With the contribution of the LIFE Programme of the European Union

LIFE 15 IPE IT 013

prepAIR

Po Regions Engaged to Policies of Air

SUMMARY

REPORT 3 COVID-19

The Effects of the Covid-19 Lockdown on PM10 chemical composition in the Po valley

With the contribution of the LIFE Programme of the European Union

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prepAIR Po Regions Engaged to Policies of Air

This document is the summary of “REPORT 3 COVID-19: STUDIO DEGLI EFFETTI DELLE MISURE COVID-19 SULLA COMPOSIZIONE CHIMICA DEL PARTICOLATO NEL BACINO PADANO - GENNAIO 2021”

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1 The CONTEXT

The LIFE-IP PrepAIR project was launched in 2017 with the aim to create tools and actions to improve air quality in the Po Basin. In the first months of 2020, the health crisis due to the COVID-19 pandemic and the consequent policy measures adopted have generated a drastic and sudden reduction of some sources of atmospheric pollution. The conditions were useful to test some actions to combat air pollution in one of the most polluted areas of Europe, and, unfortunately, also dramatically affected by the health emergency.

In this context the PrepAIR Steering Committee has decided to carry out a specific study to evaluate the effect of lockdown measures on air quality. The analysis was conducted in three different phases (www.lifeprepair.eu) and this work represents the synthesis of the third one in which the chemical composition of PM10 is examined in the special stations of the project. The study aims to verify and consolidate the preliminary conclusions of the two previous reports and to obtain new elements of knowledge necessary to set up the next planning phase in terms of air quality.

The study was conducted by analyzing in particular two periods: pre-lockdown (January 2 - March 9) and lockdown (March 10 - May 18). To fight the rise of infection two DPCM (Prime Minister Decrees) put in place strictly national lockdown measures on 9th and 11th of March 2020. Lockdown restrictions have begun to be eased across Italy the 26th of April with a recovery of limited activities and the cessation of national restrictive measures on traffic within regional boundaries.

2. CHEMICAL ANALYSIS in PO VALLEY

As part of the PrepAIR project, action A4 provided the creation of a measurement network for the PM10 chemical characterization. The data collected will allow intra-situ analysis, to verify variations in air quality parameters and source pressures, following the implementation of the regional plans for air quality. Moreover, data will be used to verify the air quality in Po basin.

The network is based on existing monitoring stations and consists of a rural site in Schivenoglia (MN) and four urban sites - Turin, Milan, Bologna and Vicenza (not in this study due to instrumental problems).

Daily filters are collected with two low-volume gravimetric samplers at each site. The following chemical analysis are carried out on PM10 filters: cations (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}) and anions (Cl^-, NO_2^-, Br^-, NO_3^-, PO_4^{3-}, SO_4^{2-}) analyzed by ion chromatography (IC); elements (Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Pb), analyzed by X-ray emission spectroscopy (XRF, X- ray fluorescence); the carbonaceous fraction (OC and EC, organic and elemental carbon) quantified by thermo-optical technique (Sunset Laboratory inc.) using the NIOSH-Ike thermal protocol.

Levoglucosan, an hydro-sugar derived exclusively from incomplete combustion of cellulose, is also analyzed through IC as an important tracer of biomass burning.

In addition to the special stations, the urban site of the city of Aosta was included in the study. The analyses differ for certain ions and elements and a different protocol (EUSAAR2 instead of NIOSH-lke) is used to determine the carbonaceous fraction. EC and OC concentrations were so corrected by a comparison between the two protocols. The results are satisfactory: the OC has a 5% difference between NIOSH-like and EUSAAR2 (m_{OC}=1,05, R^2= 0,99) while the EC of 19% (m_{EC}=0,81, R^2= 0,99).

3. MATERIAL AND METHODS

PM10 mass closure was calculated based on non-reprocessed analytical data which led to identify the following components: together with the carbonaceous fraction (OC and EC) and the main ions of the inorganic secondary, - nitrate and ammonium sulphate (NH_4NO_3 e $(NH_4)_2SO_4$) - the crustal component, the "other elements" and finally the sum of the "others ions".

Crustal matter is constructed by calculating the oxidized form of its main elements (Si, Al, Mg, Fe, etc.) following Marcazzan et al. (2002) and using the reference values of the Mason (Nature, 1966). Enrichment factor (EF) is calculated to estimate what percentage to attribute to the crustal source. In Aosta, since Si was not analyzed, a multiplicative factor derived from Mason (1966) was used, as a function of the concentration of Al.

"Other elements" is composed of the elements that do not form the crustal component (eg. copper), plus the non-terrigenous percentage of the elements that may have a mixed origin (eg. iron). Also "other ions" are the sum of the non-crustal ions (chloride, bromide, etc.), and the non-terrigenous part of the crustal ones (potassium, calcium, etc.), except the main secondary ions considered separately (ammonium, nitrate and sulphate). Finally, "ND" indicates the PM10 mass fraction not determined, as it is not covered by all the analysis carried out and therefore cannot be quantified. “ND” is mainly composed of organic matter and water.

In the analysis of each site, levoglucosan and copper were taken into consideration. Both do not contribute to the mass of the particulate matter, but are useful as tracers, the first of the "combustion of biomass burning" and the second of the abrasion of the braking material (non-exhaust traffic). Therefore, copper was used together with the EC for the analysis of the traffic source.

Lastly, the ratio between organic carbon and elemental carbon (OC/EC) was evaluated. This parameter can discriminate different sources of PM. Low value indicates a contribution from sources such as traffic (Handler et al., 2008), while high value may be linked in the colder months to primary emissions and meteorological factors. In winter, low values of the PBL (Planetary Boundary Layer) height and atmospheric stability, typical of the Po Valley, favour the condensation of volatile precursors and the production of aged particulate with secondary enrichment, or the combustion of woody biomass. In the warm months, high values are affected by the increased production of SOA (Secondary Organic Aerosol) due to photochemical activity.

4. RESULTS

Data analysis was carried out mainly considering the years 2019 and 2020 as the sampling campaigns in the PrepAIR project started in April 2018. Results were confirmed using the analysis carried out in Milan in which the database started in 2013, and in Aosta site which has information since 2017.

