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Faculty of Environmental Science and Engineering

OPTOELECTRONIC TECHNIQUES FOR ATMOSPHERIC MONITORING USED FOR THE ASSESSMENT OF NATURAL HAZARDS AND TECHNOLOGICAL RISKS

- PhD THESIS summary -

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Table of contents

Keywords: hazard and risk, optoelectronic techniques, atmospheric remote sensing, dispersion simulations

The summary contains a part of the thesis' results, general conclusions, and selective bibliography. The summary has the same notations for the table of contents, chapters, sub-chapters, figures, tables and equations as the thesis.

1. Introduction. Concept and objectives

1.1. Introduction

The magnitude of the effect humanity and its activities have on environmental change is one of the most pressing issues currently debated by all members of society. Air quality and climate change are issues that have complex socio-political implications. The potential impact of atmospheric processes and climate change on society is of crucial importance and therefore requires advanced scientific research into the causes, consequences and mitigation of such changes, in order to develop efficient strategies.

Clean air is a basic condition for the health of the population and the environment, and for maintaining biodiversity. Health effects are an issue especially in the case of combustion gases (SO_x, NO_x,) and particulate matter (PM).

Aerosols are an important component of the atmospheric mixture which influences radiative forcing through the atmosphere (Ştefan, 2004). Also there is a lack of detailed knowledge in order to exactly quantify the influence they have on Earth's radiative budget.

The variations in the Earth's energy equilibrium is known as radiative forcing, which is used for comparing the heating or cooling of the Earth's climate. According to IPCC, 2007, the anthropogenic contributions of aerosols have a cooling effect, with radiative forcing values of -0.5 [-0.9 to -0.1] W/m^2 and an indirect cloud albedo forcing of -0.7 [-1.8 to -0.3] W/m^2.

The optical parameters of aerosols can be retrieved using optoelectronic techniques, more precisely through passive and active remote sensing techniques. Passive remote sensing techniques like sun-photometry yield valuable information concerning the above mentioned aerosol properties. A worldwide network of sun-photometers (NASA-AERONET) offers valuable information from different environments, for aerosols both of natural and anthropogenic origin (Holben et al., 1998).

Increased attention is also given both at governmental level and within the international scientific community to the **primary air pollutants** like $\mathbf{SO_2}$, NO_x and NH_x. Their contribution to the formation of acid rains leads to decreased ecosystem vitality due to acidification and eutrophication of water and soils.

One of the major industrial facilities emitting mainly gases (SO_2, NO_x and CO_2) and particulate matter are the large combustion plants (LCPs).

Atmospheric modeling is widely used to support decision making on environmental matters and pollution abatement. The models used in a regulatory context are often relatively simple, as compared with a range of other more complex models that reflect atmospheric processes in more detail.

A serious limitation in modeling of atmospheric processes is due to the unavailability of data describing the vertical structure of the atmosphere, therefore the calculation of the mixing layer is possible only through rough approximations. Point monitoring systems measure concentrations only at ground level, but the vertical distribution of concentrations of pollutants is not provided. Also, ground level concentrations can be influenced by vertical transport induced by turbulent mass flux. Even more important, the direct and indirect climate impact of aerosols depends on the total aerosol load in the atmospheric column and its vertical distribution.

This thesis will try to overcome these shortcomings by using advanced 3D optoelectronic systems for the measurement of vertical distribution of aerosols (LIDAR and sun-photometry), and of high performance UV cameras for optical determination of gas particle concentrations in the atmosphere. This synergy of optoelectronic instruments supplemented by modelling techniques and in-situ measurements has a unique potential for advanced atmospheric studies and will bring a considerable added value to impact and risk assessment methodologies (Ajtai et al., 2011a).

1.2. Concept and objectives

In this context, **the concept** of the thesis revolves around new and innovative methodologies and techniques for a better understanding and analysis of natural hazards and technological risks using optoelectronic techniques for environmental monitoring. The hazard and risk concepts are of great interest for researchers, academia, industry and authorities. This wide spectrum of stakeholders generates multiple types of needs to be addressed. Therefore different types of hazard and risk analyses must be employed in order to address these needs.

A systematic identification of possible environmental hazards, as well as a rigorous analysis of the risks associated with them is required. The purpose of such a process can be divided in two major issues, on one hand to produce quantitatively accurate estimation of particular risk and a comprehensive list of possible environmental impacts, and on the other hand to develop a solid platform for making public policy decisions that is both well reasoned, and recognized as legitimate and acceptable by the socio-economical factors.

The **major objectives** of the thesis can be summarized as follows:

1. **present a theoretical background concerning hazard and risk analysis, the atmosphere and optoelectronic monitoring techniques;**

In order to mitigate natural and technological hazards an assessor must have a solid knowledge regarding the hazard and risk concept both individually and combined. Differences between terms, types of analyses employed in various situations must also be clearly defined. The theoretical part of this thesis addresses the above mentioned issues along with a detailed presentation of the environment studied, in this case the atmosphere and associated processes.

2. **propose optoelectronic techniques for natural hazard and technological risk analysis;**

A sound theoretical description of the techniques and methods employed in the identification and analysis of hazards and risks must be presented. In our case, several optoelectronic techniques for atmospheric monitoring were selected: passive and active remote sensing, UV cameras and in situ UV-fluorescence gas analyzers.

3. **present case studies supporting the use of the selected optoelectronic techniques for hazard and risk analysis;**

In order to demonstrate the appropriateness of the use of the above mentioned optoelectronic techniques, a series of case studies are presented for the identification and analysis two major types of hazards and risks:

- natural hazards: a case study on the detection and characterization of a volcanic ash intrusion using LIDAR and sun-photometers
- technological risk: three case studies on monitoring and modeling of SO_2 emissions from large combustion plants using UV cameras, dispersion modeling and in-situ SO_2 imissions monitoring

4. **develop frameworks in which the data outputted by these techniques can be used for a better integration in the assessment process and finally in the development of strategies.**

Finally, from the conclusions drawn from the case studies, two frameworks are developed, one for natural hazards and one for technological risks in order to better integrate optoelectronic techniques for atmospheric monitoring in the risk and impact assessment process as a whole.

This part is necessary because a decision maker using raw results obtained from a scientific study, may encounter difficulties in justifying or agreeing over actions to protect the environment. The results obtained by monitoring and modeling must be used and presented in a more broader perspective in order for the results of scientific research to have a maximum impact on stakeholders, regulation agencies and the general public.

The thesis is structured as follows:

Chapter 1 is the introduction of the thesis, underlining the necessity of advanced atmospheric research and proposes optoelectronic techniques for atmospheric monitoring as a tool for risk and impact analysis. The concept and major objectives of the thesis are also stated in this chapter.

Chapter 2 presents the hazard and risk concepts, describing different perspectives the scientific community has over these two concepts and underlines the major differences between the approaches. Chapter 2 also presents a classification of hazards according to their different characteristics and proposes qualitative and quantitative methods of assessment. The case studies are chosen according to an important classification: natural (volcanic ash) and technological (SO_2 emissions) hazards. The chapter also covers the concept of NATECH risks represented by technological accidents triggered by natural hazards.

Chapter 3 describes the theoretical background regarding the atmosphere in general, with emphasis on aerosols and atmospheric gases. The interaction of light with atmospheric constituents is also described here, continued with a description of the optoelectronic techniques proposed for the monitoring of hazardous components. This chapter proposes passive (sun-photometers) and active (LIDAR) remote sensing systems, in-situ gas monitors (SO_2 analyzers) and UV cameras as optoelectronic instruments for detection and characterization of hazardous properties of aerosols and SO_2 emissions.

Chapter 4 presents the major aerosol types (urban-industrial, biomass-burning, mineral dust) describing their optical and microphysical properties as well as their hazardous properties. A case study on the detection and characterization of a volcanic ash (natural hazard) intrusion is presented in order to demonstrate the validity of the approach based on active and passive remote sensing in indentifying and characterizing hazardous particle intrusions.

Chapter 5 focuses on anthropogenic hazards (SO_2 emissions), and presents 3 case studies of risk and impact assessment carried out for large combustion plants (LCP) in Romania using optoelectronic techniques (UV cameras and SO_2 gas analyzers). This chapter introduces modelling as a crucial tool for risk and impact assessment, and more important, reveals the advantages of using modelling combined with optoelectronic techniques (UV cameras) in the retrieval of LCP emission rates. Chapter 5 also proposes and demonstrates through the case studies a new assessment methodology based on SO_2 emission rate retrieval with UV cameras, followed by dispersion simulations and validation with a SO_2 point monitor. This methodology is the base of an innovative strategy for risk and impact assessment for LCPs, involving authorities, local stakeholders and industry.

2. Theoretical considerations regarding hazards and risks

The knowledge of natural and technological risks is of the utmost importance for conducting risk and impact assessment studies, as well as land use planning and emergency response planning (Török et al., 2011c, Török et al., 2009). For the overall comprehension of this issue, it is highly relevant to define the recurring concepts of this work, namely hazard and risk. This chapter focuses on these two concepts and lays the foundation for the following applicative studies in chapters 4 and 5.

2.1. The hazard concept

In 2000, Ozunu defines the concept under discussion as „a situation with the potential of an accident"; in 2001, Bălteanu states that hazard is „a threatening event, representing the possibility for a potentially damaging phenomenon to happen." According to him, damage to people, goods and the environment occur.

A hazard is composed of three basic elements (Ericson, 2005):

1. Dangerous property – the basic source of the danger that creates the hazard, i.e. a dangerous energy source etc.
2. Initiation mechanism – the event that triggers or initiates the occurrence of the hazard, transforming the hazard from a passive state to an active one.
3. Target and threat – the person, object, situation vulnerable to damage caused by the materialization of the hazard.

These three elements compose the hazard triangle shown in figure 2.1:

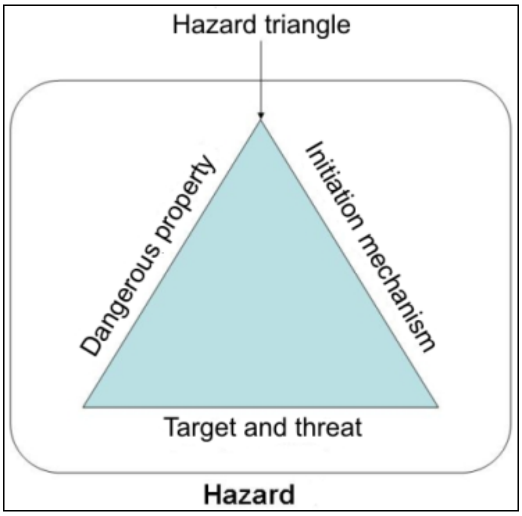


Figure 2.1 - The hazard triangle (Ericson, 2005)

2.2. Hazard vs. risk

The term ”hazard” is closely related with the term ”risk”. Not few are the cases when confusion arises between the meanings of these two concepts. Only when a hazard or phenomenon exceeds certain critical values, leading to material damage or casualties, does it become risk. Therefore, a hazard is the threat that an event might happen and not the event itself. Should it affect a human community, to a certain extent, it becomes risk.

The widely accepted definition of risk as the product between the probability for an event to happen and the negative consequences it may have is expressed as follows:

$$R= F \times C \qquad \text{(Eq. 2.1)}$$

where:

R- risk (losses/unit of time), F- frequency of occurrence (no. of events/unit of time), C- consequences (losses/event).

Another definition is given by Ozunu and Anghel in 2007:

$$R = F \times C \times V \qquad \text{(Eq. 2.4)}$$

where: R – risk; F- frequency; C – consequences; V- vulnerability (-).

2.3. Classification of hazards and risks

As previously mentioned, hazards and risks are defined and characterized in multiple ways. In this chapter, a classification is presented according to the most relevant characteristics:

- origin
- manifestation mode
- duration
- affected surface and duration of effects

The present thesis focuses on the assessment with optoelectronic technologies of two major hazards from the above mentioned classifications:

- **Natural hazards – volcanic ash by active and passive remote sensing**
- **Technological hazards – monitoring and modeling the impact of SO_2 emissions associated with large combustion plants.**

2.4. NATECH hazards

This thesis focuses both on natural and anthropogenic hazards separately. In order to develop efficient strategies for assessing risks and impact, the NATECH (natural hazards which trigger technological accidents) principle is necessary to be considered when assessing either of the two above mentioned types of hazard. Current EU regulations in the field of risk assessment and disaster management (European Commission, 2010) emphasize the necessity of a multi-risk and multi-hazard approach in all natural and anthropogenic hazard and risk studies.

Therefore, there is a growing interest in the scientific community and among stakeholders regarding natural hazards which trigger technological accidents (NATECHs). NATECHs have significant negative consequences on human health, the environment and the economy. The increase in the number of such events is closely linked with the exponential technological development of the past decades, due to the diversification of technologies, the growing number of personal exposed, and the substances used in the technological processes. The consequences of NATECH events have become more severe within this timeframe mainly due to the exposure of the population living near these facilities.

2.5. Qualitative and quantitative methods for identifying hazards and assessing risks

A systematic identification of possible environmental impacts atmospheric pollutants have, as well as a rigorous analysis of their magnitude is required. The purpose of such a process can be divided in two major issues, on one hand to produce quantitatively accurate estimation of particular risk and a comprehensive list of possible environmental impacts, and on the other hand produce a rationale for making public policy decisions that is both well reasoned, and recognized as legitimate and acceptable by the socio-economical factors.

2.5.1. Qualitative methods used in hazard analysis

A qualitative analysis implies the use of qualitative criteria, using different categories for parameters separation, with qualitative definition which establish the scale for each category. Also, qualitative decisions are made, based on the field experience, in order to assign elements into categories. This approach is subjective, but it allows a higher generalization degree, being less restrictive.

The identification of technological hazards is the basic step in risk assessment process. Hazards appear in the industry all the time, due to the process and operating conditions of the installations and the physical, chemical and toxicological properties of the substances used in these processes. This is why it is highly important to identify the substances' hazardous properties and the operating conditions that put at risk these processes, the series of events that may lead to the materialization of a hazard.

A preliminary hazard analysis is the starting point of any assessment and represents the most general type of hazard and risk assessment, resulting in most cases in qualitative risk matrixes (Table 2.7) describing risk as a product of frequency and consequences (Török et al., 2011b) (Eq. 2.1):

Table 2.7 – The risk matrix

			Consequences				
			Insignificant	Minor	Moderate	Major	Catastrophic
			1	**2**	**3**	**4**	**5**
Frequency	Improbable	**1**	**1**	**2**	***3***	***4***	***5***
	Isolated	**2**	**2**	**4**	***6***	***8***	***8***
	Occasional	**3**	**3**	**6**	***9***	***12***	***15***
	Probable	**4**	**4**	**8**	***12***	***16***	***20***
	Frequent	**5**	**5**	**10**	***15***	***20***	***25***

According to table 2.7 the risk level is qualitatively described as follows:

Table 2.8 – Risk levels

Risk levels	**Definition**
1 – 3	*Very low risk*
4 – 6	*Low risk*
7 – 12	*Moderate risk*
13 – 19	*High risk*
20 – 25	*Extreme risk*

2.5.2. Quantitative methods used in risk assessment

A quantitative analysis implies the use of numerical or quantitative data and provides quantitative results. This approach is more objective and more precise, but the precision is highly dependent on the validity of the input parameters. Therefore, the quantitative results within the risk analyses should not be taken into consideration as exact numbers, but as estimates, with a variable scale depending on data quality (Török, 2010).

