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HAZARDOUS AEROSOL EMISSIONS DURING AGRICULTURE BIOMASS BURNING SEASON IN SON LA AND BA VI REGIONS, VIETNAM

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ABSTRACT: Major national emission sources are assessed by characterization of smoke pollution arising due to traditional agriculture, domestic, and cooking activities in the regions of the biggest biomass burning. Measurement campaigns were carried in Son La and Ba Vi regions, Vietnam, during the dry seasons of 2013 and 2015-2016. PM and BC monitoring, aerosol sampling, chemical speciation were conducted to evaluate ambient smoke level, to relate the characteristics of local on-field emissions to regional aerosols, and to identify the dangerous components of smoke composition. The regions Son La and Ba Vi in February-June faced severe levels of air pollution, with critical $PM_{2.5}$ and PM_{10} concentrations up to 130 and 167 μg/m^3, respectively, significantly exceeding the air quality standards. A wide range of PM mass concentrations was categorized according to the smoke level, supported by the evolution of carbon (OC, EC) fractions as well as ionic species and molecular markers. The level of PM and BC concentrations was seen to be dependent on factors such as weather conditions and precipitation. Non-acid carbonyls, carboxylates, and aliphatic carbon compounds were evolved with increasing smoke intensity, together with carbonates in coarse size fractions, indicating a large impact of smoke emissions and soil lifted up by the intense fires. On-field emissions in both smoldering and flaming phases were assessed in near-source measurements.

Keywords: Biomass burning, air quality, aerosol characterization.

INTRODUCTION

Quantification of adverse particulate pollution is a serious problem in less-industrialized countries where environmental protection is low. Combustion has been increasingly recognized as the most important source of aerosols, containing black carbon (BC), organic carbon (OC), and inorganic fly ash. Environment impacts of combustion aerosols and adverse health effects of air pollution are remaining underestimated, with a big concern in highly populated areas and during dry seasons. They impact directly and indirectly on the Earth's radiation balance, cloudiness, and subsequently affect the regional and global climate. Various components of biomass burning (BB) aerosols are considered

as dangerous pollutants with respect to human health, causing respiratory and cardiovascular diseases.

BB activities in northern Southeast (SE) Asia comprise a variety of combustion processes, ranging from residential burning, domestic heating, and cooking while burning of agricultural residuals is a particularly important source during dry season [1]. Initiated in 2007, the Seven South-East Asian Studies (7-SEAS) campaign seeks to perform interdisciplinary research in the field of aerosol-environment interactions, particularly focusing on impacts of BB [1, 2]. Combustion emissions in SE Asia typically peak in spring when the most intense BB activity occurs. In Vietnam rice straw burning deserves an attention as the most important pollution practices in agricultural provinces. Residential cooking emissions are a key source of PM especially in winter when urban emissions are dominant. Agricultural areas surrounding cities can act as a seasonal source of pollution during intense rice straw burning in the weeks following harvest periods. However, a lack of knowledge concerning the properties of source-dependent aerosols hinders assessments of BB impact on air quality: adverse air pollution and impacts of chemically - active aerosols are remaining underestimated.

This paper is based on continued work following the 7-SEAS campaign and focuses on rural areas in Son La and Ba Vi regions, a particularly important rice - producing area in Vietnam. We present two measurement campaigns carried out during the dry seasons of 2013 and 2015-2016 and review the findings for the environmentally and healthily dangerous organic and inorganic pollutants in biomass burning emissions, described in detail elsewhere [3, 4]. Real-time measurements and sampling during haze episodes yield optical (BC) and chemical (OC, EC, ions, functionalities) characteristics for evaluation of PM constitutes. The characterization of on-field emissions is intentionally conducted in order to emphasize the specific atmospheric pollutants from the major local sources and then to compare them with those identified in Son La and Ba Vi regions during haze episodes.