The mass closure in each site (fig. 4.1) shows a similar chemical composition, indicating a homogeneity in the Po basin. Almost every component shows a reduced variability in each period, with typical differences in the transition from the winter to the warmer season: reduction of ammonium nitrate, percentage increase in ammonium sulphate and growth of crustal matter, more evident during lockdown 2020 due to a dry April. Respect to the years, fingerprints of the chemical composition of each site are substantially unchanged, with some slight peculiarities, depending on the seasonal variation. As part of the PrepAIR project, variations within the basin will be the subject of future studies to investigate the presence of site-specific peculiarities.

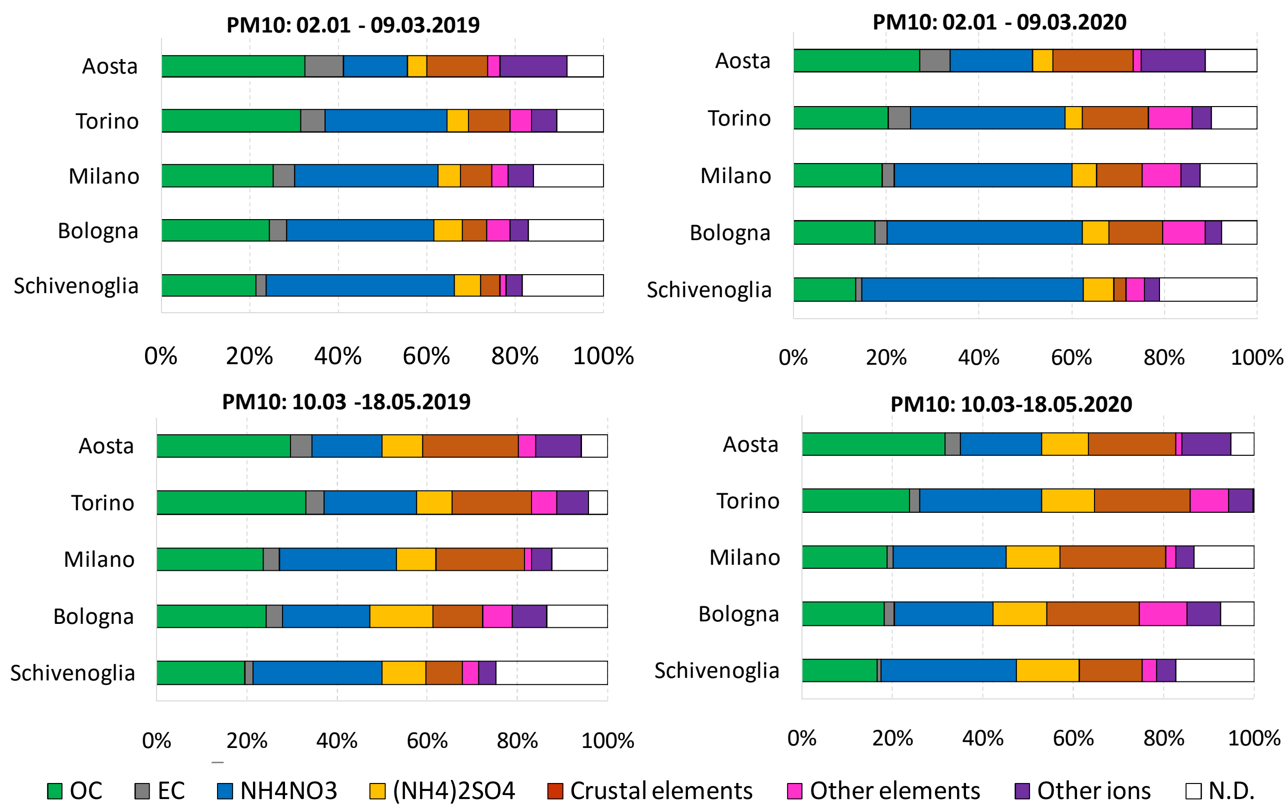

Figure 4.1 – Comparison relative mass closure of PM10 in 2019 and 2020 for both periods.

The variability of the five cities falls down within the typical one linked to annual variations: this is confirmed by the mass closure at the Milan site where a longer time series is available (fig. 4.2).