The present thesis focuses mainly on the mathematical modeling of the gas pollutants' dispersion in the atmosphere. Source models are used to define the quantitative emission scenario of substances by estimating their flow rate, the dispersion of substance after release. Dispersion models turn the outputs from source models into isoconcentration curves delimiting concentration areas and calculate the evolution of concentration in time.

3. Theoretical considerations regarding optoelectronic techniques used in atmospheric monitoring

Optoelectronics is a field of technology that brings together the physics of light and electricity. Optoelectronics focuses on the conversion of photon signals into electrical signals and vice versa (Webopedia, 2012).

3.1. The atmosphere

The Earth's atmosphere can be represented as a layer extending from the surface upwards and fading out into interplanetary space, held to the surface by gravity (Ştefan et al., 2008).

The current study will limit itself to the composition of the homosphere (low and middle atmosphere), where most of the atmospheric mass is concentrated, and the majority of the processes occur. The Earth's atmosphere contains alongside gases, liquid and solid particles.

The atmospheric gas is composed of a mixture of individual gases, most important being nitrogen, oxygen and argon (US Standard Atmosphere, 1976).

Atmospheric aerosols are represented by liquid or solid particles, of variable sizes ranging from molecule clusters to several micrometers. According to IPCC, 2007 Climate Change Synthesis Report; Intergovernmental Panel on Climate Change, the anthropogenic contributions of aerosols have a cooling effect, the value of radiative forcing being equal with -0.5(-0.9 to -0.1) Wm^{-2}.

The most important characteristics of atmospheric aerosols include size, shape, chemical composition density and hygroscopic properties. A summary of aerosol sources, effects, and lifetime was presented by Holloway and Wayne, 2010 using data from Wallace and Hobbs, 2006 (Figure 3.4):

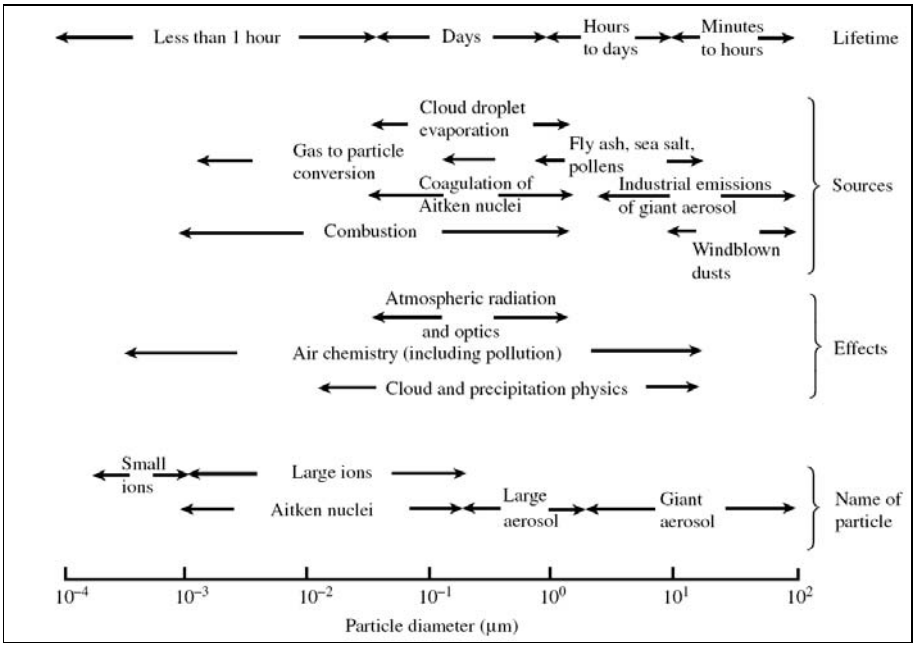


Figure 3.4 Atmospheric aerosol effects, sources, and lifetimes (Holloway and Wayne, 2010; Wallace and Hobbs, 2006)

The interaction of electromagnetic radiation with molecules and aerosols (Figure 3.6) results in a multitude of phenomena like: scattering (Mie - aerosols, Rayleigh - molecules and Raman), diffraction, refraction, fluorescence and reflection.

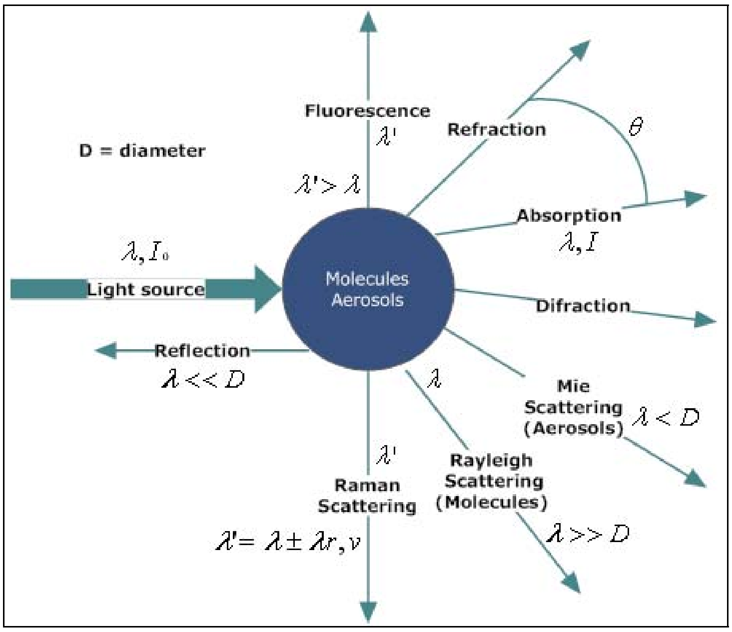


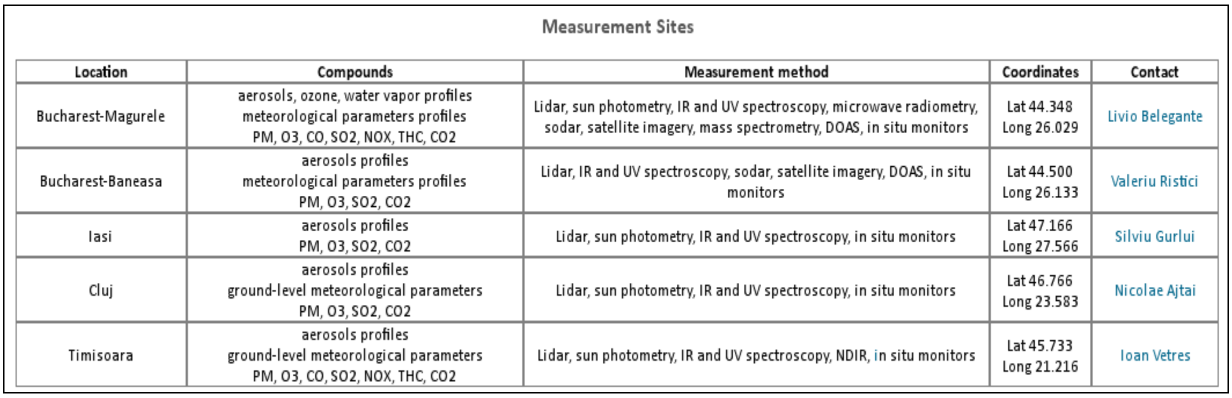
Figure 3.6 - Interaction of radiation with atmospheric compounds (adapted after Nicolae, 2006)

Relevant to this thesis and a key aspect in the processing of LIDAR and sun-photometer data, is the extinction of an incident beam of radiation through absorption and scattering processes. Both absorption and scattering occur at the same time because all materials scatter and absorb at least at molecular level.

3.2. The RADO (Romanian Atmospheric 3D research Observatory) concept

The RADO concept represents an innovative approach towards monitoring and analyzing atmospheric components and processes. The RADO network comprises instrumentation both for gases and aerosols monitoring in order to assess the important volume processes. The measurements available at sites the sites are presented in table 3.5.

Table 3.5 - RADO measurement sites (RADO, 2011):



Measurement Sites

Location	Compounds	Measurement method	Coordinates	Contact
Bucharest-Magurele	aerosols, ozone, water vapor profiles meteorological parameters profiles PM, O3, CO, SO2, NOX, THC, CO2	Lidar, sun photometry, IR and UV spectroscopy, microwave radiometry, sodar, satellite imagery, mass spectrometry, DOAS, in situ monitors	Lat 44.348 Long 26.029	Livio Belegante
Bucharest-Baneasa	aerosols profiles meteorological parameters profiles PM, O3, SO2, CO2	Lidar, IR and UV spectroscopy, sodar, satellite imagery, DOAS, in situ monitors	Lat 44.500 Long 26.133	Valeriu Ristici
Iasi	aerosols profiles PM, O3, SO2, CO2	Lidar, sun photometry, IR and UV spectroscopy, in situ monitors	Lat 47.166 Long 27.566	Silviu Gurlui
Cluj	aerosols profiles ground-level meteorological parameters PM, O3, SO2, CO2	Lidar, sun photometry, IR and UV spectroscopy, in situ monitors	Lat 46.766 Long 23.583	Nicolae Ajtai
Timisoara	aerosols profiles ground-level meteorological parameters PM, O3, CO, SO2, NOX, THC, CO2	Lidar, sun photometry, IR and UV spectroscopy, NDIR, in situ monitors	Lat 45.733 Long 21.216	Ioan Vetres

3.3. Passive remote sensing

Remote sensing can be generally described as a set of measurements made at a certain distance from the object in study. Information is usually carried by electromagnetic waves from the object to the observer (Lenoble, 1993).

In the case of passive remote sensing the source of the electromagnetic radiation is of natural origin (most of the time, the Sun). Dubovik et al. in 2000 describes aerosol features by measuring solar transmission and sky radiation with sun-photometers. The Cimel 318 A sun-photometer measures sun and sky radiances and retrieves important parameters like aerosol optical depth (AOD), Angstrom exponent, ozone concentration, water vapor concentration and also, through inversion, size distribution, complex refractive index and single scattering albedo can be computed.

3.3.1. The Cimel CE 318 A sun-photometer

The Cimel CE 318 A sun-photometer (Figure 3.11), described in Holben et al., 1998 is a ground-based sun and sky tracking automated radiometer that measures aerosol optical properties using a combination of filters and azimuth and zenith mobility. It measures sun and sky radiance in order to derive aerosol properties, ozone and total vapour column.

Figure 3.11 - NASA-AERONET #643 Cimel CE 318A sun-photometer
Faculty of Environmental Science and Engineering,
Babeş-Bolyai University Cluj-Napoca (Ajtai et al., 2012b)

3.4 Active remote sensing

For the quantitative assessment of aerosol layer distribution and concentrations, and in order to accurately determine the hazard associated with certain aerosol types (i.e. volcanic ash) through optoelectronic techniques. (Balin, 2004).The LIDAR principle is based on the emission of an electromagnetic pulse and receiving and analyzing the light backscattered towards a detector by aerosols or molecules. LIDAR systems are based on highly monochromatic lasers, controlled polarization, and a high energy per frequency unit of the spectrum (Belegante, 2011).

The simple MiniLidar System used in this thesis (Figure 3.18) gives the possibility to obtain profiles of the backscatter and extinction coefficients and vertical profiles.

Laser	Nd:YAG, 532 nm
Energy per pulse	4 µJ
Frequency	5 kHz
Duration of pulse	< 1 ns
Surface	314 cm^2
Total field of view	55 µrad
Filter band-pass	0.5 nm
Detection type	Photon counting
Acquisition time	> 0.8s
Vertical resolution	15 m
Size	220 x 1000 mm
Weight	12.5 kg

Figure 3.18 – The Cimel CAML CE 370-2 mico-LIDAR system

3.5 Point-monitoring using HORIBA APSA 370 SO_2 gas analyzer

The HORIBA APSA-370 is an ambient SO_2 gas analyzer based on UV fluorescence. The analyzer is based on the absorption spectroscopy principle (HORIBA, 2012). The atmospheric SO_2 concentrations determined using this analyzer will be compared with concentrations outputted by dispersion simulations in order to determine compliance with national legislation and finally determine the impact associated with the units emitting SO_2 in the atmosphere.

3.6. Sulphur dioxide emissions monitoring UV cameras

The SO_2 imaging camera developed by NILU uses the region from about 280 –320 nm, has a high quantum efficiency detector and operates from a laptop computer and 12V power supply (Stebel et al., 2012a).

Whenever SO_2 is present in the field-of-view of the camera, the recorded light intensity is less. By calibrating the camera using gas cells containing known amounts of SO_2, the recorded light intensity can be related directly to the path concentration (Prata et al., 2008).

The measurement procedure is based on the Beer-Bougier-Lambert law, which states that the attenuation of radiation through a layer of gas follows an exponential law that depends on the extinction coefficient for the gas.

4. The identification of natural hazards using optoelectronic techniques for atmospheric monitoring

This chapter focuses on the identification and characterization of atmospheric aerosols which are considered to be hazardous to human health, the environment, society and economics. The present approach is based on data retrieved by optoelectronic techniques, mainly LIDAR and sun photometry. The presented case study focuses on a natural hazard, volcanic ash, focusing on the detection and characterization of its optical and microphysical properties.

4.1. Atmospheric aerosols. Characterization and hazardous properties

The lack of detailed knowledge regarding optical and microphysical properties of aerosols results in difficulties in the assessment of the radiative forcing associated with them, but also of the hazards associated with certain aerosol types like volcanic ash, desert dust, biomass burning aerosol, and urban-industrial aerosol. AERONET data has the potential to narrow the uncertainties related to aerosol optical and microphysical properties (AERONET, 2010).

A fundamental step-forward in the characterization of optical properties of aerosols was made by Dubovik et al. in 2002. The study focuses on the characterization of aerosol properties in different worldwide locations and introduces a classification according to several optical parameters retrieved by the Cimel CE 318 sun photometer.

4.1.1. Urban-industrial aerosols

Urban emissions and the by-products of industries, along with exhaust products of motorised traffic are known as urban-industrial aerosols. Transport, fossil fuels combustion, metallurgy and waste combustion are some of the industrial sectors that lead to the formation of primary aerosol particles. Although not a natural source, the characterization of the optical properties of urban-industrial aerosols is important in any undertaking which focuses on the characterization of optical properties of other types of natural aerosols.