BB CAMPAIGNS IN DRY SEASON

The measurement campaign was conducted at a meteorological observation station, located on a hill at 675 m above sea level (a.s.l.) in the outskirts of Son La city, northwestern Vietnam (21.33°N, 103.9°E) during 24 February - 8 April, 2013. A MetOne BAM Continuous Particle Monitor in NASA's SMART-COMMIT mobile laboratory was used for measurements of $PM_{2.5}$ mass concentration. Numerous fires on agricultural fields, gardens, houses, and small cooking places were observed around the station during the entire measurement campaign, as shown in Fig. 1a. The most common agricultural practice was cutting off the crowns of forests on the surrounding hills and subsequent burning of the fallen trees. On the field, crop residues (e.g., cassava root and corn/bushes) were burned. Traditional stoves were used in most residential homes for indoor or outdoor cooking. Near-source sampling was performed on fields during typical agricultural residue burning and near cooking places.

Measurement campaign was carried out during a haze period from 27 May to 14 June 2015 in the mountain region of Ba Vi National Park located 48 km west of Hanoi, intensive on-field burnings were observed nearby at that time (fig.1b). The site was located 400 meters a.s.l. inside a natural forest. Aerosol equivalent black carbon (EBC) concentrations were determined using custom made portable aethalometer which measures the attenuation of light transmitted through particles that are deposited on a quartz fiber filter, as described in detail in [5]. Additional sampling campaign was conducted in the following year, from 1 to 6 June 2016. At that time the fires on fields had not started yet, thus allowing the assessing of the aerosol properties in non-BB period. For characterization of traditional rice straw burning, near-source sampling was performed on fields before and after measurement campaign.

Long- range transport to Son La and Ba Vi sites from the wider regions of South East Asia where extensive biomass burning for agricultural practices is known to take place,

was examined during BB campaigns by means of 120 h air mass back trajectories (BWT) at starting heights of 500, 1000 and 1500 meters a.s.l. which were calculated for every 24 h during the sampling period. The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (NOAA ARL READY Website) was used. Meteorological data was provided by Vietnam Metservices.

Fig. 1. a) Numerous fires in Son La region in March 2013 and b) rice-straw biomass burning activities on the fields of Ba Vi region in May 2015

SAMPLING AND ANALYTICAL METHODOLOGY

Particles equal to or less than 2.5 μm in diameter ($PM_{2.5}$) were collected on quartz fiber and Teflon membrane filters (Teflo, PALL Corp.) by a MiniVol sampler (Airmetrics) at a flow rate of 5 L/min. Size-segregated aerosol particles (PM_1) and $PM_{1\text{-}2.5}$ were collected using a 3-stage cascade impactor (Dekati Ltd.) with a flow rate of 30 L/min. Gravimetrical analyses were performed using Standard Operation Procedure (SOP). Teflon and quartz filter samples were conditioned for about 24 h in a temperature and humidity controlled room (T= 20 ± 5°C, RH=40 ± 10%) and weighted before and after sampling by a 5-digit analytical balance. Organic (OC) and elemental carbons (EC) were measured by thermo-optical transmittance (TOT) instrument (Lab OC-EC Aerosol Analyzer, Sunset Laboratory, Inc.). The samples were heated first in He and then in a mixture of 2% O_2 in He, using controlled heating ramps of the EUSAAR_2 thermal protocol. Charring correction was applied by monitoring the sample transmittance throughout the heating process [6].

Water-soluble potassium was measured by ion chromatography (IC) with conductivity detection, using a Dionex ICS-3000 system (Thermo Scientific) [7], the inorganic anions and cations were measured by capillary electrophoresis (CE) with UV detection [8], using a Capel 103 system (Lumex, Russia).

The absorbance associated with frequency of the vibrational mode of the bond was measured by Fourier Transform Infrared (FTIR) spectroscopy, thus providing the functional groups representing the various classes of organic compounds in the entire aerosol composition. FTIR spectra of filter samples were collected with an IRPrestige-21 spectrometer (Shimadzu, Japan) in a diffuse reflection mode, as described elsewhere [9]. Identification of absorption bands was carried out according to Shimadzu FTIR database and Coates's practical approach [10], as well as using authentic chemical standards.