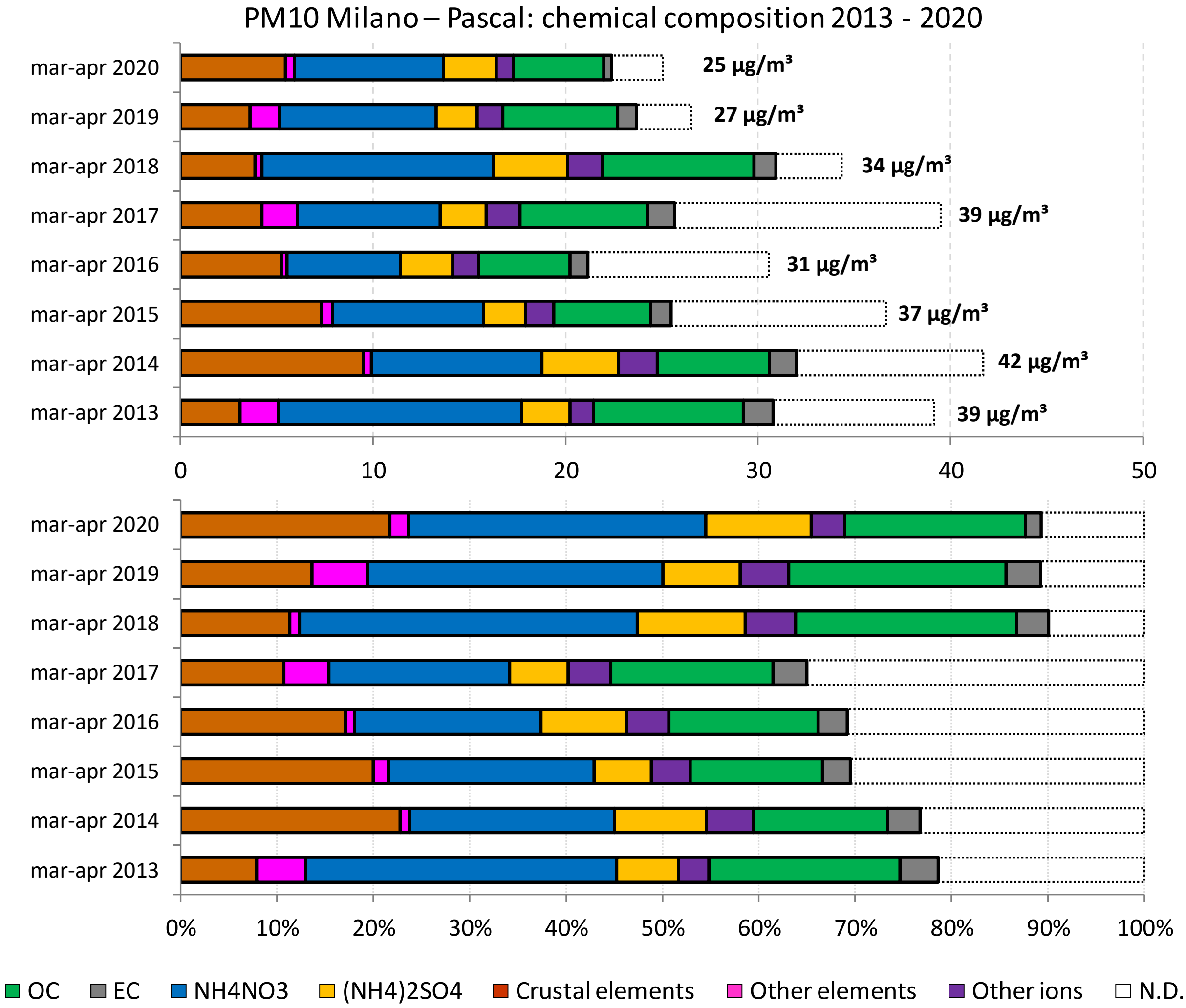

Figure 4.2 – Relative (up) and absolute (down) mass closure of PM10 from 2013 to 2020 at MI-Pascal.

Biomass burning, identified by levoglucosan, shows an important increase during the lockdown both in the Lombardy sites and in Turin (fig. 4.3). In particular, the rural site of Schivenoglia shows the greatest growth (with concentrations almost tripled, fig. 4.4) compared to the other two sites (concentrations doubled) probably due to a more consistent use of wood in a rural site. In general, the increase is presumably linked to the home confinement imposed by the lockdown measures, common to each site. The differences (no variation in Aosta and Bologna) could be due to temperatures, particularly chill in some areas, and to a diversified use of wood in the Po Valley. With the second half of April and warmer temperatures, levoglucosan concentrations decreased, in line with other years. The graph of fig. 4.3 clearly shows the growth in the 2020 lockdown period and the subsequent decrease.

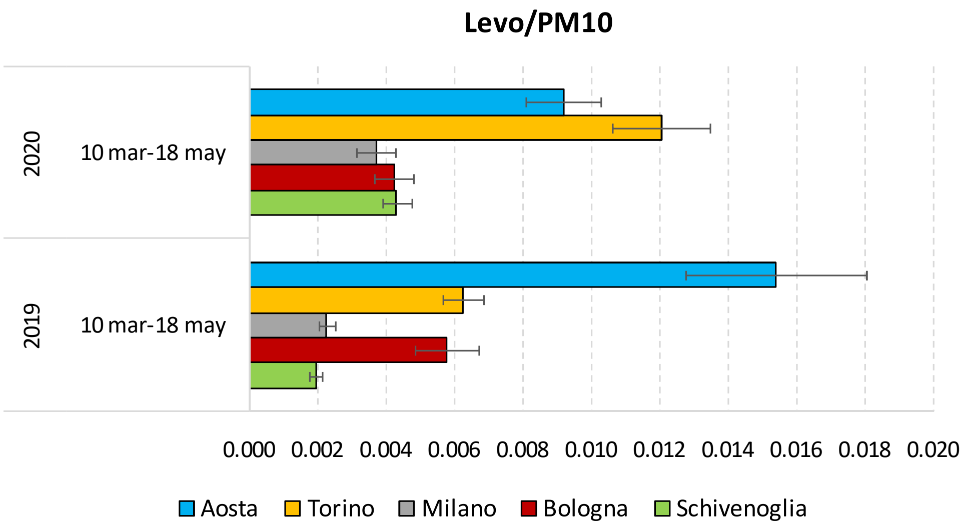

Figure 4.3 – Levoglucosan/PM10 comparison in each site during lockdown for 2019 and 2020.

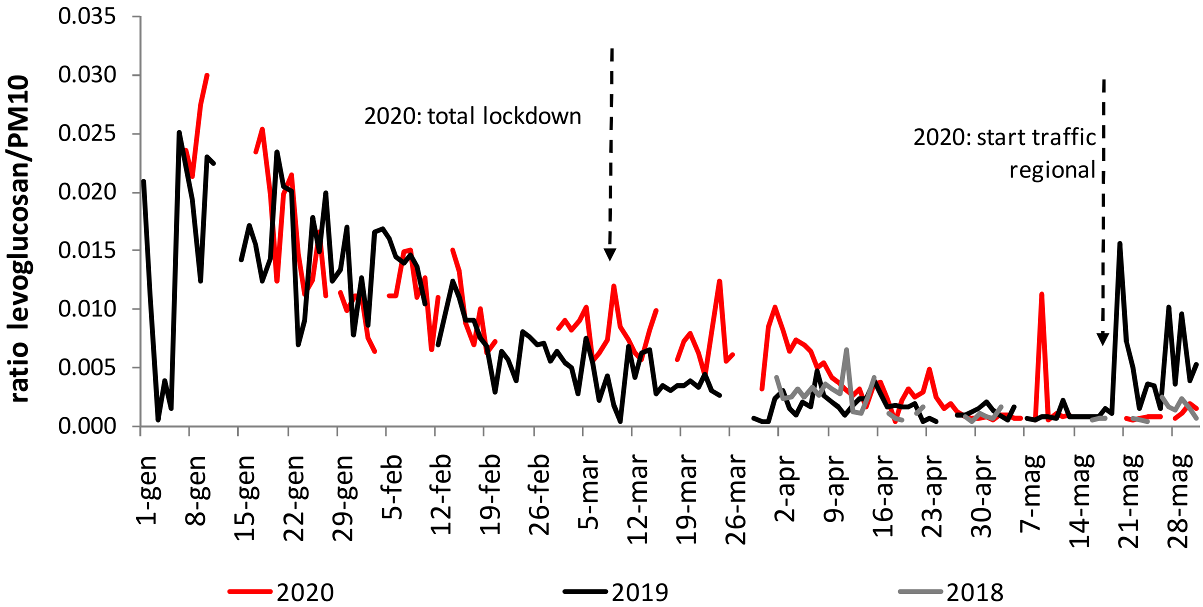

Figure 4.4 – Schivenoglia: comparison between daily trends of levoglucosan/PM10 in 2019 and 2020 (January-May).