Recent estimations of current emissions of urban-industrial aerosols range between 100 Tg/yr and 200 Tg/yr (IPCC, 2007).

Regarding size distribution, AERONET data is more conclusive, with a pronounced fine mode concentration, resulting in $\omega_0(\lambda)$ decreasing with the increase of λ. The bimodal distribution and the predominance of the fine mode were also confirmed by in situ measurements (Hartley et al., 2000). Angstrom parameter values are also high, due to the domination of fine mode particles.

4.1.2. Biomass burning aerosols

Global aerosol emissions resulted from biomass burning was estimated between 45 and 80 Tg/yr. Biomass burning is also an important source of black carbon, with an annual injection of 6- 8 Tg/yr (IPCC, 2007).

Biomass burning aerosols also have a significant large–scale impact on radiative forcing and the climate system due to its high absorption. Other important hazards manifested at a more local level include effects on air quality and visibility (Reid et al., 2005). Human health can be affected in particular cases (Pope, 2004).

The majority of the hazards associated with biomass burning are determined by its size distribution. The particle size distribution for biomass burning aerosols is mainly dominated by the accumulation mode. The size of these particles increases the hazard they pose on human health, small particles penetrating deeper into the respiratory system.

The properties associated with small particle scattering can be observed (Dubovik et. al., 2002): high values of α (1.7 - 2.0); decrease of $\omega_0(\lambda)$ with increasing λ; pronounced decrease of the asymmetry parameter to relatively low values

4.1.3. Mineral dust

Dust has a major contribution to the loading the atmosphere with aerosols, contributing to the optical extinction of light through the atmosphere.

Human activities in urban areas can significantly propagate soil particles. It has been estimated (IPCC, 2007) that up to 50% of the atmosphere's loading with dust comes from land where human activity exists. Generally, the diameter of dust particles is around 2 - 4 μm.

The major difference between the optical properties of mineral dust and the previously mentioned biomass burning and urban-industrial aerosol is the clear bimodal representation and the predominance of the coarse mode in the size distribution (for AERONET data, $r > 0.6$ μm). Angstrom exponent also has much lower values, ranging from -0.1 to 1.2, with high phase function asymmetry for the wavelengths within the Angstrom parameter range.

Given the fact that mineral dust particles are larger and significantly less absorbing, the single scattering albedo $\omega_0(\lambda)$ increases or remains neutral with the increase of λ. This is a major characteristic employed in the characterization of a dust intrusion, with values ranging from 0.92 to 0.97 ± 0.2 (Dubovik et al., 2002).

4.1.4. Volcanic ash

Generally, the volcanic ash containing more than 75% pyroclastic fragments is referred to as tuff. The ash containing less than 75% pyroclastic particles is called tufit (Guffanti et al., 2009). Although volcanic ash has a relatively small particle size, the hazards associated with its presence in the atmosphere is not negligible.

Although ash deposits are a serious hazard to crops, and can also lead to clogging of the water surfaces and the collapse of poorly constructed buildings, one of the most important risk factors concerns aircrafts. Volcanic ash has a highly corrosive effect primarily on turbines as they get temporarily or definitively blocked after aspirating the dust which melts at the high temperatures inside the reactor and then cools down and solidifies on the cooler parts of the engine, causing sudden engine failure. Ash poses a hazard also to turbine blades and causes friction scratches on the windshield, resulting in poor visibility (Ajtai et al., 2010a).

4.2. Detection of volcanic ash using LIDAR and sun-photometers

Case study. The Eyjafjallajökull eruption in April and May 2010

This case study is based on measurements of volcanic ash generated by the Eyjafjallajökull volcanic eruption in April-May 2010 made at Laboratiore d'Optique Atmosphérique (LOA), Université Lille 1 Sciences et Technologies, France with a Cimel CE 318 sun-photometer and a Cimel CAML micro-LIDAR system and processed with LOA staff within a six month research stage. This research and monitoring facility enabled the detection and characterization of volcanic ash plumes over Lille region generated by the 2010 eruption of the Eyjafjallajökull volcano in Iceland (Mortier et al., 2012).

4.2.1. AERONET observations in April-May 2010, Lille. Detection and analysis

Standard measurements performed by the CIMEL CE-318, along with data retrieved through inversion and used in this case study are composed of:

- Aerosol Optical Depth ($\tau_{ext,a}$) within the range 340 to 1640 nm;
- Angström exponent, α, computed from spectral $\tau_{ext,a}$, $\alpha = d\ell n(\tau_a)/d\ell n(\lambda)$;
- sky spectral radiances in the solar principal plane and almucantar geometries;
- volume size distribution (*dV/dln r*);
- non-spherical particle fraction;
- spectral single scattering albedo (ϖ_o);
- spectral complex refractive index;
- extinction to backscatter ratio (LIDAR ratio), S_a;

The variability of $\tau_{ext,a}$, α, and non-spherical fraction during April-May period is shown in figures 4.3 and 4.4:

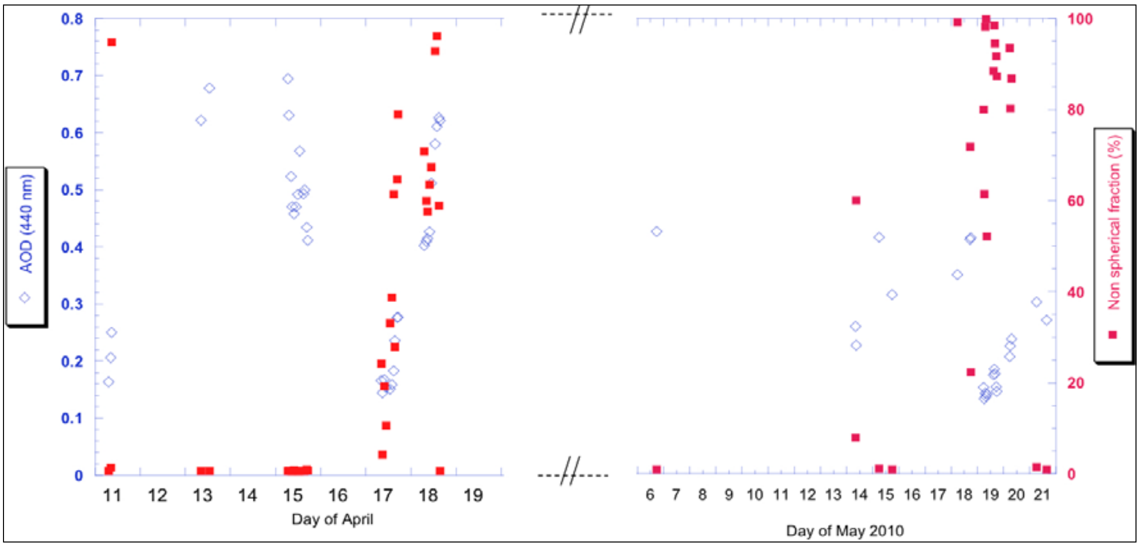


Figure 4.3 - Time series of $\tau_{ext,a}$ (440 nm) and (%) of non spherical particles for April and May 2010 over Lille

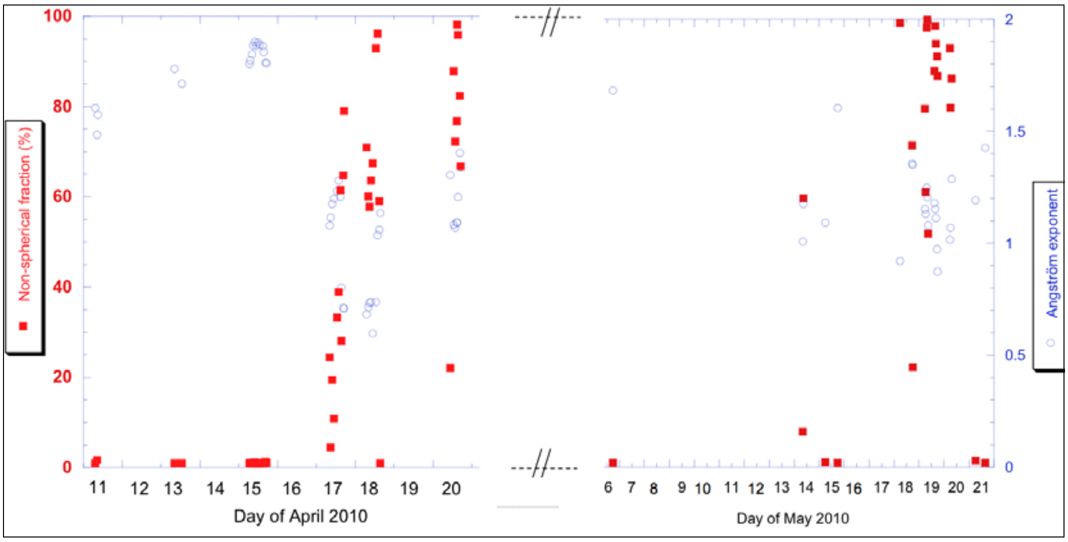


Figure 4.4 - Time series of Angström exponent, and (%) of non spherical particles for April and May 2010 over Lille

Strong changes in the size distribution retrieved from AERONET Level 2 inversion data are detected for the 15th, 17th and 18th of April, with an increase of the coarse particle distribution, as shown in figure 4.5:

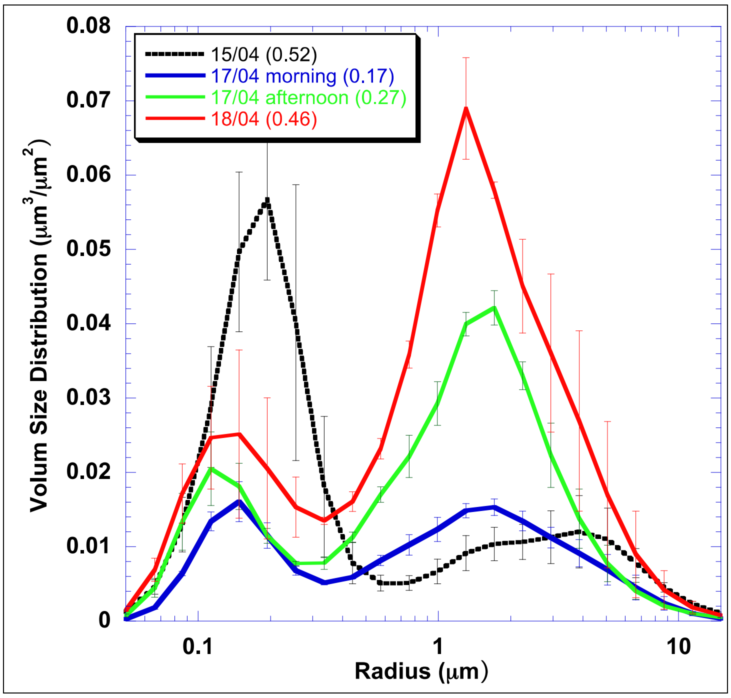


Figure 4.5 - Average size distributions (level 2) retrieved on 15, 17 and 18 April 2010. Error bars give standard deviation and indicate time variability.

The analysis of refractive index, spectral ϖ_o and LIDAR ratio indicate a modification in columnar aerosol chemical composition, which can either be explained by chemical processes and/or by a possible intrusion of an aerosol layer in the atmospheric column, at a location that cannot be located from AERONET data alone.

4.2.2. LIDAR observations in April-May 2010, Lille. Detection and analysis

LIDAR observations were made with a micro-LIDAR system operated continuously. The Cloud and Aerosol Micro-LIDAR (CAML) CE 370–2 is developed by the Cimel Company and has been extensively described in (Leon et al., 2009).

Several complex aerosol features distributed over one or multiple layers were detected from the 15th to 22nd of April 2010. In May, the volcanic ash plume was first detected between 3 and 4 km. The detection of aerosol layers in the atmosphere are confirmed in most cases by daytime detection performed with sun-photometers. In order to determine weather or not the volcanic ash is hazardous to air traffic, human health and the environment, a quantitative analysis is required in order to derive ash concentration from LIDAR measurements.

In order to minimize the impact of instrumental limitations, the extinction profiles retrieved for April 17th , April 18th, May 6th (Figure 4.12) were inverted with a fixed $S_{a,ash}$=34 sr. Moreover, for each profile, the difference between retrieved $S_{a,BL}$ and the background value retrieved from AERONET (75 sr) being a good indicator of the consistency of the approach (Mortier et al., 2012). The atmospheric layers are originating from Iceland, as shown by HYSPLIT back-trajectories (Draxler et al., 2012).

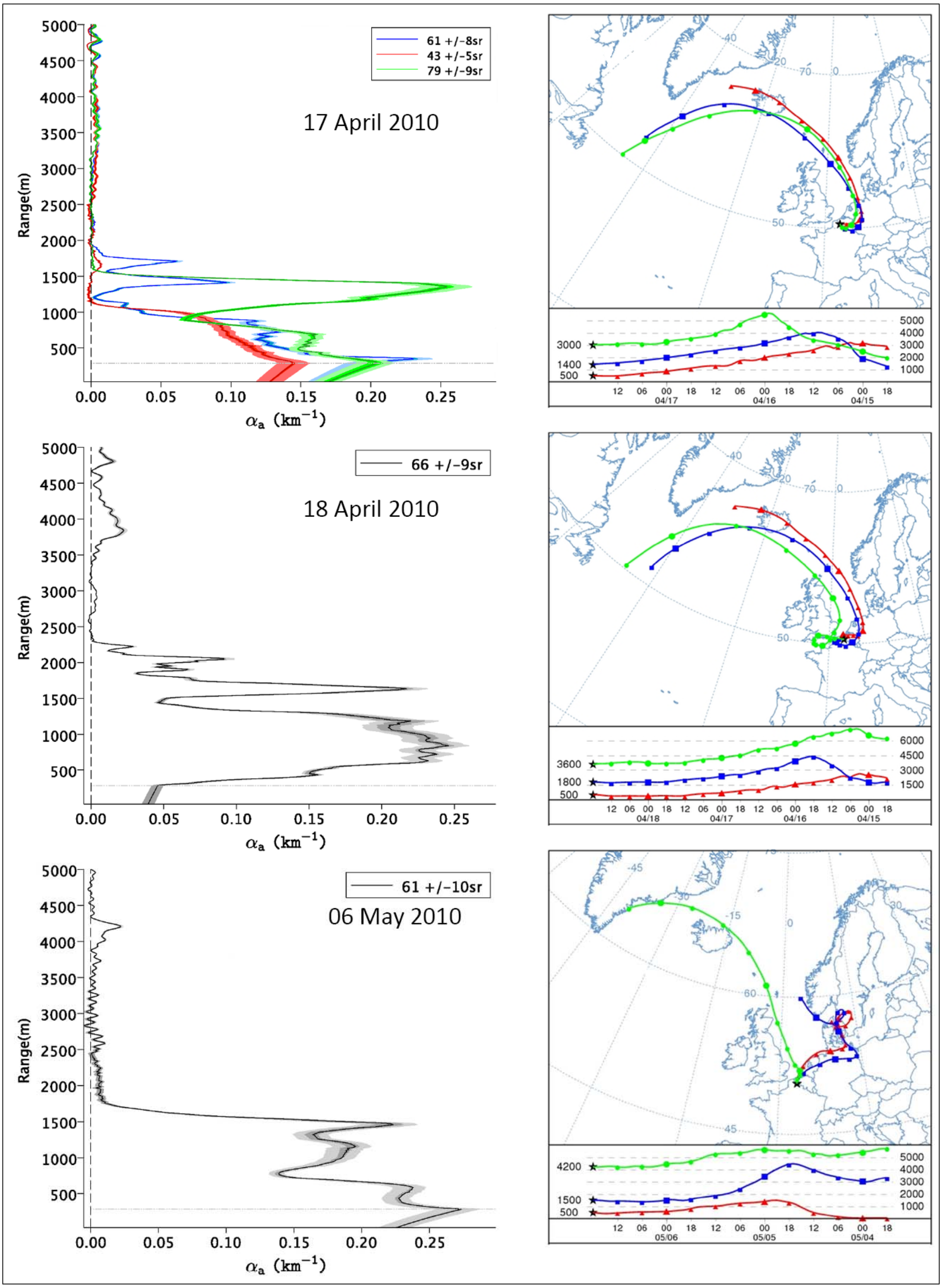


Figure 4.12 - Retrieved extinction profiles, $S_{a,BL}$ values (left)
and corresponding back-trajectories (right)

Figure 4.12 shows that in all situations single or multiple layers detected belong to air masses coming from Iceland region.