RESULTS AND DISCUSSION

PM evolution during BB periods

Evolution of $PM_{2.5}$ mass concentrations at the Son La site during February - April 2013 is

shown in fig. 2. The $PM_{2.5}$ mass concentrations exceeded the WHO (World Health Organization) 24 h guideline value of 25 μg/m^3 on 41 of the total 44 measurement days during the study period, indicating the measured PM levels to be substantially impacted by BB activities. $PM_{2.5}$ mass concentrations ranged from relatively low level of 40 to high values in excess of 80 μg/m^3, from smoke of low to high intensity. According to the air mass residence time in the extended region of SE Asia the aerosol origin is categorized into three meaningful geographic areas: South China where apart from BB, other sources such as industry may be active, the area of Indochina where extensive BB occurs and the South China Sea region where minimum anthropogenic impact is expected.

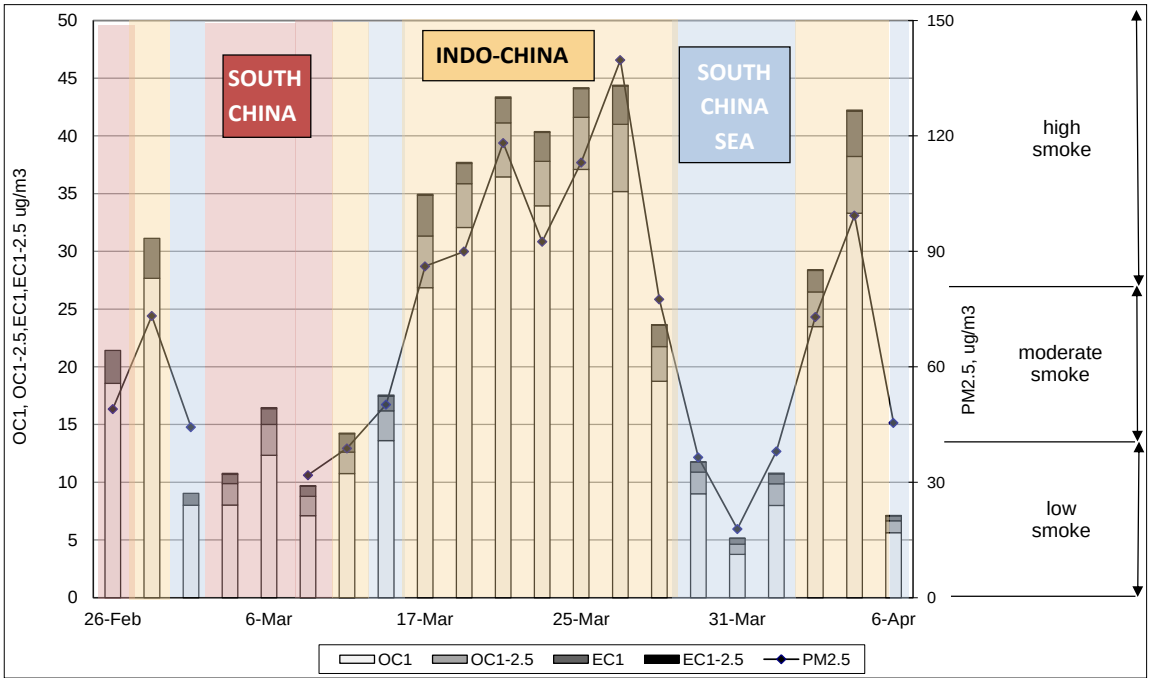

Fig. 2. Concentration of $PM_{2.5}$ mass, OC_1, $OC_{1\text{-}2.5}$, and EC_1 during the measurement campaign at the Son La site in 2013. The levels of smoke intensity are indicated

Evolution of daily mass concentrations at the Ba Vi site from 28 May to 14 June 2015 is shown in fig. 3. 24 h PM_{10} mass concentrations were on average 79±35 μg/m^3 and ranged from 39 μg/m^3 to 167 μg/m^3. It exceeded the WHO 24 h standard guideline PM_{10} value of 50 μg/m^3 on 11 of total 13 measurement days, indicating that the PM level was highly impacted by pollution. PM_{10} ranged from low levels of ≤ 54 μg/m^3 to high smoke intensity above 109 μg/m^3 (fig. 3). Unlike wind speed/direction, the relative humidity (RH) during the sampling period appeared to be inversely proportional to daily PM, varying significantly from 75 to 90%. Intensive fogs with high RH impacted smoke, leading to significant decrease of PM_{10} levels on 5 and 6 June. In addition, precipitation caused the PM_{10} levels to decrease significantly on certain days.

