathematical form to represent the diffusion and chemical modification in the atmosphere of gaseous pollutants. The works on similar problems by M. A. Doury³⁷ of the Safety Analysis Department of the French Atomic Energy Commission (CEA) and F. B. Smith³⁸ of the British Meteorological Office suggested to us that such an approach was credible.

The basic concepts of the proposed representation are conventional. The model for the diffusion of a non-reactive chemical compound that we shall describe in the first section is for example that used by the IPSN for an evaluation of external effects at regional level³⁹.

In the second part of this note we shall propose extending the basic model to the problem of the transport of a pollutant whose chemical form changes with time. Typical such pollutants are sulphur, emitted in the form of sulphur dioxide SO₂ and converted with time to the sulphate, and nitrogen, emitted as nitrogen oxides NO_x and becoming the nitrate.

1. The basic dispersion model

Let us assume that a point source discharges a pollutant into the atmosphere at a constant rate E.

It is assumed that the pollutant is dispersed in space isotropically around the source :

- the pollutant is diffused within a layer of the atmosphere characterized by a constant depth h ;

- the pollutant concentration C at a receiver is uniform throughout the thickness of this "mixing layer";
- the pollutant is dispersed centrifugally at a constant velocity u, representing the average wind speed in the mixing layer.

1.1 Pollutant remaining suspended indefinitely in the atmosphere

There are steady conditions and mass is conserved. The pollutant flux Φ which passes through a vertical cylinder of radius D centred on the source is therefore constant and equal to the emission rate E:

$$\Phi = 2 \pi D h u C = E$$

Thus the pollutant concentration C is inversely proportional to the distance D; by putting $\alpha = \frac{1}{2 \pi h u}$, the expression becomes:

$$C = \frac{\alpha}{D} E$$

Numerical value of the coefficient α

The height of the mixing layer and the wind velocity at altitude are changing meteorological variables and the estimate of the mean value to be used differs according to the models. However the order of magnitude of the parameter α is fairly well known. The following are the values used in the Harwell model⁴⁰ and by Doury.

			Harwell	Doury
Height of mixing layer	h	m	800	1000
Mean wind velocity in mixing layer	u	$m s^{-1}$	7.5	5
$\alpha = \frac{1}{2 \pi h u}$		$s m^{-2}$	2.7×10^{-5}	3.2×10^{-5}

Choice of system of units

The numerical value for the coefficient α given above implicitly assumes the use of a coherent system of units:

- emission rate g/s
- concentrations in g/m³
- distances in m

In practice it may appear more convenient to choose another system, for example:

- emission rate in tonnes/year
- concentrations in $\mu\text{g}/\text{m}^3$
- distances in km

In such a system, the new value of the coefficient α (α') is obtained by multiplying the previous figure by 10^{12} ($\mu\text{g}/\text{t}$) and dividing by 31.5×10^6 (s/year) and by 10^3 (m/km). The calculation gives:

$$\alpha' \approx 10^{-3}$$

1.2 Stable pollutant falling to the ground with a constant deposition velocity

Many pollutants do not stay suspended indefinitely in the atmosphere but are deposited on the ground by sedimentation (particles) or absorption (gases).

In this hypothesis, the pollutant flux Φ (D) passing through a cylinder of radius D centred on the source is no longer constant as it was in the previous case, but decreases as the radius D increases.

The velocity of pollutant deposition is defined as the observed ratio between the mass of pollutant falling at a particular place per unit surface area and per unit time, and the concentration of the pollutant in the atmosphere above that place.