In order to determine the risk associated with the presence of ash particles in the atmosphere, an estimate of the mass of particles per unit of volume, C_a must be made. This value and the associated uncertainty are relevant for risk assessment for aircraft engines.

The mass of particles per unit of volume C_a for the maximum of the size distribution and at the altitude where σ_{ext} has the highest value was computed. These parameters are considered representative of volcanic ash during April and May 2010 in the current analysis.

Due to the fact that the extinction coefficient depends on the value of ash LIDAR ratio, minimum and maximum possible values of ash concentration, corresponding to 12 sr and 37 sr for $S_{a,ash}$ ($S_{a,ash}$+/-12)sr were also computed (Mortier et al., 2012). This is an essential step in for one to be able to use the results within a hazard analysis.

Results are summarized in table 4.1:

Table 4.1 – The main characteristics of volcanic ash layer detected over Lille in April-May 2010

Date	**Time (UTC)**	**Altitude (m)**	**τ_{ash}/τ**	**σ_{ext} (max) (1/km)**	**C_a ($\mu g/m^3$)**	**Min-Max ($\mu g/m^3$)**
17/04	06:52	1410	0.028/0.18	0.091-0.103	115	60-170
	11:57	1665	0.007/0.13	0.007-0.008	10	5-15
	18:58	1350	0.10/0.25	0.236-0.272	300	150-450
18/04	06:46	1495	0.16/0.36	0.29-0.33	360	180-540
	20:46	1640	0.08/0.30	0.20-0.23	260	130-390
06/05	18:21	4200	0.006/0.34	0.020-0.023	25	10-40
14/05	09:55	2760	0.07/0.20	0.20-0.22	240	120-360
15/05	06:45	2890	0.07/0.40	0.17-0.20	220	110-330
18/05	11:25	3260	0.08/0.23	0.13-0.14	160	80-240
	18:05	2775	0.13/0.29	0.14-0.15	170	85-255
19/05	07:25	2655	0.04/0.11	0.074-0.082	90	45-135
	12:05	2455	0.04/0.13	0.065-0.07	75	40-110
	18:05	2295	0.03/0.14	0.058-0.063	70	35-105
	20:05	1890	0.06/0.14	0.08-0.09	100	50-150
20/05	05h05	1960	0.08/0.16	0.15-0.16	180	90-270

Results show that the highest concentration levels were detected in April with values reaching 360 $\mu g/m^3$, whereas in May concentration decreased significantly, and after May 20th 2010 no more ash was detected due to the significant decrease in eruptive activity.

The maximum of ash concentration occurred during nighttime on April 17th 2010 (23:00 UTC). A semi-qualitative analysis based the LIDAR data retrieved shows that close to midnight, at least the double of daytime concentration was detected over Lille.

In May 2010, high values of ash concentration were detected several times during nighttime (14th, 15th, 18th and 19th of April). LIDAR data from May 14th and 15th, revealed high ash mass concentrations early in the morning, respectively in the afternoon, but AERONET data were available only for May 14th.

Concentration values exhibited variable ash concentrations, an estimation of these concentrations revealed **values ranging between 10 and 360 μg/m^3** during daytime, **much lower than the air traffic hazard limit of 2 mg/m^3** (VAAC, 2012).

4.3. Optimization of the detection of hazardous particles within the RADO network

This section of the thesis proposes a semi-quantitative methodology for the assessment of hazardous particle intrusions. This methodology is based on three major components:

- identification of a particle intrusion (qualitative)
- characterization of the optical and microphysical properties in order to determine weather or not the intrusion poses a hazard. (semi-quantitative)

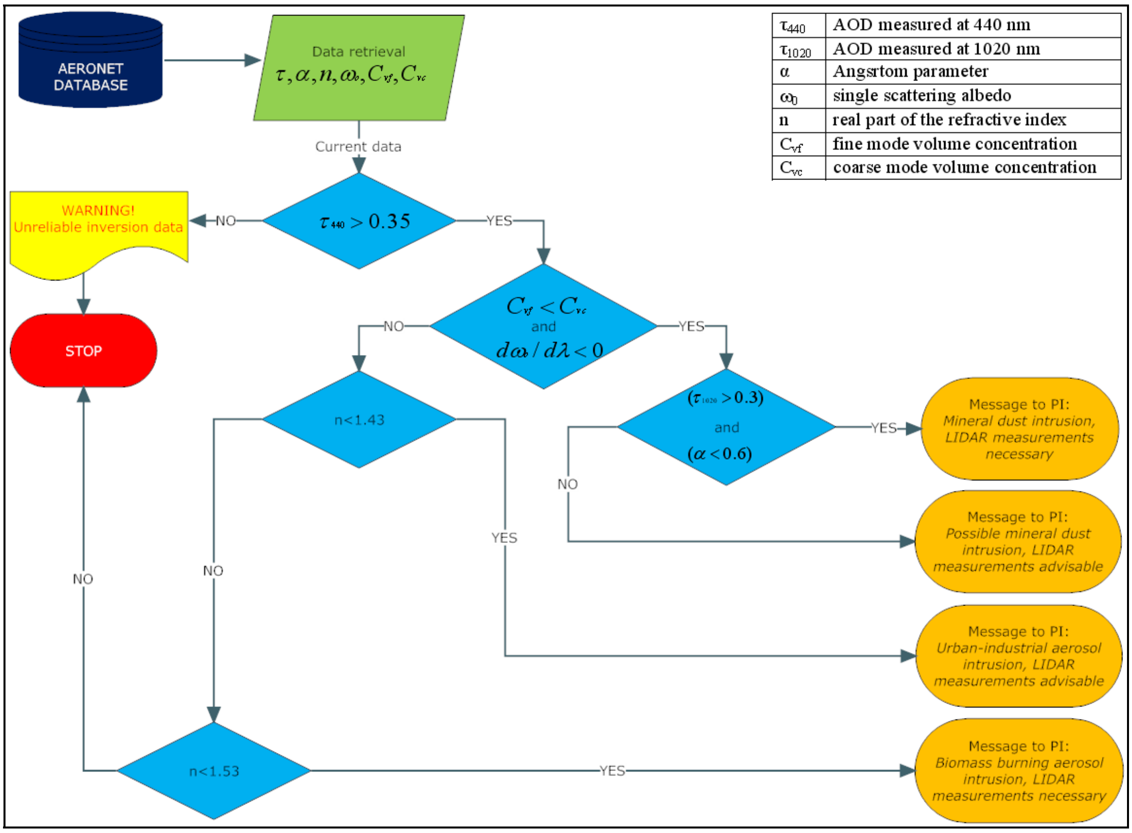


Figure 4.17 - Algorithm for hazardous particle characterization and optimization of non-continuous LIDAR measurements

- determination of concentration of hazardous particles (quantitative) a need demonstrated again the recent eruption of Grimsvoetn volcano, Iceland (Belegante, 2011, Unga et al., 2012)

5. The assessment of technological impact and risks using optoelectronic techniques for atmospheric monitoring

This chapter is dedicated to the assessment of the risk and impact associated with Large Combustion Plants (LCP), using optoelectronic techniques or environmental monitoring in combination with dedicated impact simulation software.

5.1. Large combustion plants

From a statistic point of view, large combustion plants are the main source of pollution with sulfur dioxide (approximately 67%) and nitrogen oxides (about 25%) responsible for the production of the acid rains and, respectively, soil and vegetation destruction, as well as for affecting human health.

Large combustion plants are defined by the Directive 2001/80/EC as “technical apparatus in which fuels are oxidized in order to use the heat thus generated with a rated thermal input equal to or greater than 50 MW, irrespective of the type of fuel used (solid, liquid or gaseous)”.

The Directive’s purpose is to limit the amount of sulfur dioxide, nitrogen oxides and dust emitted from large combustion plants each year. It encourages the combined production of heat and electricity (cogeneration).

5.2. Sulphur Dioxide (SO_2). Characterization and hazardous properties.

5.2.1. Characterization

The amount of anthropogenic sulphur emissions into the atmosphere is of ~80 TG/yr, which is more than the natural sources inject. Anthropogenic sulphur emissions are composed almost entirely of SO_2 emissions generated by burning coal and the smelting of sulfide rich ores (Wallace and Hobbs, 2006). The three case studies presented in this chapter focus exactly on these sources, two on coal-burning power plants, and one on the metallurgical industry.

Regarding the sinks, the main processes of removing sulphur dioxide from the atmosphere are wet and dry deposition. Around 65% of SO_2 is further oxidized to SO_4^{2-}; the rest is removed from the atmosphere by dry deposition.

5.2.2. Hazardous properties

Regarding the hazards associated, sulphur dioxide is a colorless nonflammable gas, with a pungent and irritating smell. It is classified as a hazardous substance with the following risk phrases (R-phrases): R23, R34, R50, and security phrases (S-phrases): (S1/2, S9, S26, S36/37/39, and S45). It is also classified under the Dangerous Substance Directive (Directive 67/548/EEC) as toxic (T):☠, with a lethal dose for a 30 minute exposure (LD_{50}) of 3000 pm for mice, with an IDLH (immediately dangerous for life and health) level set by NIOSH (National Institute for Occupational Safety and Health, 2012) at 100 ppm.

The presence of SO_2 in the atmosphere has adverse effects on the environment and human health, resulting in the irritation of the respiratory tract in case of low concentrations, and thoracic spasms in case of increased concentrations (Lăzăroiu, 2005). Studies have shown that exposure to SO_2 can lead to disruptions in the metabolism of sugars and other enzymatic processes (Mihăilescu, 1975).

SO_2 emissions can have a local effect, but can also be subject to long-range transport and deposition, by adhering to dust particles and aerosols, thus increasing their negative impact.

5.3 Modeling and monitoring of SO_2 emissions associated with Large Combustion Plants

The dispersion simulations for all three case studies were performed using the ISC AERMOD View software package, using the Industrial Source Complex Short Term (ISCST3) model which is a Gaussian plume dispersion model (Figure 5.4) that outputs scenarios predicting air concentrations around point or area sources using emission rates (flux) and meteorological conditions as model inputs (ISC AERMOD View, 2011).

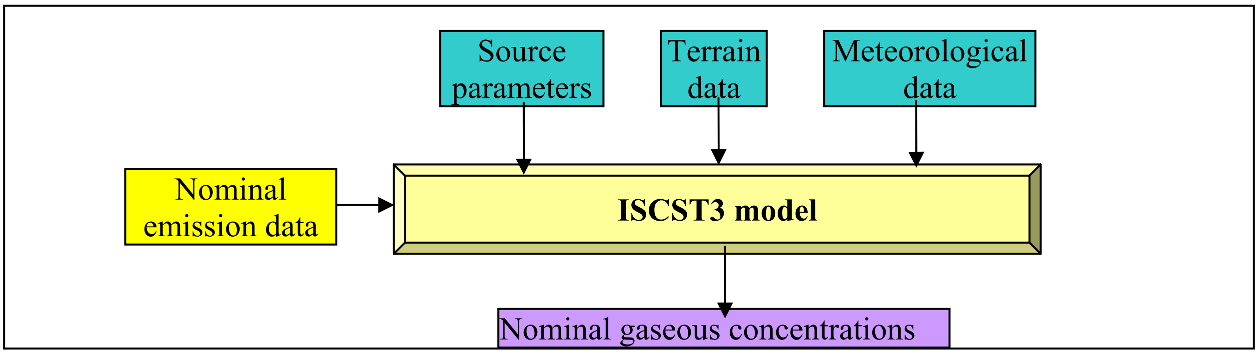


Figure 5.4 - Model framework to predict nominal gaseous concentrations (adapted from Xu et al., 2008)

The results are presented below for 3 case studies:

- **Case study 1. Modeling and Monitoring of SO_2 emissions from Mintia Large Combustion Power Plant**

  This case study was selected in order to describe the technological process of a LCP and the input parameters used for the SO_2 dispersion simulations: source parameters, meteorological data, and terrain data. These aspects will not be detailed for the other two case studies, the principle being the same, the focus being on the results obtained and the particularities of each separate case.

- **Case study 2. Modeling and Monitoring of SO_2 emissions from Rovinari Large Combustion Power Plant**

  This case study focuses on optoelectronic data gathered within the 2010 campaign at Rovinari, organized by The Romanian Atmospheric 3D Research Observatory. The focus is on the retrieval of the emission rate of a SO_2 source with UV cameras followed by dispersion simulations with the calculated emission rate, and a comparison with in-situ SO_2 imissions measurements made with a HORIBA APSA-370 SO_2 monitor.

- **Case study 3. Modeling and Monitoring of SO_2 emissions from a metallurgical plant**
  This case study was selected due to the possibility of comparing the impact of SO_2 emissions before and after the installation of a desulphurization technology. This case study brings strong evidence through the use of modeling coupled with optoelectronic point-monitoring that the use of best available technologies (BAT) previously described are a very efficient way to significantly reduce the impact associated with SO_2 emissions of large combustion plants (Mihăiescu et al., 2011).