Each site shows a reduction of EC/PM (Fig. 4.5) during lockdown. This is due to the drop in transport-related emissions and then to this primary compound. Milan and Aosta confirm this result with their time series: 31% reduction in Milan in March (2020 vs 2013-2019) and 40% in Aosta (2020 vs 2017-2019).

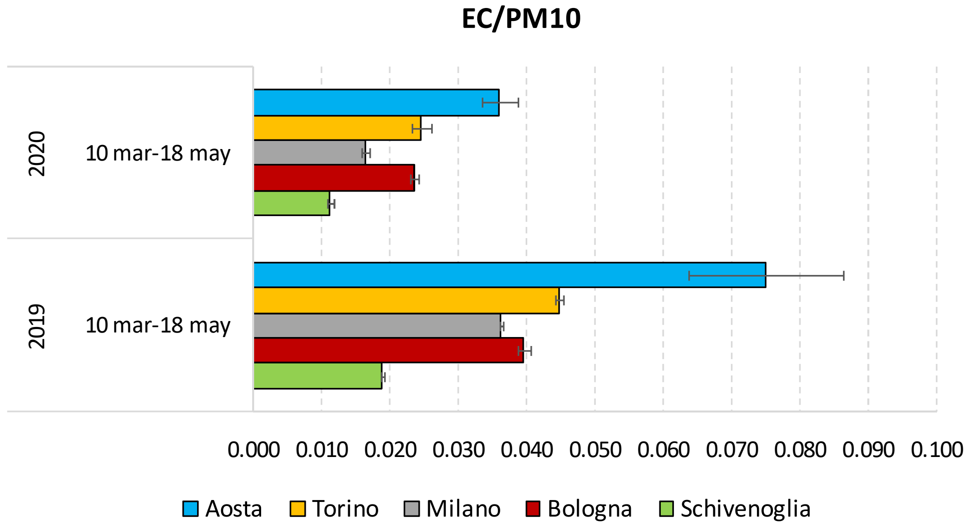

Figure 4.5 - EC/PM10 comparison in each site during lockdown for 2019 and 2020.

EC/PM10 trend in Aosta is always lower during lockdown period in 2020 both for 2019 mean and for the minimum of 2017-2019 (fig. 4.6).

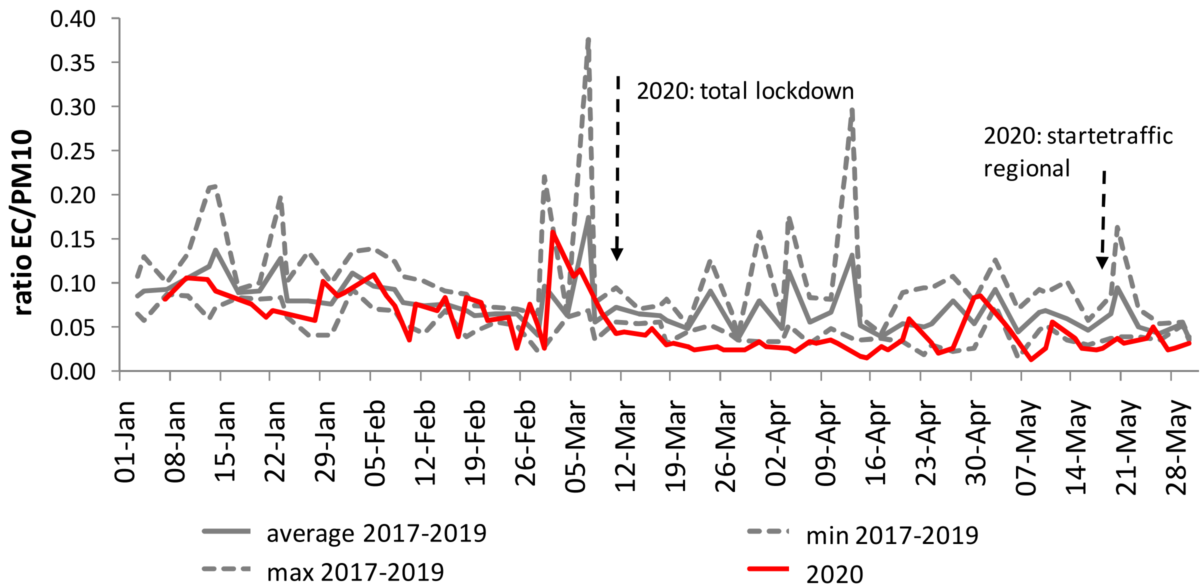

Figure 4.6 - Aosta: comparison between daily trends of EC/PM10 in 2020 and mean, maximum and minimum of 2017-2019 (January-May).

The drop in copper (Cu, as a not-exhaust traffic emissions marker) in the particulate matter is also in line with these results. Cu/PM10 ratio in the urban background site of Lombardy (fig. 4.7) clearly shows the collapse of this element since mid-March.