If this sedimentation velocity v_1 is constant, the flux decreases with distance from the source to a rate

$$\sigma_1 = \frac{v_1}{h} \quad \text{per unit time.}$$

Besides this "dry" mode of deposition, the pollutant can also be entrained or dissolved by rain and fall to the ground wet. An equivalent velocity v_2 and a rate σ_2 are assigned to this mode of wet deposition. In total, the mean rate of reduction of flux per unit time is

$$\sigma = \sigma_1 + \sigma_2$$

The mean rate of reduction in the flux per unit distance as one moves away from the source is

$$r = \frac{\sigma}{u}$$

Thus the pollutant flux $\Phi(D)$ falls exponentially as a function of D :

$$\Phi(D) = e^{-rD} E \quad (1)$$

This flux expressed as a function of the concentration is, as indicated previously:

$$\Phi(D) = 2\pi D h u C = \frac{1}{\alpha} D C$$

The expression for the concentration as a function of distance becomes:

$$C(D) = \frac{\alpha}{D} e^{-rD} E \quad (2)$$

For a chemically stable pollutant therefore, the schematic model given above can be used to calculate the concentration of a pollutant in the atmosphere as a function of emission rate and distance from the source using two parameters: the parameter α characterising the atmosphere in which diffusion takes place, and a parameter r characterising the deposition velocity of the pollutant.

Significance of the coefficient r

The form of equation (2) gives $\frac{1}{r}$ a second physical meaning:

$T_{mean} = \frac{1}{\sigma}$ is the mean time during which a molecule of pollutant remains in suspension in the atmosphere before falling to the ground;

$D_{mean} = \frac{1}{r}$ is the mean distance travelled by a molecule of pollutant in the atmosphere before falling to the ground.

1.3 Typical application: transport of particles emitted by a thermal power station

It is known that fine dust can remain in suspension in the atmosphere for long periods and be transported over long distances; an example occurs when Sahara sand falls on Northern Europe; the attention of public opinion was drawn to the phenomenon during the wanderings of the Chernobyl cloud.

1.3.1 Particle deposition velocity as a function of radius

The dry deposition velocity of a particle in the atmosphere depends first on its diameter. Different laws for deposition velocity as a function of diameter apply to different ranges of particle size.

In the range of dimensions that concerns us here, we may refer for example to the equation produced by R. Jaenicke ⁴¹, quoted by A. Renoux ⁴². The mean dwell time T in the low troposphere of a particle of radius ρ is given by:

$$\frac{1}{T} = \frac{1}{C} \left(\frac{\rho}{R} \right)^2 + \frac{1}{C} \left(\frac{\rho}{R} \right)^{-2} + \frac{1}{T_{wet}} \quad (J)$$

where, with time expressed in seconds and radii in microns,

$$\begin{aligned} C &= 1.28 \times 10^8 \text{ s} \\ R &= 0.3 \mu\text{m} \\ T_{wet} &= 6.91 \times 10^5 \text{ s} \end{aligned}$$

With the notation used previously the equation (J) can be written:

$$\sigma_1 = \frac{1}{C} \left(\left(\frac{\rho}{R} \right)^2 + \left(\frac{\rho}{R} \right)^{-2} \right)$$

$$\sigma_2 = \frac{1}{T_{wet}} \quad (\text{a constant})$$

Applying the height of the mixing layer used by Harwell, we can also write:

$$v_1 = 6.25 \times 10^{-6} \left(\left(\frac{\rho}{R} \right)^2 + \left(\frac{\rho}{R} \right)^{-2} \right) \text{ m s}^{-1}$$

$$v_2 = 1.16 \times 10^{-3} \text{ m s}^{-1}$$

1.3.2 Deposition parameter

It will be assumed that a mean diameter of 2 μm is representative of the particles emitted by a present-day thermal power station.

Using Jaenicke's formula for a particle of average diameter 2 μm , i.e. radius 1 μm , gives the following deposition velocities:

$$v_1 = 0.07 \times 10^{-3} \text{ m s}^{-1}$$

$$v_2 = 1.16 \times 10^{-3} \text{ m s}^{-1}$$

$$v_1 + v_2 = 1.23 \times 10^{-3} \text{ m s}^{-1}$$

The calculated dry deposition velocity appears very low (6 m a day) and it seems that the main cause of fall-out is atmospheric wash-out by rain.