5.3.1 Case study 1. Modeling and monitoring of SO_2 emissions from Mintia Large Combustion Power Plant

The results computed ISC AERMOD View – dispersion model ISCST3 show a significant impact of SO_2 emitted by Mintia LCP, in some areas, the maximums exceeding the legal limits (350 μg/m^3 – hourly limit, and the 125 μg/m^3- daily limit).

By overlapping the concentration map with the geo-topographic map, one can observe that the SO_2 plumes reach considerable distances on a radius of 15-20 km from the LCP. Due to the height of the stacks (220 m) and the complex topography of the area (Mures Valley, high hills nearby) the pollutant tends to accumulate in these areas, especially in unfavorable meteorological conditions for dispersion (thermal inversions, increased atmospheric stability). On the concentration maps it is worth mentioning the peak concentrations computed for the hills in the immediate vicinity of the LCP, which are at roughly the same level with the stacks (Ajtai et al., 2012c).

The ISCST3 model outputs two types of results:

- the dispersion plume for the first hourly maximum computed
- the dispersion plume for the first daily maximum computed

The dispersion map presented (Figure 5.6) in the following section for the first hourly maximum computed of 3621 μg/m^3 SO_2 on 21.09.2010, at 08:00 PM;

One can observe that the hourly legal maximum of 350 μg/m^3 SO_2 is exceeded in all three cases by an order of magnitude in the area behind the LCP. This area is characterized by hills with elevations between 300 and 680 m ASL. The elevation of the emission source is at roughly 410 m ASL, lower than the surrounding landscape, therefore explaining the presence of high concentrations in this area.

Due to the wind patterns generated by the complex topography represented by the Mures Valley and the surrounding hills, the Mures Valley is subject to dispersion favoring conditions resulting in lower SO_2 concentrations along this valley. These assumptions are generally valid in normal weather conditions, but can vary significantly in case of inversion phenomena or extremely stable conditions.

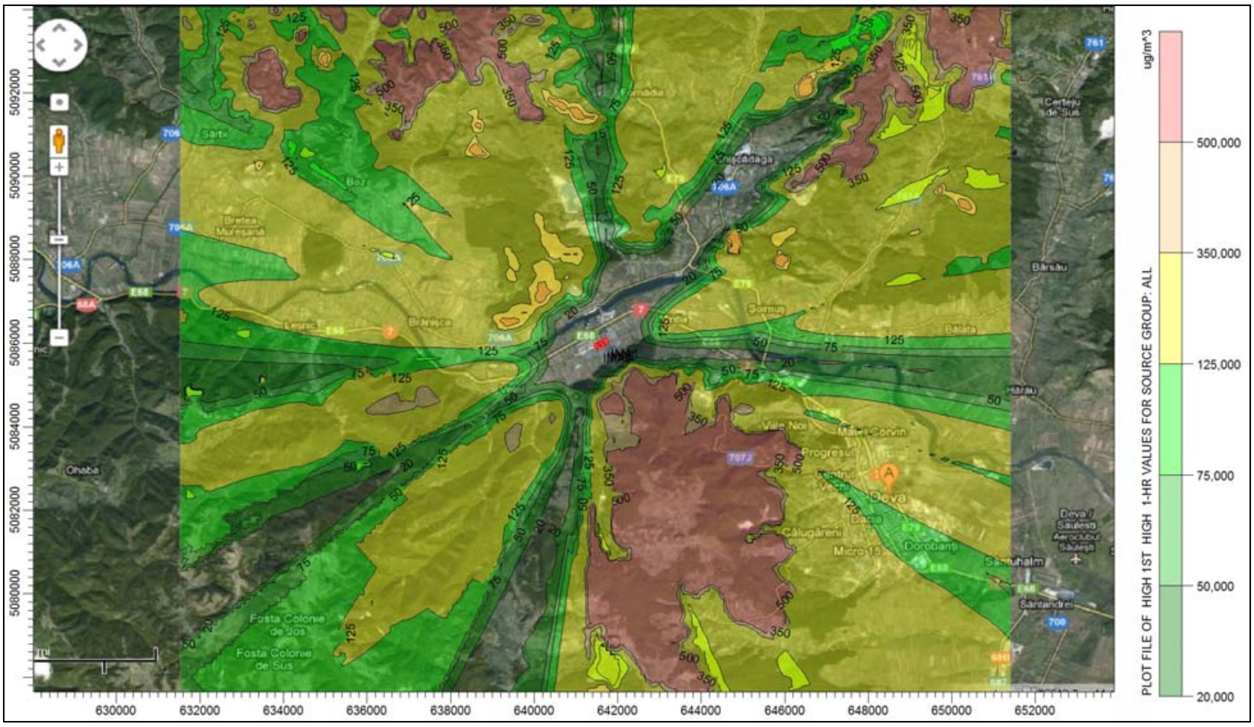


Figure 5.6 - First hourly SO_2 maximum – 3621 µg/m^3 SO_2

In case of daily maximums, one can notice again the difference of an order of magnitude between the computed concentration (1464 µg/m^3 SO_2 on 16.09.2010) and the legal limit of 125 µg/m^3 SO_2. These values are again computed for the elevated area behind the LCP. These values are though lower than the hourly maximums due to the natural turbulence processes that favor dispersion.

The hourly and daily concentration maximums computed mentioned above represent the absolute maximum concentrations reached in a point on the map. In order to better establish the impact associate with Mintia LCP, the same types of daily and hourly maximums we computed for two receptor points corresponding to the locations of the SO_2 gas monitors HD-1 and HD-2, located near the city of Deva.

Using the SO_2 imission data gathered from HD-1 and HD-2 monitoring stations of the Hunedoara Environmental Protection Agency, measured with HORIBA APSA 370, in the period 1-30 September 2010, the trends and maximums for both stations are presented. SO_2 concentrations are presented as hourly averages of the entire measurement period.

Regarding HD-1 monitoring station, three maximums were observed, all within the 125 µg/m^3 daily legal limit, with the first concentration maximum of 22.24 µg/m^3 on September 27th.

At HD-2 station, the SO_2 monitor identified maximums which do not exceed the daily legal limit of 125 µg/m^3 , with the first concentration maximum of 54.07 µg/m^3 on September 15th.

A qualitative correlation can be made between the daily concentration maximums computed and the daily mean concentrations recorded at HD-1 and HD-2.

By comparing these results periods of maximum SO_2 concentrations can be observed on the 5th - 6th, 15th -17th and 27th -28th of September 2010.

The modeling inconsistencies revealed by this case study can be most likely attributed to the incorrect estimation of the emission source's strength and temporal variation. Without accurate emission rates, most of the simulation studies must use a mediated emission rate, which may differ significantly from the real situation as demonstrated in this case**.** **A need arrises for the development of a more felxible, independent and mobile technique for the determinationof variable SO_2 emission rates.** The following case study proposes a method to eliminate these shortcomings.

5.3.2. Case study 2. Modeling and monitoring of SO_2 emissions from Rovinari Large Combustion Plant using UV-camera derived emission data

The data from this case study was collected within a 2 week field campaign in Rovinari (SW of Romania) of the Romanian Atmospheric 3D Research Observatory (RADO, 2012). This area was chosen because it is one of the largest SO_2 sources in Europe, a statement confirmed also by OMI satellite imagery (Figure 5.15).

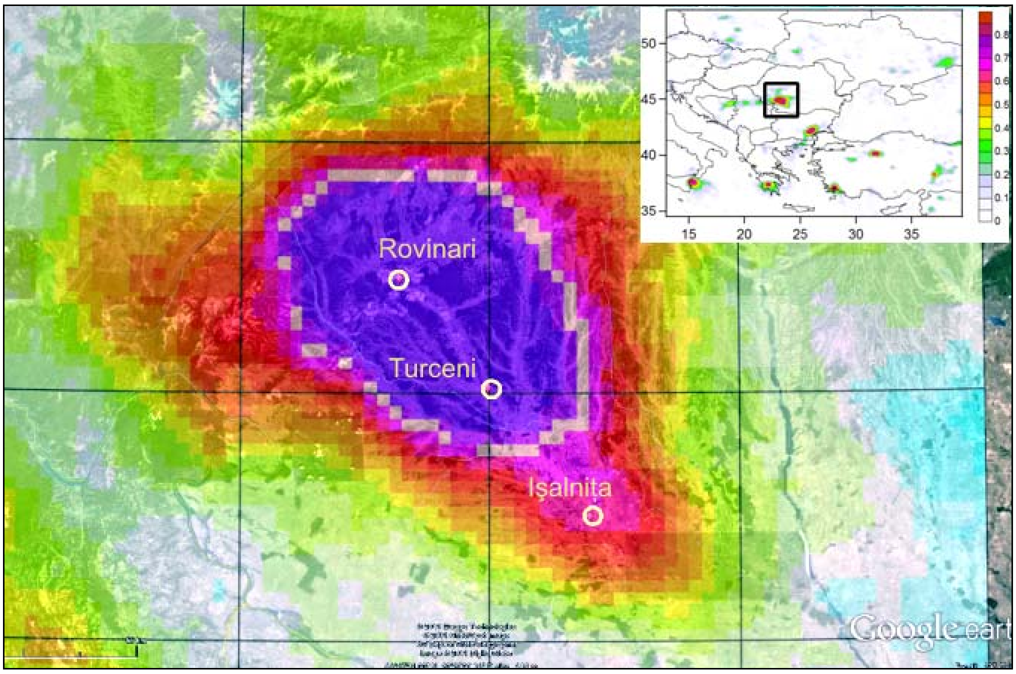


Figure 5.15 –Satelite imagery - OMI SO_2 in Dobson Units (DU)
from 2005 to 2011 (Fioletov et al., 2011)

In order to determine the emission rate of the LCP stacks an UV imaging camera was used. The day chosen for the retrieval of the emission rate was September 13th, due to the quality of the imagery and the peak concentrations of SO_2 measured by the HORIBA APSA-370 gas analyzer on that day.

Apparent absorption is retrieved using the background and average calibration constant retrieved and converted to SO_2 path concentration

$$AA = -\ln\left[(IP_{310} / IB_{310}) / (IP_{330} / IB_{330})\right] \qquad \text{(Eq. 5.9)}$$

where:

AA is apparent absorbance; IP is the plume image with dark image subtracted; IB is the background image with dark image subtracted;

In order to derive fluxes, the vertical plume speed (v_p) has been retrieved from consecutive images, by analyzing the difference in plume altitude when there is a clear feature identified. By dividing the two images by each other, gradients are retrieved.

The fluxes are then calculated by integrating the path concentration across the plume (w) and multiplying it with the determined vertical plume speed (Figure 5.19b):

$$w = \int_{x_0}^{x_1} u(x)dx \ [\mathrm{gm}^{-1}] \qquad \text{(Eq. 5.10)}$$

The x coordinate is determined from the measurement geometry, by correlating the GPS positions of the camera, the stack, along with elevations and distance between camera and stack and stack height.

In our case images were taken with a 50 mm lens with a 15.15 degrees field of view in x- as well as in the y-direction. This translates to a pixel size of about 50 cm for images for the 2 km distance between the campaign site and the plume.

As previously mentioned emission rate E (g/s) (Figure 5.19c) is obtained by multiplying the integrated path concentration across the plume (w, gm^{-1}) with the wind speed (v_p, ms^{-1}):

$$E = w * v_p \ [\mathrm{g/s}] \qquad \text{(Eq. 5.13)}$$

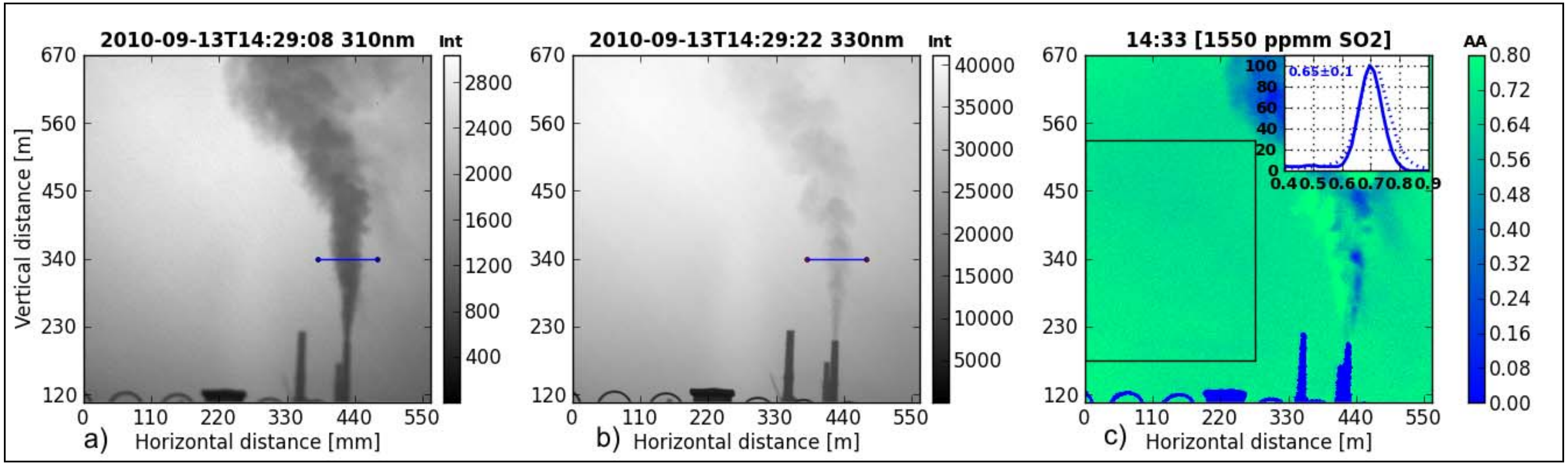


Figure 5.18 - Raw images – 310 (a) and 330 (b) nm filters (Filter FWHM = 10 nm), corrected for dark counts. (c)Illustration of the retrieval of the calibration values (Stebel et al., 2012a)

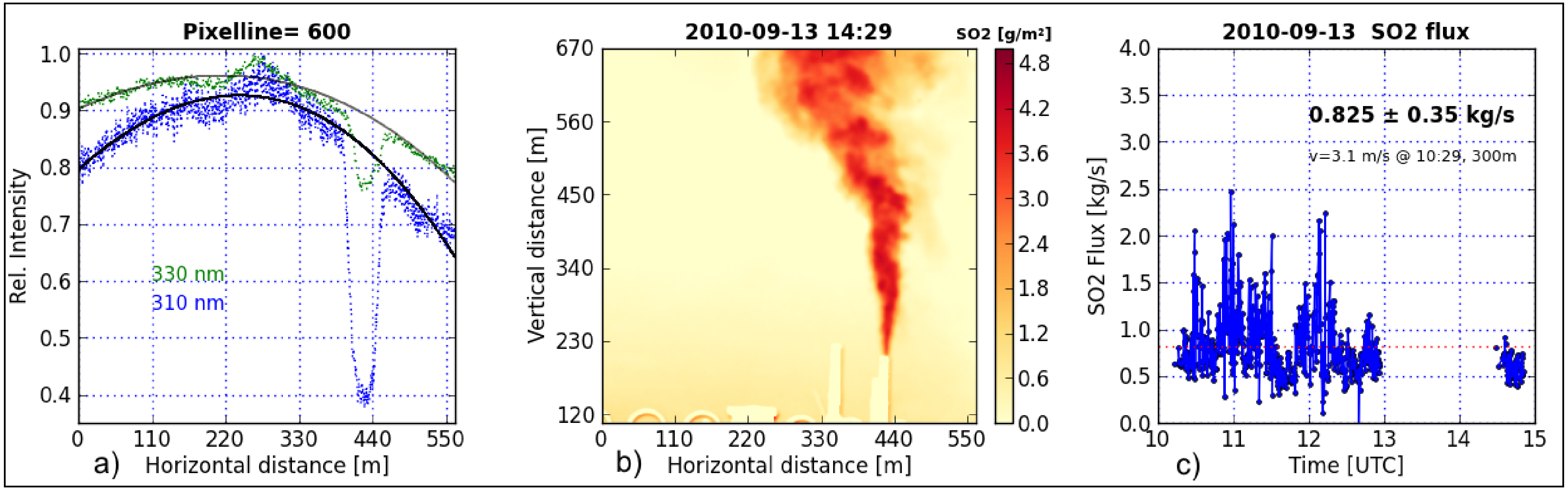


Figure 5.19 - (a) illustration of the retrieval, (b) apparent absorption retrieved (g/m^2), (c) vertical plume speed retrieved (Stebel et al., 2012a)

The computed SO_2 emission rates are presented in the table bellow for the 13th of September 2010 between 10:12 and 14:51 UTC.