BWTs demonstrated the long-range transport of air masses from Indo-China and west of Vietnam. According to FIRMS WEB Fire Mapper, the biggest fires were observed on the border of Laos and Vietnam, south of Ba Vi region, from 24 May to 31 May. On 1 June fires nearly disappeared until 3-4 June and increased again on 8 June. Fire evolution combined with air mass trajectories allows the postulation that PM_{10} maximum observed on 3 and 11 June was influenced by fire-polluted air mass transported from the south of Vietnam

and Laos. Background PM_{10} level averaged over the sampling period from 1 to 6 June 2016, was found to be near 30 μg m^{-3}. It was time when post-harvesting fires had not started yet. Since the aerosol loading during the haze episode of 2015 even on the days with lower PM_{10} concentrations was higher than 30 μg m^{-3}, it can be suggested that the regional aerosol burden in Ba Vi region in spring of 2015 was significantly influenced by agricultural fires in the surrounding area.

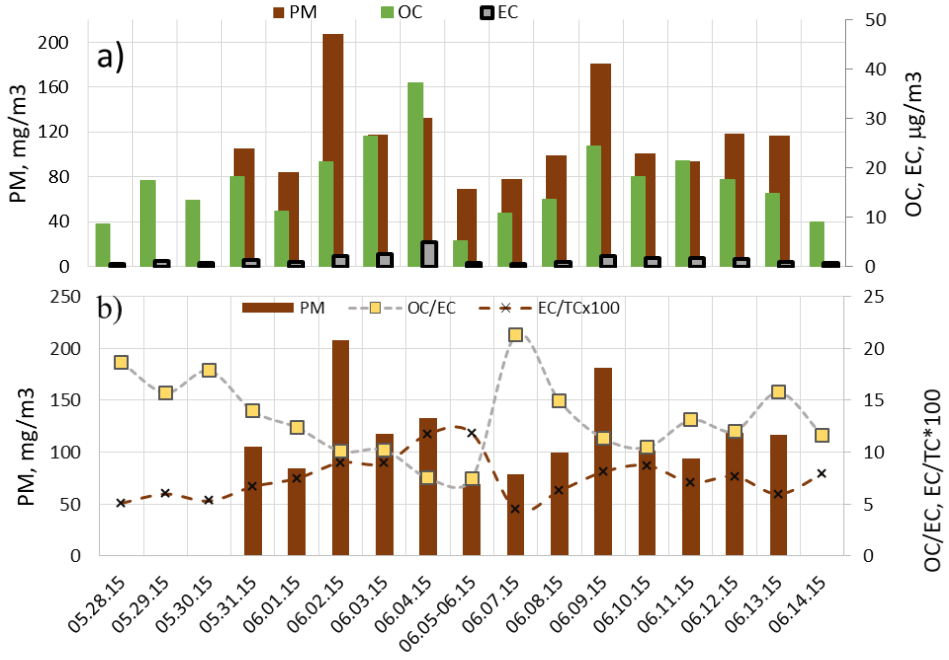

Fig. 3. PM_{10} mass concentrations and a) OC and EC, b) OC/EC and EC/TC ratios during the measurement campaign at the Ba Vi site in 2015

Carbon fractions

The parametrization of the PM mass concentrations according to the smoke intensity is supported by the evolution of aerosol constituents. Fig. 2 shows OC_1 mass concentrations in PM_1 which are well correlated with smoke evolution, comprising around 30% of $PM_{2.5}$. This finding is in good agreement with the typical carbonaceous nature of BB emissions, showing a clear dominance of OC mass fractions (up to 50%) specifically from smoldering burns [11]. The EC_1 fraction in PM_1 was found to be low in comparison with OC_1, yet it correlated well with total $PM_{2.5}$ mass concentrations (fig. 2). As large EC fractions are typically associated with the flaming phase, low EC_1 levels indicate smoldering phase combustion to be dominant during the entire measurement period. The coarser OC fractions, i.e., those in $PM_{1\text{-}2.5}$ ($OC_{1\text{-}2.5}$) and $PM_{2.5\text{-}10}$ ($OC_{2.5\text{-}10}$), were found to be much lower than OC_1 during the majority of high smoke days, demonstrating that the carbonaceous particles are almost entirely present in the fine particle fraction during the BB period.