The value of the parameter r in the formula (2) is:

$$r = 0.2 \times 10^{-6} \text{ m}^{-1}$$

This value corresponds to a mean dwell time in the atmosphere of

$$T_{\text{mean prop}} \varnothing 2 \mu = 0.65 \times 10^6 \text{ s} = 7.5 \text{ days}$$

and a mean distance of atmospheric transport of:

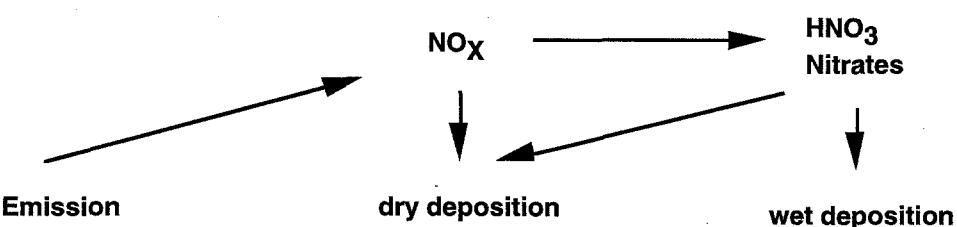
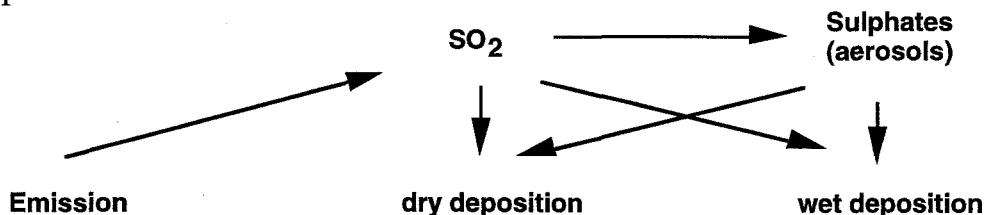
$$D_{\text{mean prop}} \varnothing 2 \mu \approx 5000 \text{ km}$$

2 Modelling the transport of a reactive pollutant

2.1 Changes in the chemical form and deposition velocity of the pollutant during transport: a simplified representation of the phenomenon

The polluting element, emitted as a component of a primary chemical compound can, as a result of actions taking place in the atmosphere, gradually turn into a component of a secondary chemical compound.

For example an increasing proportion of the sulphur emitted in the form of sulphur dioxide is transformed with time into sulphate. Similarly, although its chemistry is more complex, we shall assume, in a very simplified manner, that nitrogen emitted in the form of nitrogen oxide gradually turns into a component of nitrates.



The total concentration of the polluting element in the atmosphere is the sum of two terms, C_P the concentration of the element in the form of the primary chemical compound and C_S the concentration in the form of the secondary chemical compound:

$$C = C_P + C_S$$

2.1.1 Concentration in primary form

Let us assume that all the pollutant is emitted in the form of the primary compound and that this compound is consumed by the oxidation reaction at a constant rate τ per unit time.

The flux Φ_P and the concentration C_P of the primary pollutant are expressed in ways similar to equations (1) and (2). They differ only in the fact that the rate of reduction per unit time is now the sum of two terms:

- the deposition rate $\sigma_P = \sigma_{P1} + \sigma_{P2}$ (dry and wet deposition)
- the oxidation rate τ

By putting

$$\gamma = \frac{\sigma_P + \tau}{u}, \text{ we have :}$$

$$\Phi_P (D) = e^{-\gamma D} E \quad (3) \quad \text{and}$$

$$C_P (D) = \frac{\alpha}{D} e^{-\gamma D} E \quad (4)$$

2.1.2 Concentration in the secondary form

The change in the flux Φ_S and the concentration C_S of the secondary compound is the resultant of two processes: the production of the secondary compound by oxidation of the primary compound at the rate τ , and the elimination of the secondary compound by deposition on the ground at a rate $\sigma_S = \sigma_{S1} + \sigma_{S2}$.