Table 5.8 - Calculated hourly emission rates

Date / Time	**SO_2 emission rate (g/s)**
13/09/2010 10:12-11:00 UTC	925 g/s ± 400 g/s
13/09/2010 11:00-12:00 UTC	825 g/s ± 350 g/s
13/09/2010 12:00-12:56 UTC	775 g/s ± 325 g/s
13/09/2010 14:29-14:51 UTC	500 g/s ± 125 g/s
13/09/201010 10:12-14:51 UTC	**average emission rate = 825 g/s ± 350 g/s**

The **average emission rate retrieved via the UV cameras** of **825 g/s** [475 – 1175 g/s] from **only one emission source active** on the day of the measurements, agrees well with the **reported total average emission rate of 2321 g/s** [1160 - 3243 g/s] **for both sources.**

SO_2 imissions monitoring was performed at the Rovinari campaign site with the HORIBA ASPSA-370 SO_2 analyzer at 2.13 km from the Rovinari LCP.

The results for the 4-13 September 2010 are presented in Figure 5.20 (Ajtai et al., 2011b):

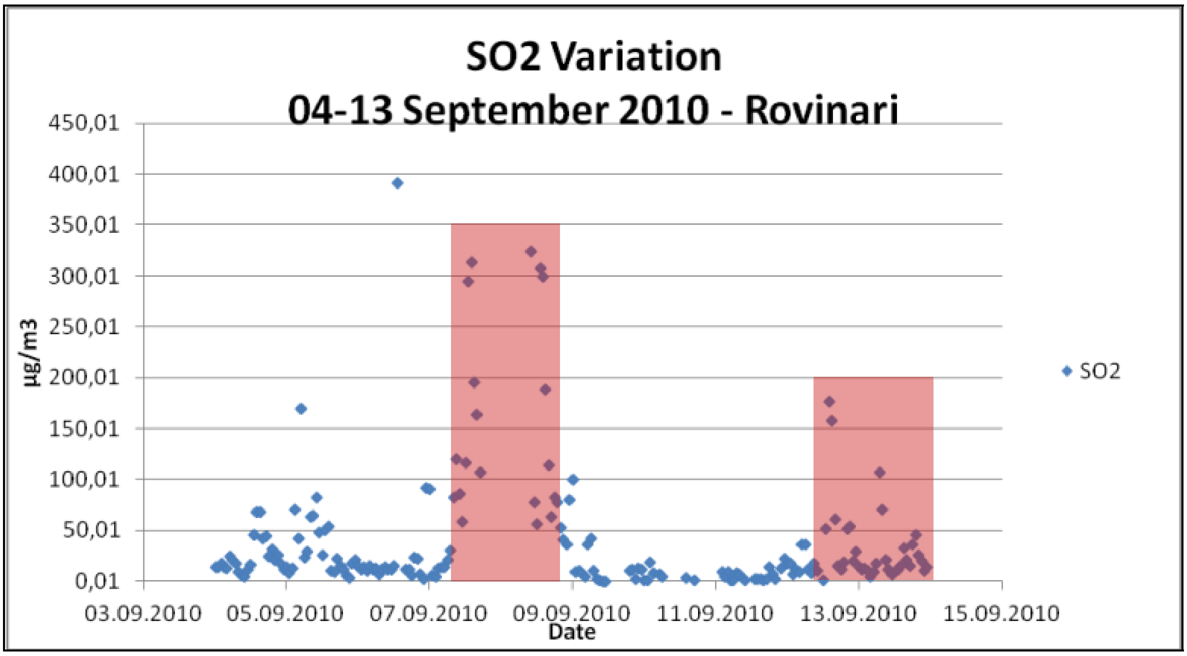


Figure 5.20 - Hourly SO_2 concentrations (µg/m^3) measured at the Rovinari campaign site

The UV camera derived emission rates of the LCP, along with other parameters such as meteorological data, terrain data and source information like stack height form the input dataset for dispersion modeling using AERMOD View's Industrial Source Complex Short Term (ISCST3) model, described in the previous case studies.

After performing dispersion simulations with the above mentioned input data the following dispersion plume was plotted. (Figure 5.21) Hourly values do not exceed the 350 µg/m^3 legal threshold values.

Results also include the concentration outputted for the point where the HORIBA APSA-370 SO_2 monitor was located (670623.00, 4974075.00).

Simulations were performed using the following emission rates:

- computed emission rate (Figure 5.21), with concentrations for the campaign location of **22.74 µg/m^3**
- computed emission rate with the maximum error added, with concentrations for the campaign location of **32.58 µg/m^3**
- computed emission rate with the maximum error subtracted, with concentrations for the campaign location of **12.90 µg/m^3**

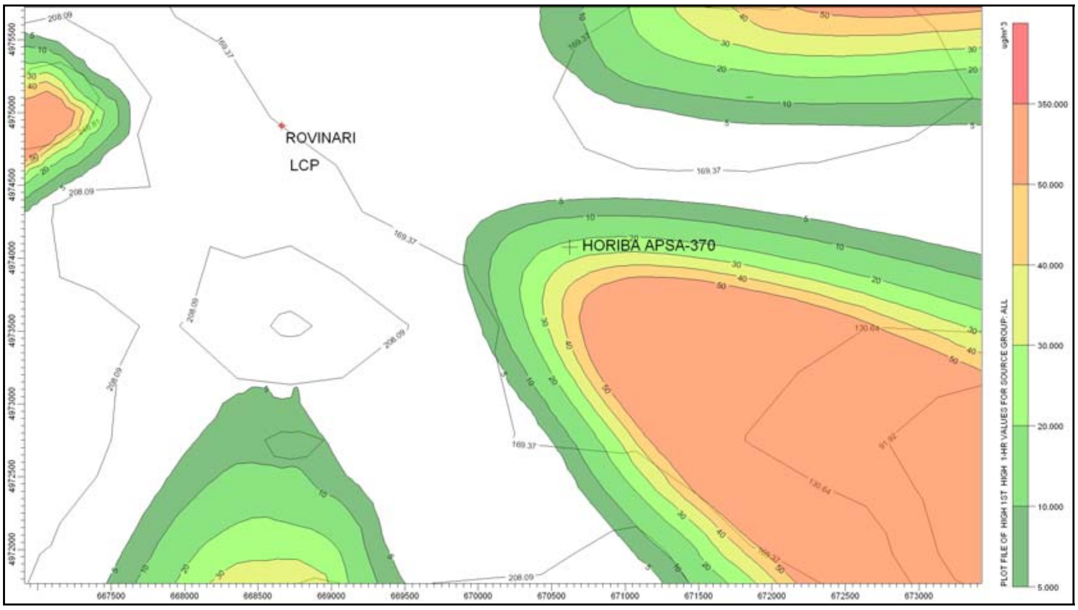


Figure 5.21 - First highest hourly concentration (µg/m^3) values for 13.09.2010 using computed emission rate

The results of the comparative study between measured and simulated imissions concentrations are presented in table 5.10:

Table 5.10 - Comparison between simulated values and HORIBA APSA-370 measurements at the Rovinari campaign site

Date/Time	HORIBA SO_2 concentration measured	Concentration outputted for the point where the HORIBA APSA-370 SO_2 monitor was located with		
		computed emission rate	**maximum** emission rate	**minimum** emission rate
13.09.2010 10:00 – 11:00	**10.69 µg/m^3**	**22.74 µg/m^3**	**32.58 µg/m^3**	**12.90 µg/m^3**

In conclusion, we can state that the dispersion simulations correlate well with the HORIBA ASPA-370 measured data, having the same size factor. This correlation is an important step in the validation of UV cameras for industrial emission rate retrieval, and demonstrates the validity of the approach based on optoelectronic techniques (UV-imaging-camera –SO_2 gas analyzer) combined with modeling software, in the present case, ISC AERMOD.

5.3.3. Case study 3. Modeling and monitoring of SO_2 emissions from a metallurgical plant before and after the installation of a desulphurization system

This case study was selected in order to reveal an important aspect in the management of industrial SO_2 emissions, regarding the implementation of best available techniques for the reduction of the impact associated with the above mentioned emissions. The case study focuses on demonstrating the reduction of the impact of SO_2 emissions of a metallurgical plant by performing dispersion simulations, analyzing imissions and compliance with national SO_2 emissions levels monitoring before and after the introduction of a desulphurization system.

The simulation results regarding to the SO_2 dispersion before the installation of the desulphurization system show maximum ground level imission concentrations much higher than the limits established in the G.E.O. No. 592/2002 for the protection of human health for 1, 3 and 24 hr averages. The maximum concentrations are shown in table 5.11.

Table 5.11 - Maximum imission concentrations obtained before desulphurization

	Maximum concentration [μg/m^3]	**Limit value for protection of human health (G.E.O. No. 592/2002) [μg/m^3]**
1 hr average	2880	350
3 hr average	1136	125
24 hr average	337	20

These maximum concentrations were obtained in stable (Pasquill class 6) atmospheric conditions in different imission points. Figure 5.24 shows in red and orange the areas where concentrations are higher than the 1 hr average limit of concern established (C > 350 μg/m^3).

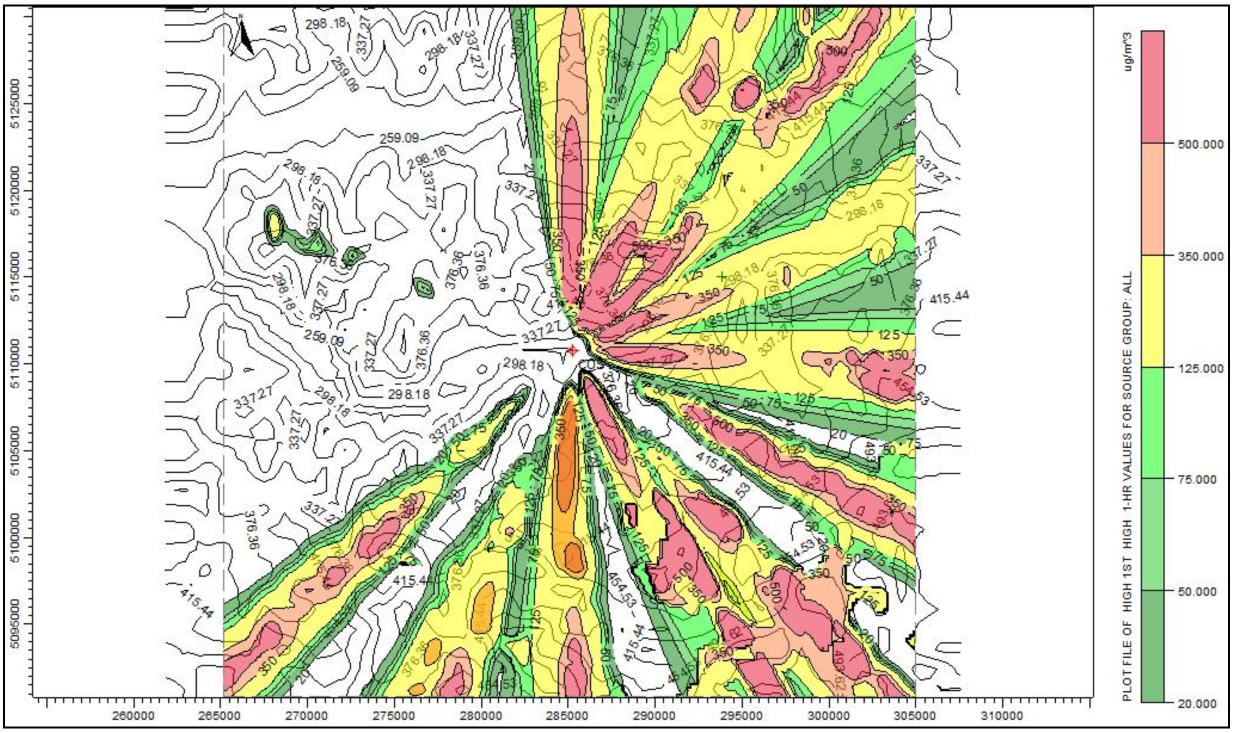


Figure 5.24 - Maximum 1 hr average concentrations obtained before desulphurization process

Simulation results were qualitatively correlated with imission concentration data measured at an automatic HORIBA APSA-370 point monitor situated at 8 km from the emission source. The maximum 1hr average concentration obtained in simulation for the monitoring receptor point is 253 μg/m^3. It represents the maximum concentration that was reached using a 1 hour averaging period.

Comparing the simulation results with the measured ones, it can be observed that the values have the same order of magnitude, but the simulation results show slightly lower values.