OC/EC ratios are widely used to describe the impact of BB on aerosol chemistry [12]. Typically, ratios of OC/EC between 4 and 33 are associated with wildfires according to studies conducted in different regions across the globe [12. 13], with higher values being associated with smoldering combustion. We found average OC/EC ratios in PM_1 to be 7.6 on days of low smoke and up to 18.3 during high smoke periods, indicating that smoldering fires were predominant during the dry season in Son La region.

The evolution of carbon fractions at the Ba Vi site was found to be in relation to smoke intensity (fig. 3a), except on 12-13 June when precipitation was significant. OC comprised around 28% of PM_{10} on 4 June and decreased to 7.5%, suggesting almost no BB impact on 5 and 6 June in comparison with other days of the haze. EC levels during the BB season were lower than OC levels, yet correlated with PM_{10}. The highest EC value was 4.9 μg m^{-3} on 4 June, comprising 7.8% of PM_{10}. Low EC levels indicate that smoldering emissions were dominant during the entire sampling period. The lowest EC level occurred on 7 June when the lowest PM_{10} mass was observed, probably because of almost all fires being extinguished on the surrounding fields following the sustained rain period in the previous days. The OC/EC ratios in PM_{10} were above 7.5 and as high as 20 during sampling period, with the highest value observed on the same day (7 June) when EC emissions were radically suppressed (fig. 3b). On average, the OC/EC ratio was 13.2±3.8, indicating that smoldering fires dominated by high OC, prevailed during the smoke period in Ba Vi region. Additionally, meteorological conditions (low wind speed, high temperatures and strong solar radiation) could favor formation of secondary organic aerosols due to condensation of heavier hydrocarbons generated by acid-catalyzed oxidation and photochemical reactions in the smoke plume [11], which could have influenced high daily OC/EC ratios as well. Real-time EBC concentrations on selected days of June of 2015 were varying from 0.5 μgm^{-3} to 8 μgm^{-3}. The highest concentration was recorded on 4 June, in good agreement with the highest EC level observed on the same day.

BB markers

Enhancement of BB marker (levoglucosan and K^+ ion) concentrations by two orders of magnitude indicates strong influence from BB emissions resulting in a regional smoke haze episode. For ambient aerosol during the BB period we found that the levoglucosan and K^+ ion concentrations in PM_1 correlated well with each other (R^2 = 0.91) and with $PM_{2.5}$ mass evolution (fig. 4), supporting the smoke parametrization during the entire BB period. The relative contributions of levoglucosan and K^+ to PM_1 mass were 2.2% and 1.7%, respectively, similar to previous observations between 0.5% and 6% of PM depending on biomass type [11, 12]. An average K^+/EC ratio of 0.2 obtained during the high smoke period was between the values for smoldering and flaming fires obtained during regional-scale biomass episode [14], and in good agreement with values reported from previous biomass burning studies [15].