For an observer moving with the flow, the variation $d\Phi_S$ in the secondary compound flux during an infinitely short time interval dt is:

$$d\Phi_S = \tau \Phi_P dt - \sigma_S \Phi_S dt$$

$$\text{or, by putting } \delta = \frac{\sigma_S}{u} \quad \text{and} \quad \theta = \frac{\tau}{u}$$

$$d\Phi_S = \theta \Phi_P dD - \delta \Phi_S dD$$

Replacing Φ_P by expression (3), we have :

$$\frac{d\Phi_S}{dD} + \delta \Phi_S - \theta E e^{-\gamma D} = 0$$

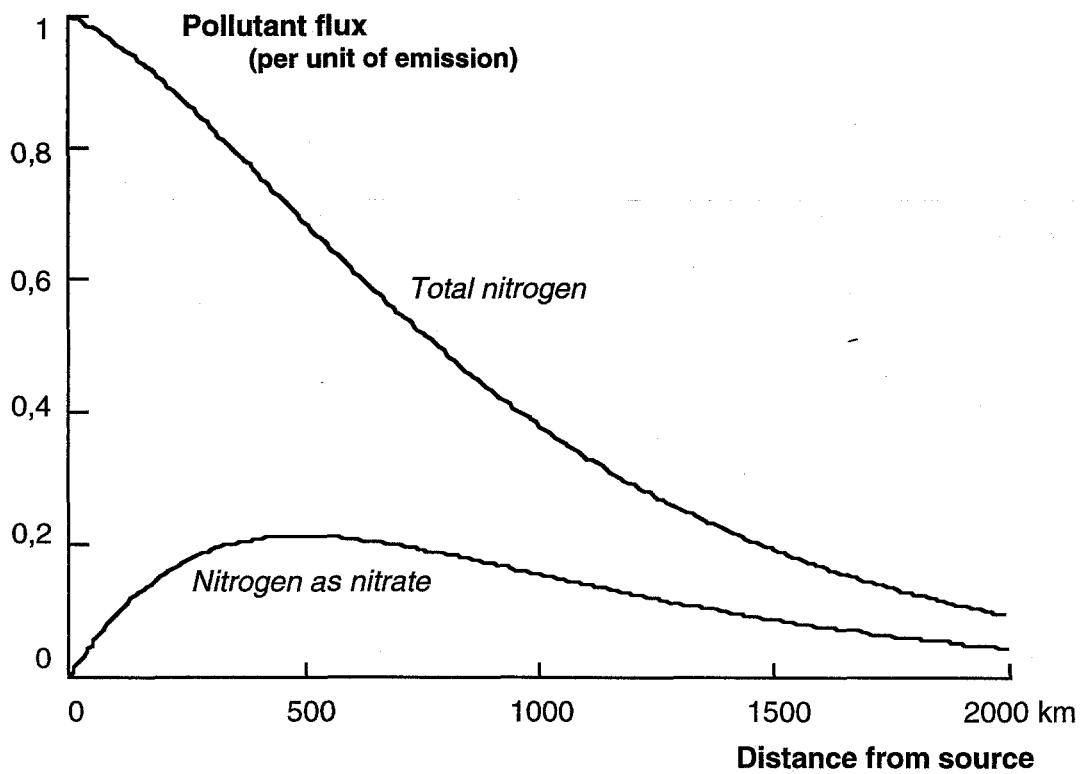
Integrating this differential equation leads to the expression:

$$\Phi_S(D) = \frac{\theta}{\delta - \gamma} (e^{-\gamma D} - e^{-\delta D}) E \quad (5)$$

and by putting $\beta = \frac{\theta}{\delta - \gamma}$

$C_S(D) = \frac{\alpha}{D} \beta (e^{-\gamma D} - e^{-\delta D}) E \quad (6)$

Plot of the function Φ / E



2.2 Estimation of parameters

When the pollutant "changes vehicle" during transport, the calculation of its concentration in the atmosphere as a function of emission rate and distance from the source requires two new parameters to be considered: besides the parameter α characterising the atmospheric medium in which diffusion takes place and the deposition velocity in the first form, it is necessary to take into account the kinetics of the change to the second form and the deposition velocity in this second form.