The simulation results regarding to the SO_2 dispersion after the installation of the desulphurization system show maximum ground level imission concentrations approx. 20 times lower than the limits established in the G.E.O. No. 592/2002 for the protection of human health for 1, 3 and 24 hr averages. The maximum imission concentrations are shown in table 5.13:

Table 5.13 - Maximum imission concentrations obtained after desulphurization process

	Maximum concentration [μg/m^3]	**Limit value for protection of human health (G.E.O. No. 592/2002) [μg/m^3]**
1 hr average	16	350
3 hr average	6	125
24 hr average	1	20

In figure 5.28 the situation of 1 hr average imission concentrations after the installation of the desulphurization system are represented.

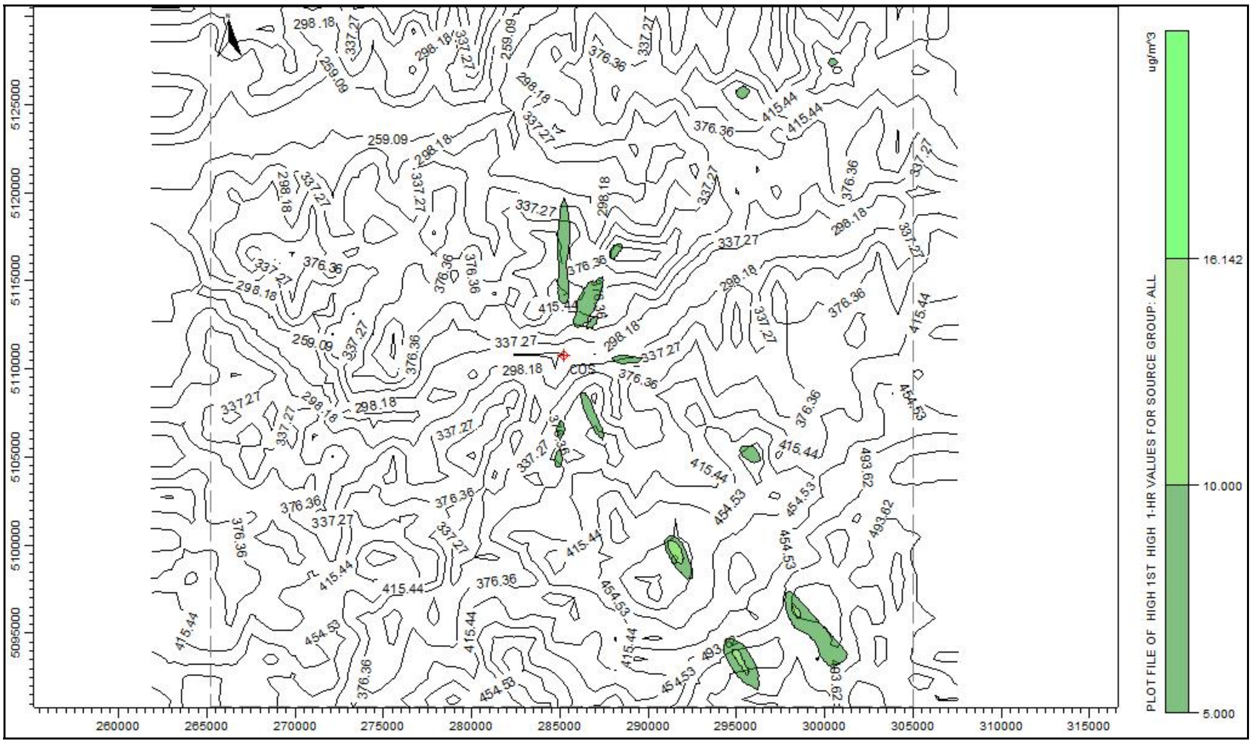


Figure 5.28 - Maximum 1 hr average concentrations obtained after desulphurization process

The simulation results, obtained for the situation before the installation of the desulphurization system, were compared with point monitoring data, showing a good qualitative correlation (Ajtai et al., 2012a). The simulations performed for the conditions after the installation of the desulphurization system show a significant reduction in the ground level concentrations of SO_2, well within the range imposed by national and European legislation.

It can be concluded that the installation of desulphurization system reduced the SO_2 emissions and imissions significantly and the obtained concentration values are well below the limits imposed by national and EU legislation.

5.4. Innovative strategy for assessing the impact of SO_2 emissions associated with LCPs

This chapter proposes an innovative strategy for the assessment of risk and impact associated to atmospheric pollutants emitted by large combustion plants. This strategy employs the methodology based on optoelectronic systems, more specifically, UV cameras and UV-fluorescence gas analyzers in close relation with dispersion modelling, described in the three case studies detailed in the previous sub-chapter (Figure 5.29). The goal of this undertaking is to provide a better understanding of specific risks and impacts associated with LCPs by developing a new and original atmospheric risk and impact assessment strategy, based on a methodology that integrates dispersion simulations and advanced optoelectronic techniques.

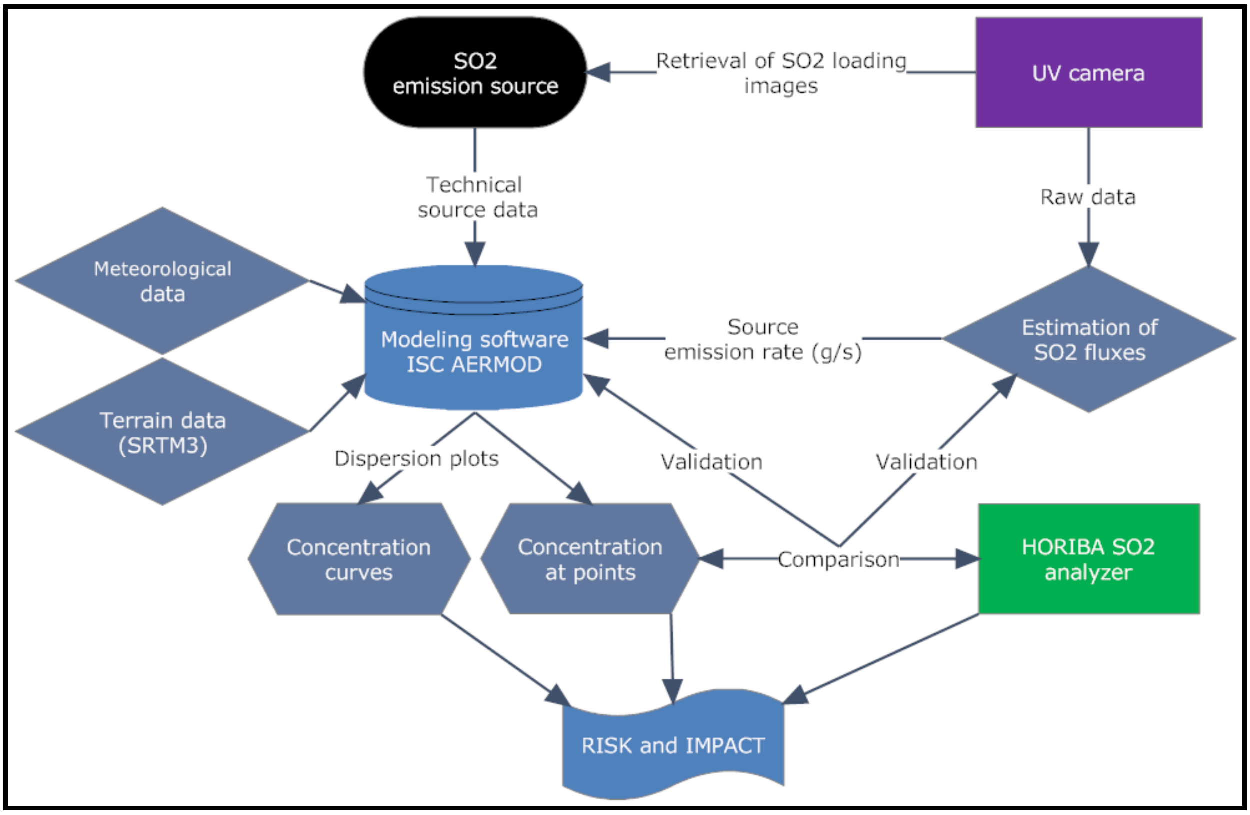


Figure 5.29 - Methodology for risk and impact assessment based on optoelectronic monitoring techniques and modeling tools

This synergy of point monitoring, plume profiles supplemented by modelling results represents a unique potential for advanced atmospheric studies and realistic impact and risk assessment.

Another major objective of the current strategy is to achieve a better compliance with the local or regional policies helps in the identification of the associated policies and institutions that can support the implementation of the strategy. Furthermore, the strategy proposes long-term targets and interim goals, and provides the guide steps for its implementation through efficient policies.

The strategy is built upon close sector cooperation and engagement with all relevant stakeholders, which provides an opportunity to discuss strategy ideas and policy proposals. The involvement of a broad range of stakeholders is essential for the development of a successful atmospheric risk and impact assessment strategy. The following groups of stakeholders will each have an important role in the development of appropriate strategies (Figure 5.30): experts in air quality and risk and impact assessment; national government agencies and legislative bodies, and local authorities; industry; intermediate groups which can help advocate for pollution reduction campaigns; those whose interests are directly affected by air quality; local community.

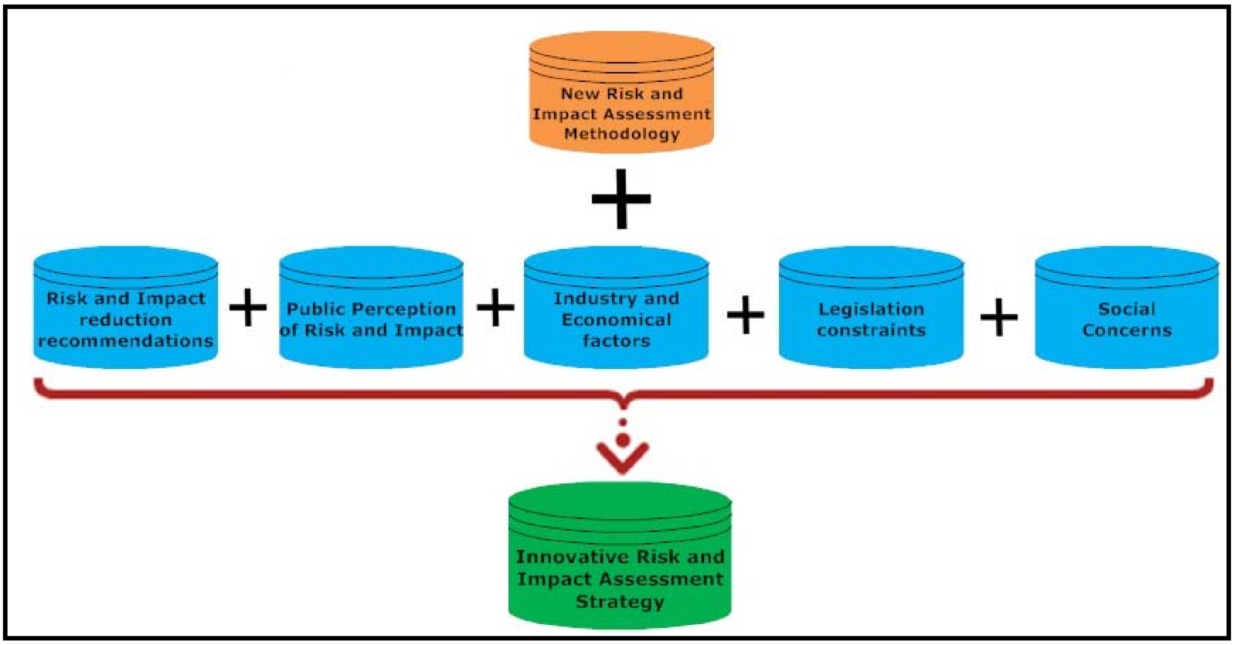


Figure 5.30 - Innovative risk and impact assessment strategy based on optoelectronic techniques and dispersion modelling

6. Final conclusions and personal contributions

6.1. Final conclusions

This thesis focused on a major issue concerning the scientific community, industry and the general public: **air quality**. In particular, it focused on **hazards and risks** posed by two types of atmospheric components: **aerosols** and **atmospheric gases**.

The **scope** of the thesis was stated as follows: **developing new and innovative methodologies and techniques for a better understanding and analysis of natural hazards and technological risks using optoelectronic techniques for environmental monitoring**.

The thesis achieved this scope through a series of **objectives**:

- by presenting a theoretical background concerning hazard and risk analysis, the atmosphere and optoelectronic monitoring techniques;
- by proposing optoelectronic techniques for natural hazard and technological risk analysis;
- by presenting case studies supporting the use of the selected optoelectronic techniques for hazard and risk analysis;
- by developing frameworks in which the data outputted by these techniques can be used for a better integration in the assessment process and finally in the development of strategies.

In order to rigorously analyze an environmental factor from the perspective of hazards and risks, the fundamental concepts of hazard and risk were presented and synthesized. This was achieved by a literature review regarding terminology, concept and usage of terms in various contexts. These contexts vary significantly form financial, social to industrial. Therefore there are slight differences in approaches for both terms, and there is a need for a classification according to various criteria. A classification of hazards and risks was presented according to the following criteria:

- origin
- manifestation mode
- temporal development
- affected surface and the duration of effects

From the above mentioned classifications two major hazard types were selected for assessment with optoelectronic technologies of:

- Natural hazards – **volcanic ash** by active and passive remote sensing;
- Technological hazards – monitoring and modeling the impact of **SO_2 emissions** associated with large combustion plants.

The identification and assessment of natural hazards and technological risks employed both qualitative and quantitative techniques. Therefore, a series of qualitative and quantitative methods of assessment were presented, including hazard identification check-lists and theoretical considerations regarding modeling tools.

A comprehensive description of the environmental factor where the natural and technological hazards occur was presented. The description of the atmosphere focused on its composition, describing aerosols and atmospheric gases, their sources, sinks and impact on radiative forcing and air quality.

Given the fact that optoelectronic techniques were employed in the analyses, atmospheric radiative transfer concepts were presented, with emphasis on light-particle interaction processes like absorption and scattering. Several optoelectronic techniques of the Romanian Atmospheric 3D research Observatory (RADO) for the monitoring of volcanic ash and sulphur dioxide were selected as analysis tools and presented along with the data types they provide.

For the monitoring of volcanic ash aerosol, passive remote sensing sun-photometers (AERONET data) combined with active remote sensing LIDAR systems were used. These techniques focus on the retrieval of the optical and microphysical properties of aerosols through direct Sun measurements and inversion products. The components of a LIDAR system were also presented alongside with in information regarding LIDAR data processing for quantitative analyses.

In order to be able to optically characterize a narrow spectrum of hazardous particles like volcanic ash, there is a need to present the optical and microphysical properties of several aerosol classes: urban-industrial aerosol (necessary for the discrimination between background aerosols and hazardous intrusions), biomass-burning aerosol and mineral dust (including volcanic ash).