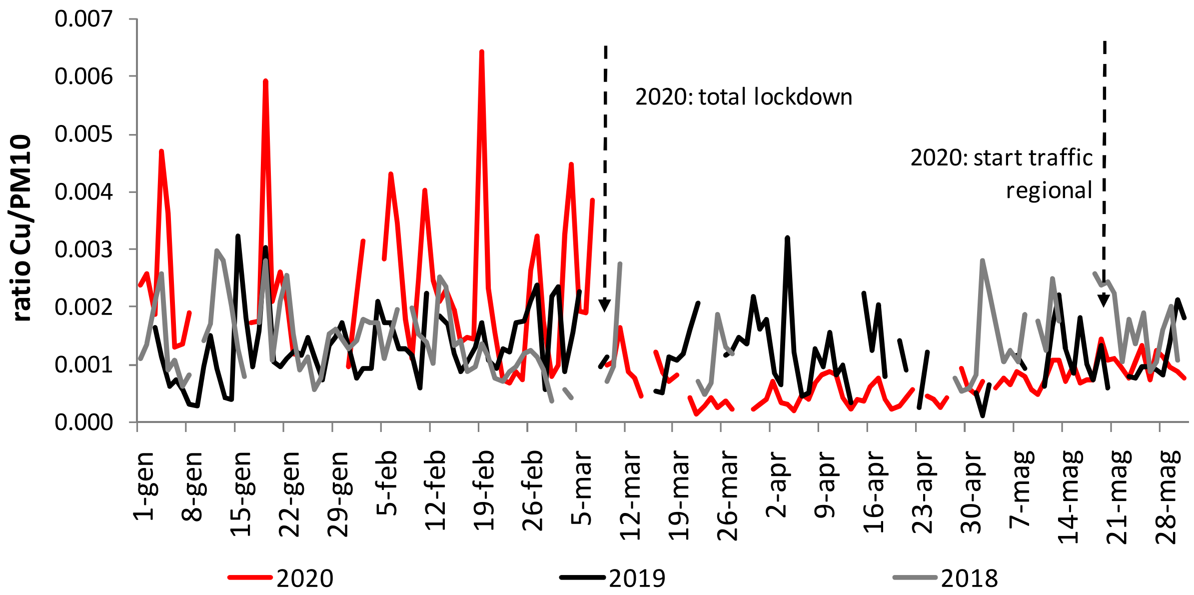

Figure 4.7 – Milano-Pascal: comparison between daily trends of Cu/PM10 in 2018, 2019 and 2020 (January-May).

The increase in OC/EC ratio is again common in all cities during the lockdown 2020 (Fig. 4.8). This growth is certainly linked to the decrease in EC, but it can be derived by increasing of the biomass burning (in some sites it shows growth) and of the SOA (Secondary Organic Aerosol) formation by photo-oxidation (not verifiable hypothesis with the available data).

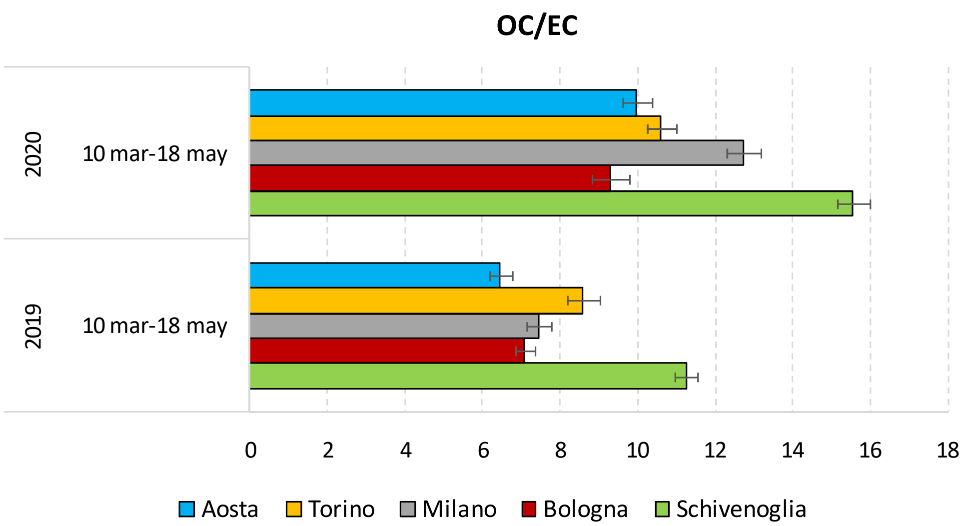

Figure 4.8 - OC/EC comparison in each site during lockdown for 2019 and 2020.

Another interesting result due to the impact of the COVID-19 policy measures is the decrease of "other elements" fraction. The Aosta site is adjacent to a steel plant and the concentration of "other elements" shows an important decline after mid-March with values close to the minimum of the 2017-2019 period (fig. 4.9).

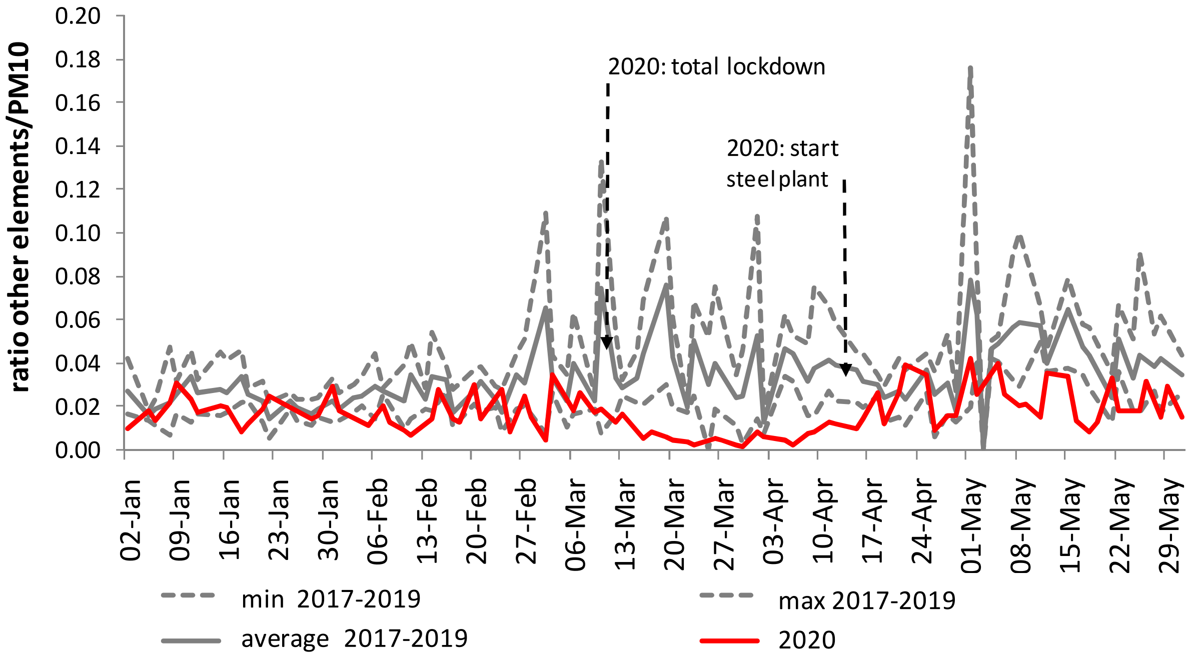

Figure 4.9 – Aosta: comparison between daily trend of "others elements" in 2020 and mean, maximum and minimum of 2017-2019 (January-May).