For a first evaluation of these basic quantities, in the case of sulphur and nitrogen, we shall rely on the estimates of the same parameters used for a more complex model, the TREND model (cf. Derwent, op. cit.).

Pollutant lifetime parameters

Pollutant		Sulphur	Nitrogen
<i>Primary compound</i>		SO ₂	NO ₂
Dry deposition velocity	v _{P1}	m s ⁻¹	9 x 10 ⁻³
Dry deposition rate	$\sigma_{P1} = \frac{v_{P1}}{h}$	s ⁻¹	11.25 x 10 ⁻⁶
Wet deposition rate	σ_{P2}	s ⁻¹	0.8 x 10 ⁻⁶
Oxidation rate τ		s ⁻¹	4.45 x 10 ⁻⁶
$\theta = \frac{\tau}{u}$		m ⁻¹	0.59 x 10 ⁻⁶
<i>Secondary compound</i>		Sulphates	Nitrates
Dry deposition velocity	v _{S1}	m s ⁻¹	2.8 x 10 ⁻³
Dry deposition rate	$\sigma_{S1} = \frac{v_{S1}}{h}$	s ⁻¹	3.5 x 10 ⁻⁶
Wet deposition rate	σ_{S2}	s ⁻¹	10 x 10 ⁻⁶
<i>Parameters of the model</i>			
$\gamma = \frac{\sigma_{P1} + \sigma_{P2} + \tau}{u}$		m ⁻¹	2.2 x 10 ⁻⁶
$\delta = \frac{\sigma_{S1} + \sigma_{S2}}{u}$		m ⁻¹	1.8 x 10 ⁻⁶
$\beta = \frac{\theta}{\delta - \gamma}$		- 1.5	1

Value of the parameter β according to the type of compounds taken into account

Standard practice is to sum the emissions and primary concentrations of sulphur and nitrogen in terms not of the masses of the elements but of SO₂ and NO₂ respectively.

In the hypothesis where the secondary product from sulphur consists of a mixture of equal proportions of SO₄(NH₄)₂ and SO₄HNH₄, the mass of compound formed is the mass of SO₂ entering the reaction multiplied by a

factor of 1.93. The concentration of sulphate as a function of an emission expressed as SO_2 will therefore be obtained by replacing β in formula (6) by:

$$\beta' \text{ sulphate} / \text{SO}_2 = -2.9$$

Similarly, if the secondary product, as regards nitrogen, consists of NO_3NH_4 the mass of compound formed is the mass of NO_2 entering the reaction multiplied by a factor of 1.74. The concentration of nitrate as a function of an emission expressed as NO_2 will be obtained by replacing β in formula (6) by:

$$\beta'' \text{ nitrate} / \text{NO}_2 = 1.7$$

2.3 A test of the credibility of the model using EMEP evaluations

The Norwegian Meteorological Institute (MSC-W) was kind enough to provide us with special data giving the geographical distribution in Europe of fall-out of sulphur and nitrogen (oxides) emissions from the region of Nantes calculated using the EMEP model.

The areas of constant fall-out density form rings, more or less regular in shape, around the source. These data were smoothed to eliminate the variation of fall-out as a function of direction. Thus the assumption was made that an area of constant density could be characterised by a single distance from the source; this distance is calculated as being equal to the radius of the circle whose area equals that of the zone bounded by the outer border of the area of equal density.