For the above motioned classes several parameters retrieved via passive remote sensing were used for this characterization:

- **optical parameters**: aerosol optical depth (τ), and Angstrom parameter (α);
- **microphysical parameters** derived through inversion:
  - size distribution (*dV/dlnr*);
  - non-spherical particle fraction;
  - spectral single scattering albedo (ϖ_o);
  - spectral complex refractive index (n+*i*k).

These parameters prove to be very useful in the characterization the properties of aerosols, but due to a limitation of the sun-photometer, the fact that it only provides columnar data, no data is available regarding the location of the layer(s) within the atmospheric column.

LIDAR systems can overcome this limitation and therefore a combined LIDAR/sun-photometer approach was used to characterize the hazardous volcanic ash intrusion presented in the case study.

The case study on natural hazards focused on the identification and characterization of a volcanic ash intrusion during the April-May 2010 eruption of the Icelandic volcano Eyjafjallajökull. It revealed volcanic ash intrusions on 22 days during April and May 2010, the atmospheric layers originating from Iceland, as shown by HYSPLIT back-trajectories.

By analyzing the retrieved AERONET data the following conclusions were drawn:

- size distribution has a pronounced coarse mode (particles radius around 1.5-2 μm);

- the real part of the refractive index strongly varied from 1.46 to 1.55;

In order to retrieve the ash concentration, a key aspect in determining the risk associated with hazardous particles, LIDAR measurements were used.

Concentration values exhibited variable ash concentrations, an estimation of these concentrations revealed **values ranging between 10 and 360 μg/m^3** during daytime, **much lower than the air traffic hazard limit of 2 mg/m^3**.

Using the demonstrated techniques from the presented case study, a semi-quantitative methodology the assessment of hazardous particle intrusions was developed in order to be applied within the Romanian Atmospheric 3D Research Observatory. **The case study demonstrates that by using active and passive remote sensing it is possible to identify, characterize and determine the threat associated with a hazardous aerosol particle intrusion.**

Anthropogenic sulphur emissions are composed almost entirely of **SO_2 emissions** generated by burning coal and the smelting of sulfide rich ores. The three case studies presented in this chapter focus on these specific sources**, two case studies on coal-burning power plants**, and **one on the metallurgical industry.**

For the monitoring of sulphur dioxide emissions and imissions a combination of UV cameras (NILU Envicam-1), UV-fluorescence gas analyzers (HORIBA APSA-370), and dispersion models (AERMOD - ISCST3) was used.

Modeling is introduced alongside optoelectronic techniques as a tool for determining the impact associated with SO_2 emissions. The dispersion simulations for all three case studies were performed using the ISC AERMOD View software package, using the Industrial Source Complex Short Term (ISCST3) model. The case studies focus on the evolution of the air quality in the area of large combustion plants from the point of view of SO_2 emissions and imissions.

The case study on modeling and monitoring of SO_2 emissions from Mintia Large Combustion Power Plant described the technological process of a LCP and the input parameters used for the SO_2 dispersion simulations such as: source parameters, meteorological data and terrain data. The dispersion simulations show the **need of the implementation of desulphurization techniques (BAT)** for all the generating groups of Mintia LCP. The proximity to the city of Deva reaffirms this need, the simulation results and the imission monitoring data presented revealing the impact SO_2 emissions from Mintia LCP have on the city.

The modeling inconsistencies revealed by this case study can be most likely attributed to the incorrect estimation of the emission source's strength and temporal variation. **A need for the development of a more flexible, independent and mobile technique for the determination of variable SO_2 emission rates arises**.

The following case study addresses this issue through **modeling and monitoring of SO_2 emissions from Rovinari Large Combustion Power Plant using UV camera-derived emission rates.** This case study is based on optoelectronic data gathered within the 2010 campaign at Rovinari, organized by The Romanian Atmospheric 3D Research Observatory. The focus was on the **retrieval of the emission rate of a SO_2 source with UV cameras** followed by **dispersion simulations** with the calculated emission rate, and a **comparison with in-situ SO_2 imissions** measurements made with a HORIBA APSA-370 SO_2 monito**r.**

The dispersion simulations results correlate well with the HORIBA APSA-370 measured imission data, having the same size factor. This correlation is an important step in the **validation of UV cameras for industrial emission rate retrieval,** and demonstrates the validity of the approach based on optoelectronic techniques (UV-imaging-camera – SO_2 gas analyzer) combined with modeling software, in the present case, ISC AERMOD.

The last case study based on modeling and monitoring of SO_2 emissions from a metallurgical plant was selected due to the possibility of **comparing the impact of SO_2 emissions before and after the installation of a desulphurization technology**. This case study brings strong evidence through the use of modeling coupled with optoelectronic point-monitoring that the use of **best available technologies (BAT)** previously described are a very efficient way to significantly reduce the impact associated with SO_2 emissions of large combustion plants.

The simulation results, obtained for the situation before the installation of the desulphurization system, were compared with point monitoring data, showing a good qualitative correlation (Ajtai et al., 2012). The simulations performed for the conditions after the installation of the desulphurization system show a significant reduction in the ground level concentrations of SO_2, well within the range imposed by national and European legislation. **It can be concluded that the installation of desulphurization system reduced the SO_2 emissions and imissions significantly** and the obtained concentration values are well below the limits imposed by national and EU legislation.

The optoelectronic data gathered with these equipments were used in the **development of a risk and impact assessment methodology** with applicability in all sectors especially the economic and administrative ones which gain access to a high level scientific research, but also take active part in the **development of an innovative risk and impact assessment strategy**.

This strategy provides a better understanding of specific risks and impacts associated with LCPs by employing the new and original atmospheric risk and impact assessment methodology validated by the case studies using in an integrated way, dispersion simulations and advanced optoelectronic techniques.

By correlating emissions data, in-situ continuous point monitoring and UV camera path remote sensing data obtained along the plant perimeter, the approach presented in this thesis represents **a step forward towards certification of optoelectronic techniques for monitoring of anthropogenic pollutant emissions and as a tool for efficient risk and impact analysis.**

6.2. Personal contributions

This thesis proposes an original approach to assessing hazards and risks, by integrating optoelectronic techniques for atmospheric monitoring in the hazard and risk analysis process (**Ajtai, N.**, Török, Z., Stefanie, H., Ozunu, A., (2011a), *Integrated technologies for improving atmospheric risk and impact assessment models and studies*, Book of abstracts, 3rd iNTeg-Risk Conference in conjunction with 20th SRA-Europe Meeting, Stuttgart, Steinbeis Edition, ISBN 978-3-941417-65-6, pp 42).

Personal contributions to the present thesis included in **1 published book, 6 ISI published papers, 3 ISI papers submitted for publication, and 5 papers published in conference proceedings of international conferences** can be summarized as follows**:**

- literature review regarding hazards and risks:
    - Török, Z., **Ajtai, N.**, Ozunu, A., (2011b), *Aplicații de calcul pentru evaluarea riscului producerii accidentelor industriale majore ce implică substanțe periculoase*, Editura EFES, Cluj-Napoca, ISBN 978-606-526-078-8;
    - Stezar, I.C., Modoi, O.C., Török, Z.**, Ajtai, N.**, Crisan, D.A., Cosara, G.V., Senzaconi, F., Ozunu, A., (2011), *Preliminary investigation and risk assessment of contamination on an industrial site in Maramures County*, Environmental Engineering and Management Journal January 2011, Volume 10/2011, no.1, p. 65-73, ISSN: 1582-9596;
- presentation of the major qualitative and quantitative hazard and risk analysis methods:
    - Török Z., **Ajtai, N.**, Turcu, A.T., Ozunu A. (2011c), *Comparative consequence analysis of the BLEVE phenomena in the context on Land Use Planning; Case study: The Feyzin accident, Process Safety and Environmental Protection*, ISSN: 0957-5820, Imprint: ELSEVIER;
    - Török, Z., **Ajtai, N.**, Ozunu, A., Cordoş, E., (2009), *Chemical Risk Area Estimation as a Tool for Efficient Emergency Planning*, Studia Universitatis Babeş-Bolyai Chemia, 2009, ISSN: 1224-7154;
- presentation of the atmospheric environment and a synthesis of the radiative transfer concepts necessary for the identification and characterization of hazardous particles;
- synthesis on major sources and sinks of aerosols and associated radiative forcing;
- characterization of hazardous particles according to their optical and microphysical properties using data from Dubovik et al. 2002;
- analysis of AERONET sun-photometric data from Lille and Romania:
    - **Ajtai N.**, Ştefănie H., Stoian L.C., Oprea M.G., (2010a), *The volcanic ash and its impact on European air transport industry. A case study on the detection and impact of the the Eyjafjallajökull volcanic ash plume over North-Western Europe between 14th and 21st April 2010*, AES Bioflux 2(1):57-68;
    - Mortier, A., Goloub, P., Podvin, T., Deroo, C., Chaikovsky, A., Blarel, L., Tanre, D. **Ajtai, N.**, *Detection and Characterization of Volcanic Ash Plumes over Lille during Eyjafjöll Volcano Eruption*, submitted for publication in Atmospheric

Physics and Chemistry/Atmospheric Measurement Techniques Special Issue, Observations and modeling of aerosol and cloud properties for climate studies (ACP/AMT Inter-Journal SI), ISSN: 1867-1381);

  - **Ajtai, N.**, Stefanie, H., Ozunu, A., (2012b), *Description of aerosol properties over Cluj Napoca derived from AERONET sun photometric data,* submitted for publication in Environmental Engineering and Management Journal, ISSN: 1582-9596
  - **Ajtai, N.**, Stefanie, H., Costin, D., Ozunu, A., (2010b) *Comparative study of regional aerosols from columnar sunphotometric data in Romania*, "Papers presented at the 4th Workshop on Optoelectronic Techniques for Environmental Monitoring", pp 37, ISSN 2066-8651;
  - Unga, F., Dănilă, M.N., Gurlui, S., Dimitriu, D., **Ajtai, N.**, Timofte, A., Cazacu, M.M., (2012), *Optical parameters characterization of a volcanic ash intrusion over Northern Romania following the Grímsvötn volcano eruption in May 2011*, International Student Conference on Photonics, Book of Abstracts;
- analysis of volcanic ash concentration using a combined sun-photometer/LIDAR technique during a 6 months research stage at Laboratiore d'Optique Atmosphérique (LOA), Université Lille 1 Sciences et Technologies, France:
  - Mortier, A., Goloub, P., Podvin, T., Deroo, C., Chaikovsky, A., Blarel, L., Tanre, D. **Ajtai, N.**, *Detection and Characterization of Volcanic Ash Plumes over Lille during Eyjafjöll Volcano Eruption*, submitted for publication in Atmospheric Physics and Chemistry/Atmospheric Measurement Techniques Special Issue, Observations and modeling of aerosol and cloud properties for climate studies (ACP/AMT Inter-Journal SI), ISSN: 1867-1381;
- elaboration of a semi-quantitative methodology for the assessment of hazardous particle intrusions based on the identification of a particle intrusion, characterization of the optical and microphysical properties in order to determine weather or not the intrusion poses a hazard and on the determination of concentration of hazardous particles (quantitative);
- development of an algorithm for hazardous particle characterization and optimization of non-continuous LIDAR measurements;
- synthesis on large combustion plants regarding: plant types, fuels combusted, energy distribution;
- legislative synthesis on air quality, integrated pollution prevention and control, large combustion plants, national and European emission levels, national and European strategies for limiting of pollutants emitted by large combustion plants, best available technologies (BAT):
  - Mihăiescu, R., Mihăiescu, T., **Ajtai, N.**, Török, Z., Ozunu, A., (2011), *Air quality modelling as a tool used in selecting technological alterantives for developing a*

new abrasive facility, AES Bioflux 3(2):123-128, Online ISSN 2065-7647, Printed ISSN 2066-7620;

- synthesis on sulphur dioxide based on characterization physical-chemical and hazardous properties (Risk Phrases and Security Phrases), presentation of sources, sinks and "hot-spots";
- data collection within the 2010 Rovinari field campaign using UV cameras and UV-fluorescence gas analyzers (HORIBA APSA-370);
- analysis of the data collected with the HORIBA APSA-370 in Rovinari:
  - **Ajtai, N.**, Török, Z., Costin, D., Ştefănie H., Ozunu, A., (2011b), *Preliminary results of Modeling and Monitoring of SO_2 emissions from Rovinari Large Combustion Power Plant in September 2010*, Papers presented at 5th International Workshop on Optoelectronic Techniques for Environmental Monitoring, ISSN 20066-8651);
- SO_2 dispersion simulations using ISC-AERMOD for:
  - Mintia large combustion plant : **Ajtai, N.**, Deaconu, L., Ozunu, A., (2012c), *Sulphur dioxide emissions modeling and monitoring, originating from large combustion power plant Mintia, Hunedoara*, submitted for publication in Environmental Engineering and Management Journal, ISSN: 1582-9596;
  - Rovinari large combustion plant: **Ajtai, N.**, Török, Z., Costin, D., Ştefănie H., Ozunu, A., (2011b), *Preliminary results of Modeling and Monitoring of SO_2 emissions from Rovinari Large Combustion Power Plant in September 2010*, Papers presented at 5th International Workshop on Optoelectronic Techniques for Environmental Monitoring, ISSN 20066-8651;
  - Metallurgical plant: **Ajtai, N.**, Török, Z., Ozunu, A., (2012a), *Air quality modelling Of SO_2 emissions associated to metallurgical processes*, Studia Universitatis Babes-Bolyai Chemia, no 57, pp 57-65, ISSN: 1224-7154;
- retrieval of the SO_2 path concentration for the emissions of the Rovinari large combustion plant:
  - Stebel, K., Prata, F., Dauge, F., Amigo, A., **Ajtai, N.**, (2012a), *UV multispectral imaging cameras for validation of SO_2 emissions*, Proceedings of the EUMETSAT Meteorological Satellite Conference, Sopot, Poland, 03-07 September 2012
  - Mărmureanu, L., Deaconu, L., Vasilescu, J., **Ajtai, N.**, (2012), *Combined optoelectronic methods used in the monitoring of SO_2 emissions and imissions*, paper submitted for the 9th edition of the ELSEDIMA international conference, 25-27 October 2012, Cluj-Napoca, Romania; paper submitted for the 9th edition of the ELSEDIMA international conference, 25-27 October 2012, Cluj-Napoca, Romania;
- study of the effects generated by the implementation of a desulphurization system (BAT) at a metallurgical plant:

  - **Ajtai, N.**, Török, Z., Ozunu, A., (2012a), *Air quality modelling Of SO_2 emissions associated to metallurgical processes*, Studia Universitatis Babes-Bolyai Chemia, no 57, pp 57-65, ISSN: 1224-7154);
- development of a new methodology for risk and impact assessment based on optoelectronic monitoring techniques and modeling tools and of a innovative strategy for assessing the impact of SO_2 emissions associated with LCPs.

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