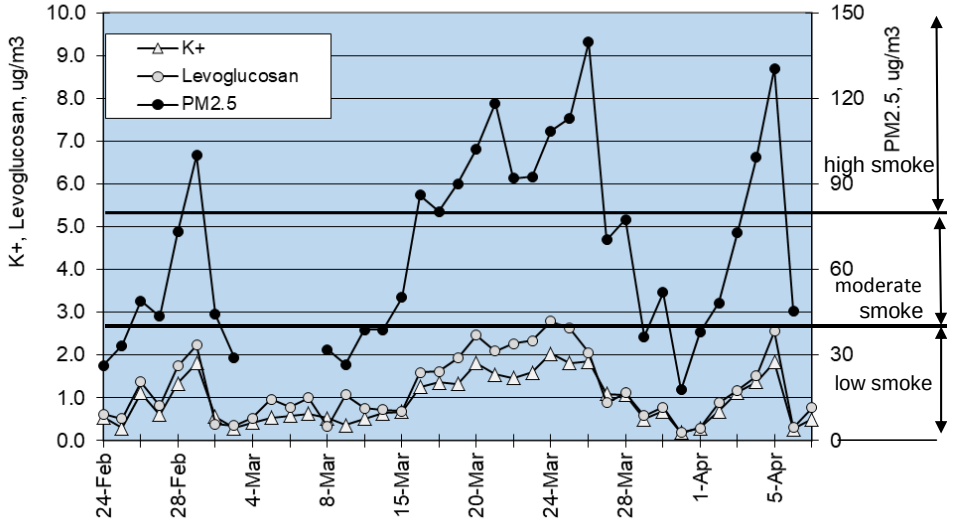

Fig. 4. Concentrations of $PM_{2.5}$ mass, K^+, and levoglucosan in PM_1 at Son La site in 2013

Molecular BB marker mannosan (Man) derived from the thermal decomposition of hemicellulose, showed the same temporal pattern as levoglucosan (Lev), as evidenced in the high correlation, R^2 = 0.98, between the ambient concentrations of two markers. Based on Lev/Man ratios from previously reported source emission studies and the results from the near-source measurements of locally burned species in Son La region, the average Lev/Man ratio of 14.6 in PM_1 indicates a mix of hard wood and grasses/straw with minor contributions from softwood burning as the sources for the sampled BB aerosol.

Mass concentrations of three organic and nine inorganic ions during the haze episode at the Ba Vi site were summarized in fig. 5. On days of highest smoke, total ion concentration comprised 47% of PM_{10}, and correlated well with the smoke evolution on other days. The highest levels, (> 5 μm.m^{-3}), are found for CH_3COO^-, Ca^{2+}, Na^+, and Cl^- ions. High levels of Ca^{2+} ions, also observed in coarse particles in Son La region, are associated with soil dust resuspension due to long-lasting smoldering on the fields. Na^+ and Cl^- could be linked to aged sea salt as the Ba Vi site is located about 100 km west of the Bac Bo (Tonkin) Gulf. Cl^- ions were also prominent in the pattern of smoke emissions from rice straw burning, and therefore they could impact the regional abundance due to intensive fires. The relative contribution of K^+ to PM_{10} was high, on average from 31 May to 9 June as high as 4%, and up to 6.9% on days of high smoke , similar to observations during wildfire smoke events [11, 12].

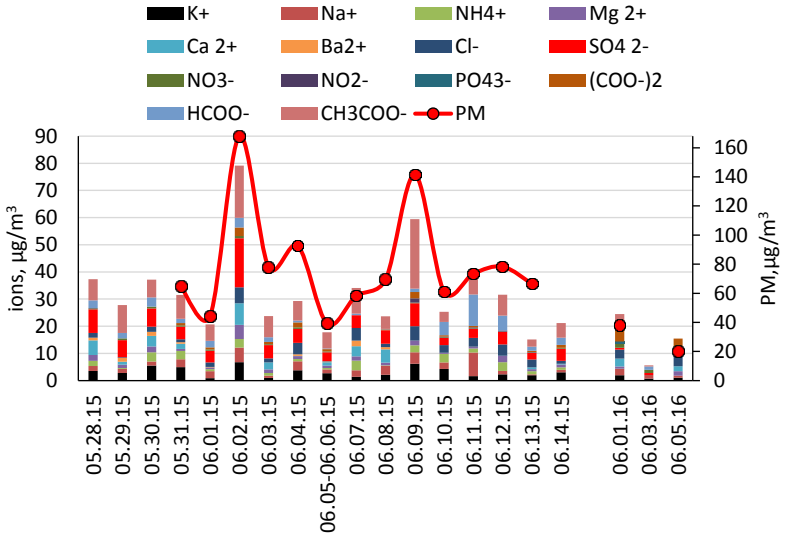

Fig. 5. PM_{10} mass concentrations, organic and inorganic ions on the Ba Vi site in 2015 and 2016