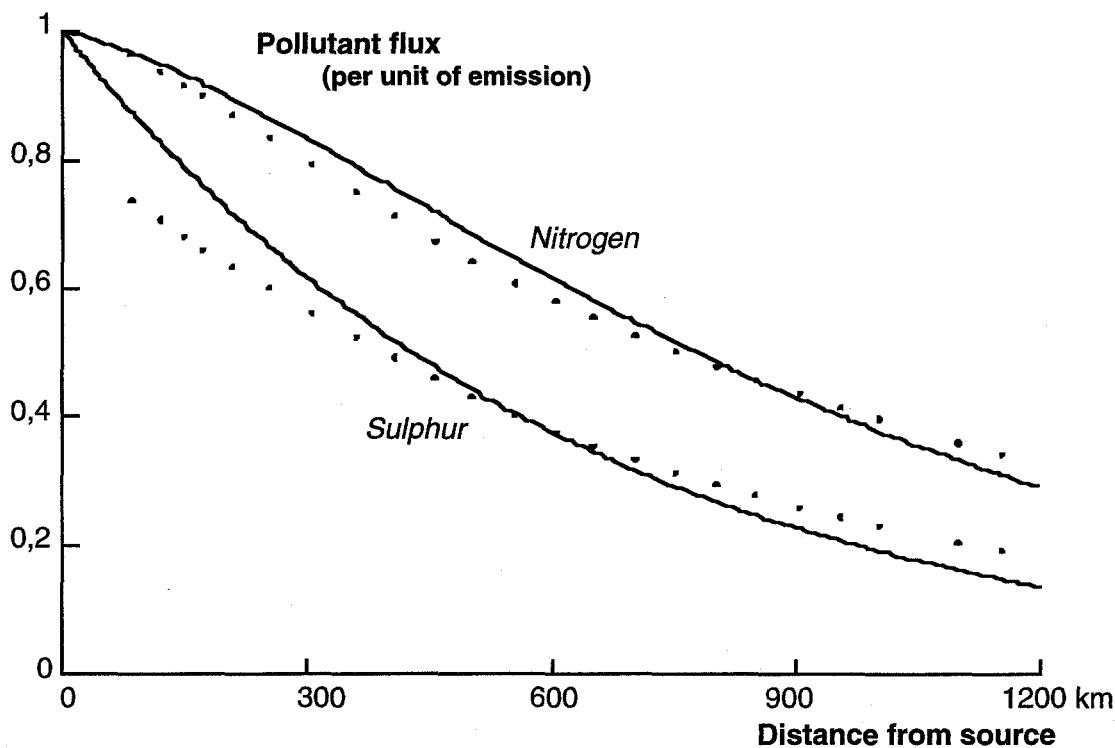
In this way it is possible to calculate the cumulative amounts of pollutant falling on successive areas of decreasing density, i.e., at increasing distances from the source. It is also possible to calculate the proportion of the emitted pollutant which remains suspended in the atmosphere at a given distance from the source. The following graph shows EMEP data specific to the Nantes emission zone, treated in this way (the points represent a series of zones of equal density).

This graph shows (solid lines) the estimate of the ratio

$$\frac{\Phi(D)}{E} \quad \text{or} \quad \frac{\Phi_P + \Phi_S}{E}$$

from the simplified model we presented and using the numerical values for the parameters proposed above as an initial approach.

The closeness of the two estimates can be regarded as encouraging for the mathematical model proposed. The model correctly represents the distribution of areas on the ground as a function of the fall-out levels to which they are exposed.



The model correctly represents the distribution of areas on the ground as a function of the fall-out levels to which they are exposed.

It now remains to be shown that the model accurately reflects the relative concentrations of the primary and secondary forms of pollution. We hope to have data in the future for testing the model from this point of view and, if necessary, calibrating it by optimising the values of the parameters used.

At the same time it will be possible to improve the model by incorporating directional factors in the distribution of pollution (introducing the pattern of winds on a regional scale).

3 Calculation of mean pollutant concentrations in a simple case

We shall consider a source of atmospheric pollution located at the centre of a circular area. In such a situation, with a circle of radius R around a source with constant emission E , the expression for the mean pollutant concentrations is obtained directly by integrating the expressions (2), (4) and (6):

$$C_{\text{part mean}}(R) = \frac{2\alpha}{R^2} \frac{1 - e^{-r/R}}{r} E$$

$$C_{\text{P mean}}(R) = \frac{2\alpha}{R^2} \frac{1 - e^{-\gamma R}}{\gamma} E$$

$$C_{\text{S mean}}(R) = \frac{2\alpha\beta}{R^2} \left(\frac{1 - e^{-\gamma R}}{\gamma} - \frac{1 - e^{-\delta R}}{\delta} \right) E$$

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