SIA (Secondary Inorganic Aerosol) analysis highlights a homogeneity within the Po basin, as well as shown in the graph in fig. 4.10, with the only difference for the Aosta station that has lower contributions of the ammonium ion (absolute concentrations in Table 4.1). Air masses transport is a very frequent phenomenon in the Aosta Valley region, and it explains how most of the inorganic secondary measured at the site comes from remote origin (mainly from the Po valley), with a minor local contribution. Average annual contribution of 25% on PM10 was estimated by PMF (Positive Matrix Factorization) about transported nitrates and sulphates-rich aerosol in the city of Aosta (Diémoz et al., 2019). In remote sites, where there are fewer local emissions, the relative importance of this contribution is greater.

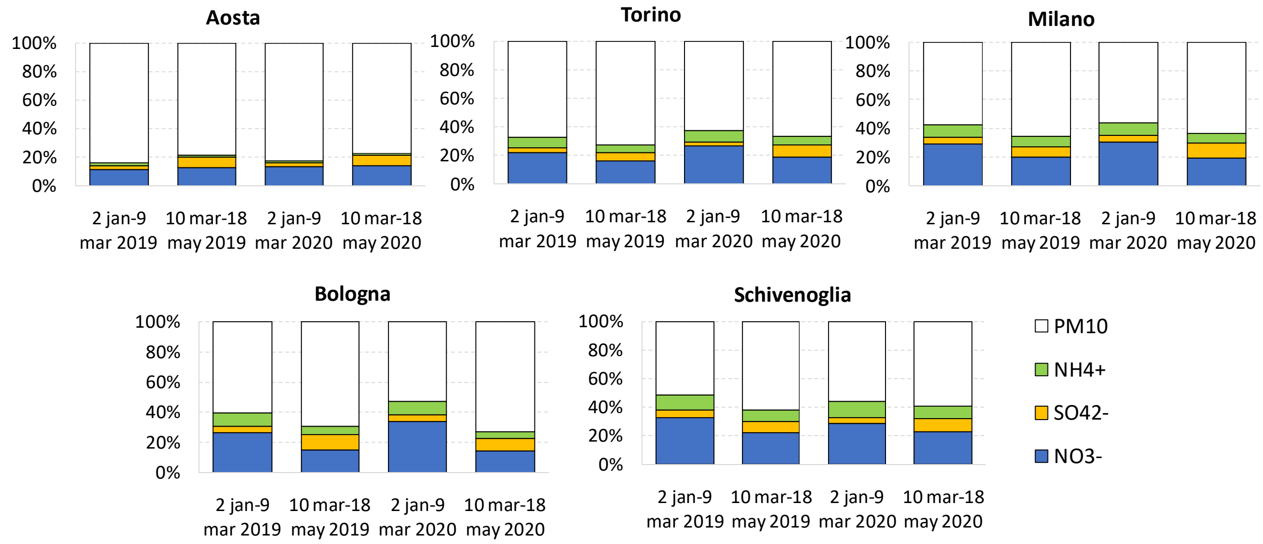

Figure 4.10 – Sulphate, ammonium and nitrate of PM10 in each site during both periods in 2019 and 2020.

Nitrate ion shows a slight increase during the lockdown period in all sites from 2019 to 2020, except in Milan, with the maximum concentration measured in Schivenoglia (5.6 µg/³). Comparing the data with a longer time series, such as the one in Milan or Aosta, these variations seem to fall down within the inter-annual variability range. The sulphate ion also increases in each site in 2020 except in Bologna where no variation was observed.

		2019				2020			
		NO_3^-	**SO_4^{2-}**	**NH_4^+**	**PM10**	**NO_3^-**	**SO_4^{2-}**	**NH_4^+**	**PM10**
		µg/m³				µg/m³			
AO	2 jan-9 mar	2.2	0.6	0.3	20	3.0	0.7	0.2	23
	10 mar-18 may	1.5	0.8	0.2	12	2.0	1.1	0.2	14
TO	2 jan-9 mar	12.2	2.0	4.1	56	15.9	1.6	4.6	60
	10 mar-18 may	3.4	1.2	1.2	21	4.0	1.8	1.3	21
MI	2 jan-9 mar	15.5	2.4	4.7	53	14.0	2.2	4.1	46
	10 mar-18 may	4.3	1.5	1.6	21	4.3	2.3	1.5	22
BO	2 jan-9 mar	11.0	2.0	3.6	42	14.5	1.9	3.8	42
	10 mar-18 may	2.5	1.7	0.9	16	3.2	1.7	1.0	22
SCHI	2 jan-9 mar	16.8	2.6	5.4	51	13.8	1.9	5.4	48
	10 mar-18 may	4.9	1.8	1.8	22	5.6	2.4	2.1	25

Table 4.1 – Concentrations of sulphate, ammonium, nitrate and PM10 in each site for both periods.

SIA is one of the predominant components in PM10 mass balance in the Po valley (e.g. up to 54% in Schivenoglia). Understanding his formation from main precursors is one of the main goals. Unlike the decrease of NO_X (as seen in the previous reports published by PrepAIR), due to the collapse of traffic source, NH_3 shows an opposite behaviour: during the lockdown ammonium concentrations increase in many stations because agricultural-livestock activities have never stopped. Its trend is in agreement with SIA ones which has not shown significant decreases compared to the previous year, despite the consistent reduction in nitrogen oxides.

Figure 4.11 shows a comparison between the temporal trend of ammonia and ammonium nitrate in both years. NH_4NO_3 concentration was calculated as daily average of the concentrations measured in different sites while NH_3 concentration represents the daily average between the station of Turin (Piedmont) and Corte de Cortesi (an agricultural sites of Lombardy adjacent to a swine farm). Although it is premature making considerations between sources and concentrations at the receptor, a good agreement is observed between the two trends, especially in specific weather-climatic conditions (situations favourable to the accumulation of air masses rich in ammonia). This could suggest a role of agriculture on concentrations during the lockdown, given the invariance of SIA and PM10 in 2020 compared to 2019. Further considerations require additional studies on processing of the meteo-climatic component and chemical transformations of ammonia in the atmosphere as well as accumulation processes to verify some hypotheses already analyzed in Lombardy in previous years (Colombi et al., 2018; Dal Santo et al., 2018).