Organic composition

Parameterization of smoke intensity during the BB period is useful for quantification of chemical composition of ambient aerosols collected during days of similar smoke levels. The prominent absorption bands of acid and non-acid carbonyls, carboxylates, and aliphatic carbon in $PM_{2.5}$ during the whole BB period are similar to those found in the on-field and cooking emissions (fig. 6a, 6b). Non-acidic carbonyls in addition to carboxylic acid groups were previously found in forest fire emissions and were assigned to aldehyde/ketone, ester/lactone, and acid anhydride groups [16]. A band of aliphatic carbon in low smoke periods may be a feature of the urban background, because alkanes dominate functionalities of diesel emissions [17]. The band of ammonium NH^+ (3350-3100 cm^{-1}) is prominent in ambient aerosols.

Dust functionalities are specific features of coarse particles identified by the prominent

band of silicate ions (SiO_4^{4-}) in various aluminosilicates with the variable position near 1000 cm^{-1}. Carbonates (CO_3^{2-}) were identified in the range 880-860 cm^{-1}, and their presence in the $PM_{2.5-10}$ size fraction was confirmed by TOT measurements of carbonates in form of carbonate carbon. The SO_4^{2-} absorption band (580-700 cm^{-1}) was well identified together with those of NH_4^+ in coarse particles. The lack of correlation with the smoke levels indicates the predominance of regional sources for sulfates and ammonium, in addition to emissions from local industry. As reported by Huang et al., (2013), only 30-40% of the sulfur deposition in Vietnam originates from domestic sources, with the rest contributed mainly by southern China and Thailand.

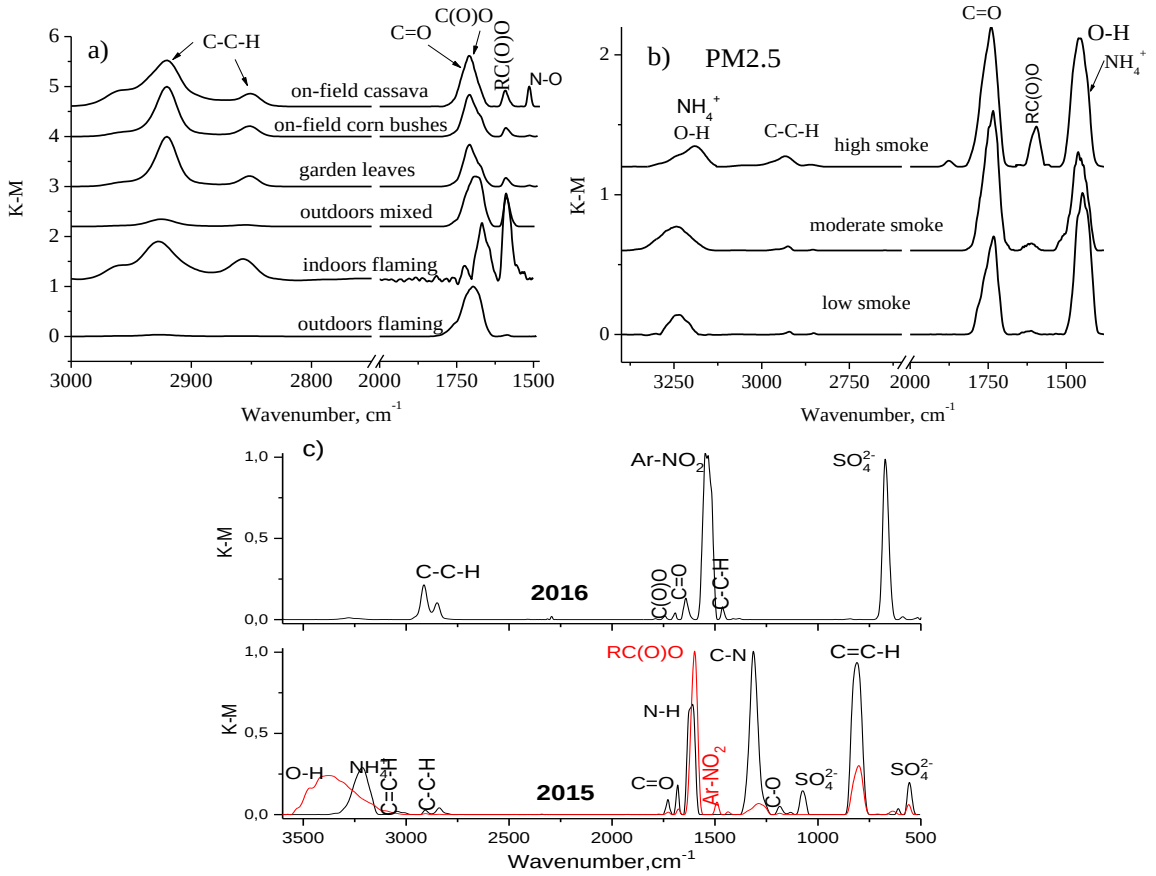

Fig. 6. FTIR spectra in a) cooking and on-field burning emissions in Son La region, and b) ambient aerosols at the Son La site in 2013 in $PM_{2.5}$ size fraction, and c) at the Ba Vi site in 2015 and 2016 in PM_{10} size fraction. Smoke intensity is parametrized according to fig. 2. Absorption bands are indicated

At the Ba Vi site, the most prominent FTIR absorption bands on days with the lowest PM_{10} levels of 40-60 μg m^{-3} (5-7 June) were of N-H (1602 cm^{-1}) and C-N (1308 cm^{-1}) groups in aromatic amino compounds (fig. 6c). They indicate the abundance of biogenic groups in bioaerosols [18], which is a reasonable feature of ambient environments after and during rain periods. The presence of aromatics in low smoke aerosols is confirmed by C=C-H (810 cm^{-1}) bending and C=C-H stretching (3027 cm^{-1}) vibrations, probably due to the regional pollution by aromatic compounds. The bands at 3209 cm^{-1} and 1194 cm^{-1} in low smoke identify sulfate ammonium, also indicating the possible impact of cooking emissions and

secondary aerosol formation at regional level. The dominant abundance of organic acid ions confirms the FTIR identification of carboxylate ions in the pattern of rice straw burning and ambient aerosols. Fire emissions during the haze episode impact aerosol chemistry significantly. The spectrum collected on 2 June at a PM_{10} level of 165 $\mu g.m^{-3}$ demonstrates strong bands of hydroxyls (3383 cm^{-1}), carboxylates (1595 cm^{-1}), and aromatic nitro groups (1489 cm^{-1}), similar to the characteristic absorption in the functional pattern of rice straw burning, whereas the bands of C-N and C=C-H groups, presumably from biogenic and cooking sources, were significantly decreased.

CONCLUSIONS

Impact of traditional biomass burning activities on regional air quality is a major environmental concern. Son La and Ba Vi areas in February-June face severe levels of air pollution, with critical PM concentrations significantly exceeding the air quality standards. Significantly lower PM_{10} mass concentrations and strong difference in aerosol composition before post-harvesting activities suggested that agricultural burning represents a large contribution to air quality degradation in agriculture areas of Vietnam. Analysis of collected data showed the emissions of environmentally-dangerous organic and inorganic pollutants for interpreting the impact of the sources of adverse PM in urban and rural areas of high population.

Results presented above have been undertaken within framework of 7-SEAS campaign and Russian-Vietnam bilateral project. For effective air quality management and mitigation it is necessary to quantify the contribution from the local sources and long range transport. In the near future focusing firstly on Hanoi city, we propose building on existing recent and new measurements taken/undertaken as part of the 7-SEAS campaign as well as possibly new strategic measurements and source apportionment in combination with new regional and urban-scale modelling studies developing current modelling capabilities to enhance our understanding of Hanoi air quality. Recent emission inventories will be used for urban modelling which will include constructing a fine-scale spatial distribution of cooking emissions, as well as using existing traffic and industrial source emissions. This will enable us to quantify the sources of PM and gaseous air pollution in Hanoi and their seasonal variation, and identify the impact of further potential growth and various mitigation scenarios.

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