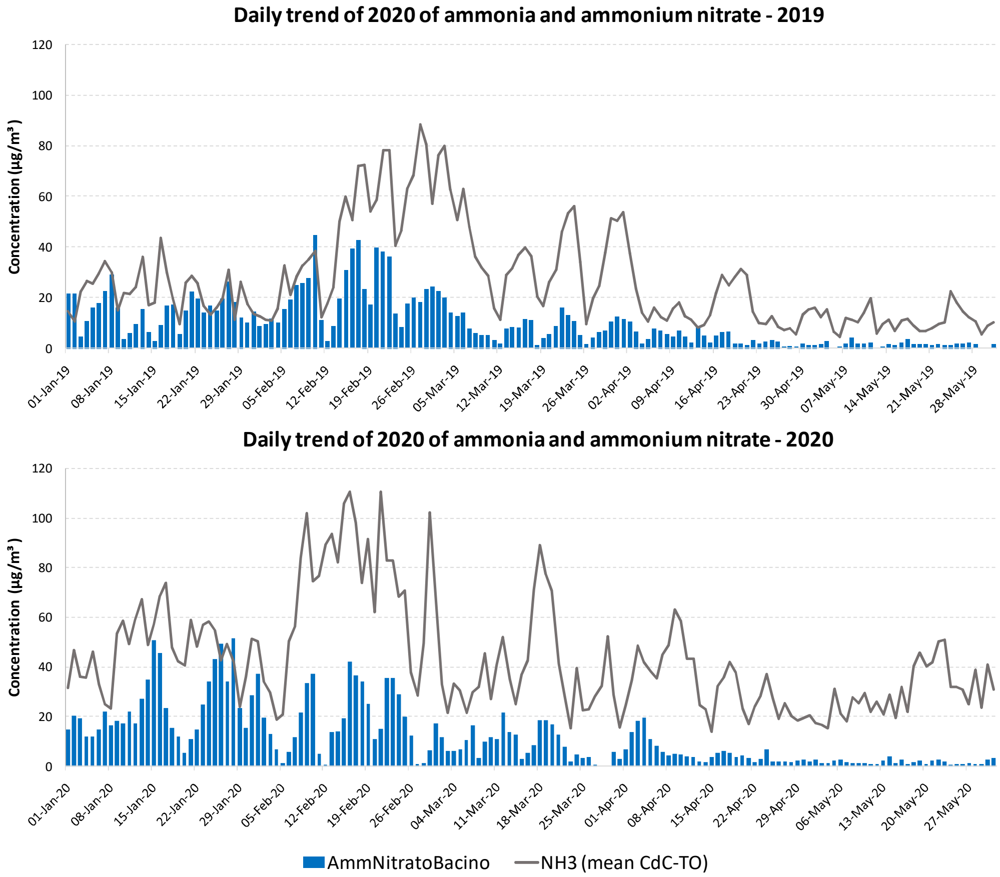

Figure 4.11 – Temporal trend of ammonia (average between Corte de Cortesi and Torino) and ammonium nitrate (average of Milano Pascal, Torino, Bologna, Schivenoglia).

The last figure (Fig. 4.12) is a summary of the main results for March-April period. The comparison between the variability of the chemical composition in the period 2013-2019, represented by the boxplot, and the average of 2020 highlights several results: in Milano Pascal many traffic-related components, several metals and EC show a considerable reduction, outside the boxplot variability or an increase, like levoglucosan tracer, while inorganic ions fall within the range of variability, between the first and third quartiles of the last 7 years.

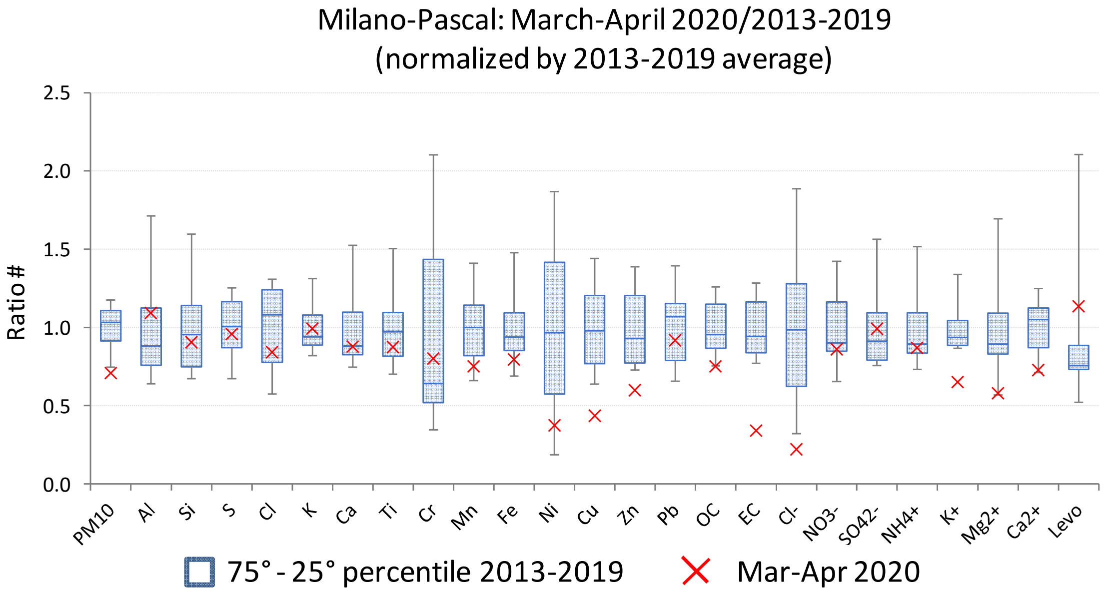

Figure 4.12 – Milano: comparison between PM10 chemical components in 2013-2019 (boxplot) and 2020

5. CONCLUSIONS

Aim of the study

The first two reports of the PrepAIR project on the effects of the lockdown on air quality (https://www.lifeprepair.eu/) highlighted the drastic reduction of the determinants that occurred in the Po basin area following the restrictive measures taken in the context of the COVID19 pandemic, which caused the emission reduction of both NO_X (which reached a maximum weekly decrease of 40%) and primary PM10 (maximum decrease 20%). Furthermore, the considerable decrease in the concentrations in air of gases (both primary and secondary), resulting from the emission reductions, and PM10 trend (which showed discontinuous variations during the lockdown period, more linked to weather conditions), was also analyzed. Aim of this third report was to investigate the reason for PM behaviour and if, and in what way, its chemical composition has changed.

Knowledge Status

It has long been known how the mass of PM10 in the Po Valley, albeit with different contributions depending on the studied area, is mainly composed of organic carbon and secondary inorganic species and subsequently, with reduced percentages, other ionic species, crustal matter and elemental carbon. Secondary fraction is certainly the major contribution to PM and can be both inorganic, dominated in winter by ammonium nitrate while in summer, due to its volatilization, by sulphate, and organic, by condensation of volatile precursors in the cold season and by photo oxidation in the warmer one. The formation of this component in the Po Valley is very complex (Gilardoni et al., 2011) and the geography of the territory and the meteorological conditions (atmospheric stability, low temperatures, high humidity and thermal inversions), together with the availability of precursors, have a more important role than the variations of the single sources. The trend of PM over time (Air quality in Italy, SNPA - ED. 2020) in the Po Valley has shown reductions in the primary contribution to PM, thanks to a series of interventions implemented by the administrations, while this decrease it does not seem visible for that part of particulate matter that forms in the atmosphere (Air quality in Italy, SNPA - ED. 2020, insights part). Another important contribution to PM is woody biomass burning (BB) which, for example, in Emilia-Romagna is estimated to reach up to 35% of the PM2.5 concentration in the cold season (Scotto et al., 2021 - submitted). Furthermore, from recent studies, the combustion of BB, in addition to involving an important contribution to the primary PM, has also been shown to contribute to the production of a secondary organic part (Paglione et al., 2019; Kodros et al., 2020).

Results

In this study, PM10 chemical composition of five stations in Po valley were analyzed, four of which are part of the PrepAIR project (special stations of Turin, Bologna, Milan Pascal and Schivenoglia). Two main periods were examined: pre lockdown (January 2 - March 9) and lockdown (March 10 - May 18) during 2019 and 2020 and where possible, in Milan Pascal and Aosta, with a longer time

series.
Both periods showed a very low variability of almost all components in each site, with typical differences in the transition from winter to warmer season. SIA analysis (sum of nitrate, ammonium and sulphate), which is the component of PM10 in the Po valley, highlighted a homogeneity within the basin, with the only difference in Aosta which showed much lower contributions for the ammonium ion. Most of the inorganic secondary measured at the Aosta site is of remote origin (mainly from the Po basin).
The comparison in the 2020 lockdown period compared to 2019 highlights:

- no evident reduction of secondary compounds in each site;
- decrease in elemental carbon and copper in each site, components largely linked to traffic emissions;
- increase in levoglucosan, biomass burning tracer, in most stations;
- increase in OC/EC ratio (due to lower values of EC, increase of BB for domestic heating and maybe also SOA)

These results are supported by the analysis of the Milan and Aosta longer time series. For example, the drastic drop in vehicle traffic is evident: reduction of elemental carbon up to 40% in Aosta (compared to 2017-2019 average) and to 31% in Milan (compared to 2013-2019 average) in 2020.

Interpretative hypothesis

The main precursors of the secondary inorganic aerosol are SO_2, NH_3 and NO_X. Sulphur dioxide has dropped dramatically in the last 20 years, but it still available as the last two ones by agriculture and combustion (mainly traffic, heating and industries). During the lockdown 2020 ammonia in several sites has shown higher values than in the previous year, however agricultural-livestock activities have never stopped, and nitrogen oxides remained available anyway (in the Po Valley the average NO_2 remained around 10-25 $\mu g/m^3$ during the months of the lockdown), despite the considerable drop (up to 40%, maximum weekly decrease in emissions).

Therefore, both NO_X and NH_3 were available enough to support secondary aerosol formation.

In addition, levoglucosan, BB source tracer, in three of the five sites analyzed, showed an increase during the total lockdown period (in the rural site the levoglucosan showed values almost tripled compared to the same period in 2019), probably due to the measures of limitation of movement of people that forced them into the house, as well as the decrease in temperatures in some areas.

Plans Implications

The results of the study showed that the reduction of a part of the pollutants is not enough to determine an appreciable variation in the formation of secondary particulate and confirmed that the necessary interventions to reduce particulate matter must not only be coordinated at the Po valley level, but also must concern all the activities that contribute to the production of precursors (mainly agriculture and all combustion, such as traffic, biomass burning, industrial sector and services) acting incisively on emissions.

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Po Regions Engaged to Policies of Air

THE PREPAIR PROJECT

The Po Basin represents a critical area for the quality of air, as the limit values of fine powders, nitrogen oxides and ozone set by the European Union are often exceeded. The northern Italian regions re included in this area as well as the metropolitan cities of Milan, Bologna and Turin.This area is densely populated and highly industrialized. Tonsof nitrogen oxides, powders and ammonia are emitted annually into the atmosphere from a wide variety of polluting sources, mainly related to traffic, domestic heating, industry, energy production and agriculture. Ammonia, mainly emitted by agricultural and zootechnical activities, contributes substantially to the formation of secondary powders, which constitute a very significant fraction of total powders in the atmosphere.Because of the weather conditions and the morphological characteristics of the basin, which prevent the mixing of the atmosphere, the background concentrations of the particulate, in the winter period, are often high.In order to improve the quality of the air in the Po Valley, since 2005 Regions have signed Program Agreements identifying coordinated and homogeneous actions to limit emissions deriving from the most emissive activities.The PREPAIR project aims at implementing the measures foreseen by the regional plans and by the 2013 Po Basin Agreement on a wider scale, strengthening the sustainability and durability of the results: in fact, the project involves not only theregions of the Po valley and its main cities, but also Slovenia, for its territorial contiguity along the northern Adriatic basin and for its similar characteristics at an emissive and meteoclimatic level.The project actions concern the most emissive sectors: agriculture, combustion of biomass for domestic use, transport of goods and people, energy consumption and the development of common tools for monitoring the emissions and for the assessment of air quality over the whole project area.

DURATION

From February 1st2017 to January 31 2024.

TOTAL BUDGET

17 million euros available to invest in 7 years: 10 million of which coming from the European Life Program.

COMPLEMENTARY FUNDS

PREPAIR is an integrated project: over 850 million euros coming from structural funds and from regional and national resources of all partners for complementary actions related to air quality.

PARTNERS

The project involves 17 partners and is coordinated by the Emilia-Romagna Region – General directorate for the territorial and environmental care.

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